Modelling heat and mass transfer in fluidised bed coating processes

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MODELLING HEAT AND MASS TRANSFER IN FLUIDISED BED COATING PROCESSES

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Introduction

Microencapsulation is defined as a technology whereby active ingredients or mixtures thereof are packaged or coated in miniature, sealed capsules. As a result, useful and novel properties can be conferred to the encapsulated ingredient. Main benefits of microencapsulation include controlled release, increased shelf life, taste masking, improved processability or the conversion of liquid into solid (powder-like) products. Microencapsulation of food ingredients and additives enables the food technologist to select products with improved or totally new properties. To date, many techniques have been developed to achieve encapsulation of solid, liquid or even gaseous ingredients. Fluidised bed coating is one among the more commonly applied encapsulation techniques. Although fluidised bed coating was originally developed as a pharmaceutical technique, it is increasingly being applied in the food industry. However, there is a large difference in the economics of both industries: whereas the pharmaceutical industry is characterised by low volume production and high profit margins, the food industry has to handle high capacity, low profit margin products. Therefore, the food technologist is obliged to cut production costs.

Process models should be considered important tool in optimising the fluidised bed coating process. Currently, there is a lack of knowledge on how the different heat and mass transfer processes between the three phases involved – gas, solid (particles) and liquid phase – interact with each other. Furthermore, the quantitative link between the fluidised bed coating microprocesses and the occurrence of side-effects, and its effect on the quality of the encapsulated product are not yet fully understood. Process models could not only help in clarifying these ‘missing links’, but could also be applied in advanced process control strategies or in scale-up studies.

The thesis is built up of five chapters. In Chapter 1, the theoretical background is provided to be used in the development of the different process models throughout this thesis. First, a literature review is given concerning microencapsulation and fluidised bed coating. Also, a brief introduction to fluidisation is given. Emphasis is made on bubbling fluidised beds, as this is the predominant fluidisation regime in fluidised bed coating processes. Finally, the theory of heat and mass is briefly discussed and will mainly focus on heat and mass transfer occurring in bubbling fluidised beds.

In Chapter 2, an overview is given of the different existing models for fluidised bed coating. The models are classified into black-box, grey-box and white-box models according to the extent to which the fundamental heat and mass transfer mechanics are implemented, as opposed to the use of empirical correlations that match input/output process data. Also, a brief introduction to population balance models is given.

Chapter 3 starts with the development of a new model for the batch fluidised bed coating process. This one-dimensional model was based on the discretisation of the fluidised bed into
different layers or control volumes, in which the dynamic heat and mass balances for air, water vapour, core particles and coating material were established. Next, a calculation method is presented which involved a Monte Carlo technique for the simulation of the particle exchange in combination with the first-order Euler’s method for solving the heat and mass balances. The model was shown to be capable of predicting both the dynamic coating mass distribution and the one-dimensional thermodynamic behaviour of the fluidised bed during batch operation. The simulation results were validated using the results from tests on a Glatt GPCG-1 fluidised bed unit in the top-spray configuration. Chapter 3 finishes with a model sensitivity analysis where the effect of changes in the input variables on the model-predicted thermodynamic behaviour of the fluidised bed was studied.

Chapter 4 continues to expand upon the model developed in Chapter 3. Careful analysis of the proposed model revealed a number of shortcomings of the existing model, more specifically with regard to the assumed distribution of coating solution among the simulated particle population. To overcome these shortcomings, it was necessary to extend the model with a third phase, namely the droplet phase. The calculation method developed in Chapter 3 will be further expanded to allow the simultaneous simulation of the three phases (liquid, gas and solid phase). Next, the extended model was validated using experimental temperature distributions retrieved by means of a purpose-built fluidised bed unit and using spray drying losses found in literature. Once the model has been validated, a model sensitivity analysis was finally performed to study the effect of the different input variables on the thermodynamic behaviour of the bed, including the temperature and concentration gradients of both the solid and gas phases, the size of the spraying region, the occurrence of spray drying losses and the growth rate distribution of coating layer.

In the last chapter, a framework is presented to model the batch fluidised bed coating process using a two-dimensional axisymmetric discretisation scheme as opposed to the one-dimensional discretisation schemes that were used in Chapters 3 and 4. Several factors necessitated the use of two- or three-dimensional models including the need to incorporate radial temperature and concentration gradients (as opposed to just axial gradients in one-dimensional models), to implement ordered particle movement and to model the spraying region as a cone-shaped volume (rather than a flat layer in one-dimensional models). Chapter 5 finishes with the presentation of some preliminary results obtained from the two-dimensional model and the comparison with experimental temperature and concentration distributions using the experimental set-up, described in Chapter 4.
1. Theoretical background

1.1. Introduction

In this chapter, a brief discussion will be given concerning the application of microencapsulation techniques in the food industry. As this work is focused on fluidised bed coating as a means of producing encapsulated food ingredients, some of the basic physical aspects involved in fluidised bed coating shall be discussed, including fluidisation, heat and mass transfer in fluidised beds and droplet/particle dynamics.

1.2. Microencapsulation

1.2.1. Introduction

Microencapsulation is defined as a technology whereby active ingredients or mixtures thereof – in solid, liquid, or gaseous form – are packaged in miniature, sealed capsules (Sparks, 1981; Dziezak, 1988; Shahidi and Han, 1993). The resulting miniature packages, called ‘microcapsules’, range in size from several tenths of a micrometer to several millimetres and have a multitude of different shapes, depending on the materials and methods used to prepare them (Balassa and Fanger, 1971; Sparks, 1981; Jackson and Lee, 1991). Capsules that are larger than 5 millimetres are often called macrocapsules, while encapsulated particles smaller than 0.2 micrometre are denoted as nanocapsules (King, 1995). The process of microencapsulation thereby confers distinct physicochemical capabilities compared to the original non-encapsulated ingredient (Barbosa-Canovas et al., 2005) to allow the release of the microcapsule’s content at controlled rates under specific conditions (Todd, 1970; Desai and Park, 2005).

Historically, the development of the first commercial applications of microencapsulation date back to the early 1950’s with the invention of ‘carbonless’ copy paper by Green (1955). Ever since, encapsulation has become a topic of growing interest in a wide range of industrial and scientific areas, ranging from pharmaceutics to agriculture (Arshady, 1993). Although the technology of microencapsulation and controlled release was mainly developed in the pharmaceutical industry, it is being increasingly used in the food industry (Greenblatt et al., 1993; Dewettinck and Huyghebaert, 1999; Gibbs et al., 1999). The use of microencapsulation as an enabling technology providing tailor-made and fine-tuned ingredients gained increasing interest which can be demonstrated by the exponential growth in the number of publications and patents covering the use of microencapsulation technologies in food applications (Gouin, 2004).

Although the food industry and pharmaceutical industry share the techniques for the production of microencapsulated ingredients and active components, there is a large difference in the economics of their markets. Whereas the pharmaceutical industry is...
characterised by low volume production and high profit margins, the food industry has to handle high capacity, low profit margin products, subsequently rendering the food industry less tolerable to costly encapsulation production technologies (Dezarn, 1995; Gibbs et al. 1999). Thus, cost-effectiveness of microcapsule preparation techniques and materials is of paramount importance in the continual development and application of encapsulated food products.

1.2.2. Microcapsule properties

The microcapsule structure is divided into the core – also called fill, substrate or internal phase – which is the interior content of the capsule, and the coating material – also denominated as wall, shell, membrane, carrier or coat – which consists of one or more external layers that cover the core material (Barbosa-Canovas et al., 2005).

The microcapsules can be divided into several arbitrary and overlapping classifications according to their structural properties (Arshady, 1993; Gibbs et al., 1999; Barbosa-Canovas et al., 2005). Distinction is made between three classes: single particle structures, aggregate structures and multi-walled structures (Figure 1.1). In multi-walled capsules, several coating layers of different composition are concentrically deposited around the core to serve multiple purposes relating to production, storage and the release of these microcapsules. The aggregate structure originates when multiple distinct cores are embedded within the same microcapsule. A special type of aggregate structures is the matrix-type, where numerous cores are dispersed in a continuous matrix of wall material. Also typical to matrix-type microcapsules is that some cores are exposed at the surface of the capsule, thereby affecting the release properties of the core material. Matrix-type microcapsules are generally the result of spray drying of emulsions or solid-in-liquid dispersions (Reineccius, 1988; 1989). As the properties of matrix-type structures are fundamentally different (i.e., release properties), some authors prefer to denote them with the term ‘microspheres’ or ‘microparticles’ (Brazel, 1999).

![Figure 1.1. Various (idealised) microcapsule morphologies (after Arshady, 1993; Gibbs et al., 1999)](image-url)
Coating materials consist of a wide variety of natural, semi-synthetic or synthetic filmforming polymers. The majority of FDA-approved (Food and Drug Administration) materials for use as coating material in foods are based on edible polysaccharides (Nisperos-Carriedo, 1994), proteins (Gennadios et al., 1994) and, lipids and resins (Hernandez, 1994). The choice of coating material depends on the type of encapsulation process used, the physicochemical properties of both the core and coating material, the compatibility between the core and the coating material (i.e. non-reactivity and insolubility between core and coating), the type of release mechanism and ultimately, the cost of the wall material (Jackson and Lee, 1991).

Ultimately, microcapsules have been designed to release their contents at a specific time and location, at a specified rate or as the result of specific stimulus. A variety of release mechanisms exist (Pothakamury and Barbosa-Canovas, 1995; Reineccius, 1995), but they can be categorised among three different groups. A first release mechanism is fracturation, in which the coating is fractured or broken due to external forces such as pressure or shear force, for example, during chewing. A second type of release is the diffusion-controlled release. In diffusion-controlled release, the coating is not necessarily broken down, but the diffusivity of the core material in the coating polymer could be altered by, for example, the action of solvents, the application of heat or changes in pH in the microcapsule’s environment. Finally, in a third type of release, the actual coating wall is degraded or broken down either by dissolution, melting or the interaction of wall-degrading enzymes.

1.2.3. Purpose of microencapsulation in food technology

Considering the food industry, there is a wide range of reasons and purposes for the use of microencapsulated products. Some of the major reasons are summarised in the following, but non-exhaustive list (Versic, 1988; Greenblatt et al., 1993; Dezarn, 1995):

- To convert the physical state from liquid products to solid, enabling improved handling and storage.
- To control the release of the capsule’s content at a specific time, at a specific rate or in a targeted location (i.e., mouth, small intestine). Other examples include delayed and sustained release.
- To improve the stability, and ultimately increase the shelf life, of the encapsulated core material against degenerating environmental factors, such as light, oxygen, moisture and temperature.
- To mask undesirable flavours.
- To separate incompatible or reactive components in ingredient mixes.
- To promote improved handling and processability of the core materials by, for example, increasing product flowability, reducing hygroscopicity, altering product density, shape and size, suppressing dust formation or improving solubility.
1.2.4. Microencapsulation techniques

To date, a wide range of techniques have been developed to microencapsulate food ingredients. In general, the selection of a specific encapsulation method depends on economics, sensitivity of the core material, desired size of the microcapsule, physical and/or chemical properties of both core and coating material, type of application for the encapsulated food ingredient and the release mechanism (Jackson and Lee, 1991). The different microencapsulation techniques are usually classified among chemical and mechanical processes. This classification can be somewhat misleading, since so-called mechanical processes may actually involve a chemical reaction and so-called chemical processes may rely exclusively on physical phenomena. Therefore, Thies (1996) proposed a classification into type A and type B processes. Type A processes usually involve a liquid phase in a stirred tank or tubular reactor, while type B processes utilize a gas phase at some stage of the encapsulation process.

Considering the variety of encapsulation techniques and their applicability in the food industry, a number of outstanding reviews have been published (Balassa and Fanger, 1971; Sparks, 1981; Dziezak, 1988; Jackson and Lee, 1991; Arshady, 1993; Bakan, 1994; Dezarn, 1995; Thies, 1996; Gibbs et al., 1999; Depypere et al., 2003; Gouin, 2004; Barbosa-Canovas, 2005; Desai and Park, 2005). The different encapsulation techniques include extrusion, solvent extraction, coacervation, cocrystallisation, spray drying (or spray cooling/chilling), fluidised bed coating, pan coating and inclusion complexation. In the food industry, some of the more commonly applied encapsulation techniques include fluidised bed coating, pan coating and spray drying (Janovsky, 1993; Brazel, 1999). However, as the focus in this research work lies solely on fluidised bed coating, only this technique will be discussed in detail.
1.3. Fluidised bed coating

1.3.1. Introduction

Fluidised bed coating, also referred to as air suspension coating, was developed as a pharmaceutical technique to coat or encapsulate active constituents. The process was originally developed by Dale Wurster in the 1950’s for coating pharmaceutical tablets, hence the term ‘Wurster process’ (Jones, 1991; Arshady, 1993). Since then, many design modifications have been proposed with the aim of improving coating quality and reducing operational costs. However, all these different fluidised bed coating techniques have the same basic working principle in common with the Wurster process, which is the application of the coating material by means of atomisation or spraying onto fluidised core particles. Some of the predominant fluidised bed coating designs will be discussed in this section.

Fluidised bed coating is very flexible as it is suitable for many core (including crystals, agglomerates with a wide range of particle sizes) and coating materials (i.e. water soluble coatings, suspension, emulsions, molten waxes, fats,…) (Jones, 1985, 1994), it allows the mechanical formation of multiple layers on the core particles (Fukumori and Ichikawa, 2004) and the fluidised bed unit enables different unit operations such as drying, granulation or agglomeration. Probably because of the aforementioned flexibility of the fluidised bed coating process, it has found widespread usage in the food industry. Currently, it accounts for the second largest production in encapsulated materials in the food industry (Barbosa-Canoivas, 2005).

1.3.2. Working principle

1.3.2.1. Fluidisation

Fluidisation arises when an upward moving gas (i.e., process air), drawn though a bed of particles, reaches sufficient velocity to suspend the particles. The particle bed then assumes fluidlike properties, hence the term ‘fluidisation’ (Dewettinck and Huyghebaert, 1999). In order to maintain the bed’s fluidisation behaviour, the gas velocity has to be situated between the so-called minimum fluidisation velocity and the terminal velocity, which is the gas velocity at which pneumatic transport of particles occurs (Guignon et al., 2002). More details concerning fluidised bed behaviour are given in Section 1.4.

1.3.2.2. Droplet production and atomisation

One of the important properties of a fluidised bed is that the suspended particles expose their entire surface to the fluidising air, thus resulting in optimal heat and mass transfer. The coating polymer to be applied could be an aqueous or organic solvent-based solution or even...
a melt and is continuously sprayed into the fluidised bed, usually by means of a pneumatic or binary nozzle which may be submerged in or positioned above the bed (Jozwiaskowski et al., 1990; Link and Schlünder, 1997; Zank et al., 2001; Nasr et al., 2002). The fluidising air also supplies the evaporative capacity to remove the solvent, leaving behind the dissolved material on the surface of the fluidised particles. Depending on the position of the nozzle with respect to the fluidised bed, distinction is made between top-spray, bottom-spray and tangential-spray. In the latter, one or more nozzles are submerged into and positioned tangentially to the fluidised bed (Jones, 1985).

The use of compressed air in a binary nozzle (Figure 1.2) results in very strong shear forces at the liquid-gas interface, producing droplets with a size ranging from 10 to 40 µm (Lefebvre, 1988; Guignon et al., 2002). Pneumatic nozzles are characterised by their specific construction, the liquid flow rate, the rheology of the coating liquid (viscosity, surface tension,...) and, the flow rate and pressure of the atomising air (Schaefer and Wørts, 1977; Lefebvre, 1988; Liu and Litster, 1993b; Juslin et al., 1995a, b; Dewettinck and Huyghebaert, 1998). These variables, in turn, will influence the spray’s microstructure, being the droplet size distribution, and macrostructure which includes the droplet spatial distribution, droplet velocity spatial distribution, shape of the spraying pattern and spray penetration depth (Guignon et al., 2002). It is important to note that a certain ratio of particle to droplet size needs to be respected. According to Liu and Litster (1993b), this size ratio should be at least 10.

![Figure 1.2. The pneumatic or binary nozzle with external liquid/gas mixing.](image-url)
To correlate droplet size with nozzle and coating liquid related parameters, Lefebvre (1988) proposed the following model for the calculation of the surface weighted average droplet diameter or Sauter mean diameter \( d_{dr} \), given in µm, of a pneumatic or binary nozzle:

\[
d_{dr} = 0.48 d_{or} \left( \frac{\gamma_{sol}}{\rho_{at} \Delta P_{at} d_{or}} \right)^{0.4} \left( 1 + \frac{M_{sol}}{G_{at}} \right)^{0.4} + 0.15 d_{or} \left( \frac{\mu_{sol}^2}{\rho_{sol} \gamma_{sol} d_{or}} \right)^{0.5} \left( 1 + \frac{M_{sol}}{G_{at}} \right)
\]  

(1.1)

As can be seen in Eq. (1.1), the following variables are included in the droplet size correlation: liquid flow rate, \( M_{sol} \) in kg s\(^{-1}\), atomisation air flow rate, \( G_{at} \) in kg s\(^{-1}\), atomisation air pressure, \( \Delta P_{at} \) in bar, diameter of the nozzle’s liquid orifice, \( d_{or} \), density, \( \rho_{sol} \) in kg s\(^{-1}\), surface tension, \( \gamma_{sol} \) in N m\(^{-1}\), and viscosity of the coating solution, \( \mu_{sol} \) in Pa s. Alternatively, Masters (1991) proposed the following correlation:

\[
d_{dr} = \frac{585000 \gamma_{sol}}{v_{at, noz} \sqrt{\rho_{sol}}} + 597 \left( \frac{\mu_{sol}}{\sqrt{\gamma_{sol} \rho_{sol}}} \right)^{0.45} \left( \frac{1000 \rho_{at} M_{sol}}{\rho_{sol} G_{at}} \right)^{1.5}
\]  

(1.2)

The actual production of the droplets occurs in a small region near the nozzle orifice, where the liquid jet is broken apart by the large shear forces, created by the release of compressed air around the liquid jet (Nasr et al., 2002). To determine the distance from the nozzle where stable droplets (in terms of constant diameter) are produced, the dimensionless Weber number is used. The dimensionless Weber number, \( We \), is the ratio of the droplet kinetic energy to the droplet surface energy (Orme, 1997; Frohn and Roth, 2000):

\[
We_{dr} = \frac{\rho_a \left( v_a - v_{dr} \right)^2 d_{dr}}{2 \gamma_{dr}}
\]  

(1.3)

In Eq (1.3) are \( v_{dr} \) and \( v_a \) the velocities of the droplet and of the fluid (gas) in which the droplet moves, respectively. The variable \( d_{dr} \) represents the droplet diameter, \( \rho_a \) the density of the fluidum (air), while \( \gamma_{dr} \) is the droplet surface tension. The higher the Weber number, the higher the kinetic energy compared to the droplet surface energy and thus, the higher the probability for droplet breakage due to droplet deformation. Three droplet breakage mechanisms exist: normal stresses lead to ‘bag’-type break-up, while tangential stresses to the droplet result in ‘stripping’ break-up (Liu and Reitz, 1993; Abdelghaffar et al., 2005). A third type of break-up is the ‘catastrophic’ droplet break-up and occurs at very high shear forces (or large Weber numbers). According to Sazhin et al. (2003, 2005), ‘Bag’-type breakage occurs when the droplet Weber number,

\[
We_{dr} > 6
\]  

(1.4)

On the other hand, ‘stripping’ break-up occurs when
Using the droplet range between 10 and 40 µm and atomisation air pressures between 0.5 and 3.5 bar characteristic to fluidised bed coating, the region where droplet breakage occurs is limited to the first 10 mm closest to the nozzle.

Besides droplet breakage, coalescence could also occur when droplets collide in the nozzle-produced spray. Brazier-Smith et al. (1972) defined the coalescence efficiency for two colliding droplets as

\[
\eta_{\text{coal}} = \min\left(1, \frac{2.4 f(x)}{We_{dr}}\right) \tag{1.6}
\]

In Eq. (1.6) is the function \( f(x) \),

\[
f(x) = x^3 - 2.4x^2 + 2.7x \quad x = \frac{d_{dr,1}}{d_{dr,2}} \tag{1.7}
\]

With \( d_{dr,1} \) and \( d_{dr,2} \) being the diameters of the two colliding droplets and \( d_{dr,1} > d_{dr,2} \). In Eq. (1.6) is \( We_{dr} \) the average Weber number of the two colliding droplets,

\[
We_{dr} = \frac{\rho \left(v_a - v_{dr}\right)^2 \left(d_{dr,1} + d_{dr,2}\right)}{4\gamma_{dr}} \tag{1.8}
\]

1.3.2.3. Droplet impact and particle wetting

During the approach of a droplet towards a particle, different mechanisms, such as diffusion and inertia, may result in the separation of the droplet trajectory and the fluid streamline (Figure 1.3), leading to an impact (Link and Schlünder, 1997; Guignon et al., 2002). The type of droplet collision mechanism depends on both size and velocity of the impacting droplet and the receiving particle. Assuming typical droplet sizes in fluidised bed coating ranging between 10 and 40 micrometers, droplet collection caused by inertia is the predominant collision mechanism. Whether a droplet is able to reach the surface of a fluidised particle, depends on a number of factors, including droplet velocity, density and droplet size compared to size of the receiving particle (Guignon et al., 2002).

The ability of a droplet to come into contact with the particle is expressed by the impingement efficiency, \( \chi_{dr} \), which is determined as the ratio of the effective, \( \pi d_{im}^2 / 4 \), to the geometrical cross-sectional area, \( \pi d_p^2 / 4 \), as shown in Figure 1.3:
The theoretical background

Modelling heat and mass transfer in fluidised bed coating processes

\[ \chi_{dr} = \left( \frac{d_{in}}{d_p} \right)^2 \]  

(1.9)

\[ \text{Droplet trajectory separation from fluid streamline} \]

\[ \text{Particle} \]

\[ \text{Fluid streamlines} \]

\[ \text{\( d_e \)} \]

\[ \text{\( d_{in} \)} \]

**Figure 1.3.** The effective cross-sectional area for defining the droplet impingement efficiency.

The impingement efficiency can be calculated using the following empirical equation (Löffler, 1988; Zank et al., 2001; Heinrich et al., 2003a):

\[ \chi_{dr} = \left( \frac{St_{dr}}{St_{dr} + a'} \right)^{b'} \]  

(1.10)

In Eq. (1.10) is \( St_{dr} \) the droplet dimensionless Stokes number. The Stokes number is a quantification of droplet agility in moving fluids. If \( St_{dr} \ll 1 \) then the droplets have ample time to respond to changes in fluid velocity (i.e. the change in gas flow directly around the particle’s surface). If \( St_{dr} \gg 1 \) then the droplets will have essentially no time to respond to fluid velocity changes (Crowe et al., 1997). The Stokes number is calculated as (Heinrich et al., 2003a):

\[ St_{dr} = \frac{\rho_{dr} d_{dr}^2 |\Delta \nu_{dr}|}{9 \mu_a d_p} \]  

(1.11)

With \( \rho_{dr} \), the droplet density (in kg s\(^{-1}\)), \( d_{dr} \), the droplet diameter (in m), \( \Delta \nu_{dr} \), the relative velocity between the droplet and the fluid (air) surrounding the particle, \( \mu_a \), the viscosity of the fluid (air) and \( d_p \), the diameter of the receiving particle.

The parameters \( a' \) and \( b' \) in Eq. (1.10) depend on the particle Reynolds number, \( Re_p \), as shown in Table 1.1. From Eq. (1.10) and Table 1.1 it can be seen that the impingement efficiency decreases with decreasing droplet diameter or with decreasing gas velocity.
Theoretical background

1.3.2.4. Particle growth mechanisms in fluidised bed processing

Previous studies, carried out by Smith and Nienow (1982) and by Maronga and Wnukowski (1997a, 1998), have shown that particles are only wetted in a small region in the proximity of the spraying nozzle. This so-called ‘wetting zone’ (or ‘spraying region’, or ‘coating volume’) corresponds to the droplet penetration depth of the spray. In this ‘wetting zone’, droplet formation, droplet/particle impact and adhesion, spreading of the droplet on the particle surface and evaporation occur almost simultaneously (Guignon et al., 2002). The repeated cycle of wetting and drying when the fluidised particles move from and towards the wetting zone, results in the creation of a uniform and layered structure around the individual particles.

When liquid binders or dissolved coating polymers are added to a fluidised bed, two different particle growth modes are to be distinguished, depending on the mass rate of binder addition and the evaporative capacity of the bed (Hemati et al., 2003; Maronga and Wnukowski, 1998). An overview of the different phenomena occurring in liquid-sprayed fluidised beds is shown in Figure 1.4.

When wetted particles collide, a wet bridge is formed in between them. Depending on the liquid bridge strength and the kinetic energy of the colliding particles, these bridges may persist beyond the point of solidification or drying and consequently, dry agglomerates or clusters are formed (Smith and Nienow, 1983; Becher and Schlünder, 1997; Saleh et al., 1999). This type of particle growth is termed ‘agglomeration’. The different stages of the agglomeration process are also illustrated in Figure 1.5b. If excessive amounts of coating or binder liquid are sprayed into the fluidised bed, the rate of liquid bridge formation could become too high, resulting in the formation of large, wet lumps with subsequent defluidisation (or collapsing) of the bed.

When fluidised beds operate in dryer conditions, i.e. when the evaporative capacity of the bed is increased or when the rate at which the liquid binder or coating solution is added to the bed is reduced, the collected coating solution on the particle surface will sufficiently dry before collision. Consequently, the formation of liquid bridges is prevented (Link and Schlünder, 1997). This type of particle growth is termed ‘layering’, which is shown in detail in Figure

Table 1.1. Parameters $a’$ and $b’$ for the calculation of the droplet impingement efficiency.

<table>
<thead>
<tr>
<th>$Re_p$</th>
<th>$a’$</th>
<th>$b’$</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt; 1</td>
<td>0.65</td>
<td>3.7</td>
</tr>
<tr>
<td>10</td>
<td>1.24</td>
<td>1.95</td>
</tr>
<tr>
<td>40</td>
<td>1.03</td>
<td>2.07</td>
</tr>
<tr>
<td>60</td>
<td>0.506</td>
<td>1.84</td>
</tr>
<tr>
<td>&gt;&gt;100</td>
<td>0.25</td>
<td>2</td>
</tr>
</tbody>
</table>

Modelling heat and mass transfer in fluidised bed coating processes
1.5a. In case of weak bridge formation, it is possible that the produced agglomerates are fragmented, a process which could be considered layered growth, but coating imperfections are likely to occur.

**Figure 1.4.** Possible phenomena taking place during the fluidised bed coating process (after Saleh et al., 1999; Maronga and Wnukowski, 1998).

**Figure 1.5.** Fluidised bed coating (a) and agglomeration (b) mechanisms (Courtesy: Glatt GmbH), corresponding to indicated pathways (A,B) in Figure 1.4.
1.3.3. Batch fluidised bed coating

As already stated, classification of fluidised bed coating processes is primarily made based on the position of the nozzle. Three major configurations could be distinguished in batch fluidised bed coating: bottom-spray, top-spray and tangential-spray configurations.

1.3.3.1. Top-spray fluidised bed coating

The top-spray system has been successfully used to coat materials as small as 100 µm (Jones, 1988a, 1988b). Typical to top-spray configurations, is the product container, which is an un baffled, inverted, truncated cone, as shown in Figure 1.6. Preconditioned air is drawn through the uniform air distributor. The resulting particle motion is unrestricted and presumed random. The nozzle is positioned above the fluidised bed and sprays the coating liquid countercurrently (or downward) onto the fluidised particles.

![Diagram](https://example.com/diagram.png)

**Figure 1.6.** The top-spray fluidised bed coating configuration

Due to the vigorous and random fluidisation behaviour, controlling the distance the droplets travel before impacting on the core particles is difficult. Consequently, premature droplet evaporation can be quite severe and coating imperfections can occur. Coating quality and the extent of premature droplet evaporation are related, because, the higher the degree of drying, the more viscous the droplets get before impacting the particle substrate, resulting in a degradation of the coating solution’s film-forming properties (Jones, 1988a). The spray dried coating material is collected by the filter system at the top of the reactor and should be considered a loss (Dewettinck and Huyghebaert, 1998). Since a top-spray produced film (or coating layer) always involves imperfections to a certain extent, this method is less suited for
controlled release products, but is nowadays primarily used for barrier (protective) coatings (Eichler, 1989).

1.3.3.2. Bottom-spray fluidised bed coating

This method is also known as the Wurster system. Originally designed to coat pharmaceutical tablets, the process is now widely used for substrates as small as 50 µm (Cheng and Turton, 2000a, b). The coating chamber contains an unbaflled, open-ended cylinder known as the partition, as shown in Figure 1.7. The size of the gap between the partition and the air distributor is adjustable. The aim of the partition is to create ordered particle movement.

Next to the partition, the air distributor is a key element in maintaining this ordered particle movement. The inner section of the air distributor – which corresponds to the cross-sectional surface of the partition projected onto the air distributor – has been provided with large-diameter holes, compared to the outer section of the air distributor (Figure 1.7). Consequently, higher air velocities are created in the centre of the reactor, through the partition. Particles entering the central partition are pneumatically transported through the partition (Jones, 1988a). Once the particles have exited the central partition, they fall back along the outer section of the reactor chamber – which has much lower air flow rates due to the typical construction of the air distributor – allowing recirculation of particles (Saadevandi and Turton, 2004). Droplet/particle contact is established in the partition by placing the nozzle in the centre of the air distributor, which sprays concurrently to the particles.

Compared to the top-spray configuration, the bottom-spray configuration produces coating films which are more uniform and have a superior quality (less imperfections), thus bottom-spray is better suited for the production of controlled release encapsulated products. This could be explained by the fact that the particle motion is controlled – i.e., the number of passages and the time spent during each passage through the spraying region – as opposed to the top-spray configuration, where particles motion is presumably random and uncontrolled. Furthermore, droplets are deposited concurrently, assuring minimum droplet travel distance. Due to the extremely short droplet travel distance, premature droplet evaporation is almost absent. The film-forming droplets can spread out at the lowest viscosity, producing a very dense film with a superior physical quality (Mehta and Jones, 1985).
1.3.3.3. **Tangential-spray fluidised bed coating**

The third, and most recently developed fluidised bed coating configuration is the tangential-spray configuration. Instead of using an air distributor, a solid, rotating disc is installed at the base of the reactor (Figure 1.8). A narrow gap exists between the reactor wall and the rotating disc, through which the process air is drawn. The particle motion resulting from the combination of the lift force created by the upward flowing process air, gravity, and the friction force created by the rotating disc, could best be described as a spiralling helix (Jones, 1988b).

Beneath the surface of the rapidly tumbling bed, one or more nozzles are positioned to spray the coating liquid tangentially to and concurrently with the flow of particles. As a result of short droplet travel distances (concurrent spray) and the rapid tumbling of the particle bed, high quality coating films are obtained which are comparable or even superior to those produced by the Wurster process (Jones, 1988a).
1.3.3.4. Design modifications

In recent years, a number of design modifications have been proposed, aiming at either improving coating quality and/or process efficiency. In a patent by Hüttlin (1990), the traditional air distributor has been replaced by a series of overlapping and slanted plates, arranged in a similar fashion as the blades of a gas turbine. As a result, air passing through this type of air distributor is deflected to create a toroidal gas flow. Hence, the particle bed assumes a similar toroidal or swirling motion, where the angular force of the bed expels the rising particles to the reactor wall. The centre of the bed is characterised by low gas velocities, allowing particles to recirculate towards the air distributor. This type of fluidised bed is also denoted as ‘swirling’ fluidised beds (Kamil et al., 2005). The ordered particle movement ensures controlled particle passage through the spraying region of the bed. Nozzles are installed in the bottom of the bed and their spray is directed concurrently with the particle flow across the modified air distributor.

In a recent development, Innojet Technologies proposed a rather different approach to control particle circulation (Hüttlin, 2005). The air distributor, the so-called Booster Orbiter consists of overlapping concentric rings. The process air flows through the slits formed by the overlapping rings, eccentrically towards the reactor wall. As a result, particles are directed towards the reactor wall in the lower part of the fluidised bed and lifted upwards, along the reactor wall. Through the effect of gravity, the core particles subsequently fall back in the centre on the bottom plate in a toroidal movement. This kind of particle movement has exactly the opposite direction of particle movement when compared to a traditional Wurster configuration. In combination with a nozzle with rotating head (Behzadi et al., 2005) – which creates a flat, horizontal spray curtain – the inventors claim to attain superior coating quality in terms of coating uniformity and homogeneity.
Other design modifications or novel fluidisation techniques have focussed on improving the fluidisation behaviour of fine and cohesive particles (< 100 µm). Cohesive particles (cfr. Section 1.4) tend to fluidise poorly, exhibiting channeling, lifting as a plug and forming rat holes when aerated (Watano et al., 2003). So far, several authors have successfully managed to fluidise cohesive particles by adding kinetic energy to the bed through agitation or vibration (Beeckmans and Macwilliam, 1986; Marring et al., 1994; Watano et al., 1995; Kuipers et al., 1996; Mawatari et al., 2001). A relatively new development is the so-called centrifugal fluidised bed, where the centrifugal force – as opposed to gravity – can be adjusted (by means of rotational speed of the bed) to overcome the cohesive forces between the particles. The working principle of a centrifugal fluidised bed coater/granulator is shown in Figure 1.9. The authors claim to be able to fluidise particles as small as 15 µm (Watano et al., 2003).

![Figure 1.9](image)

**Figure 1.9.** Schematic diagram of a centrifugal fluidised bed for coating and/or agglomeration (after Watano et al., 2003).

### 1.3.4. Continuous fluidised bed coating

The ever-growing need to cut production costs and to increase the throughput of coating operations in the food industry, has prompted the design of continuous fluidised bed coating systems (Teunou and Poncelet, 2002).
In processes where particle size enlargement is involved (i.e., agglomeration and granulation), the batch fluidised bed design could be easily modified to enable continuous operation. Using air classification, particles that have reached a desired size could be automatically discharged from the bed.

However, in coating operations, where the increase in particle size is small to negligible (especially in film coating), removal of coated particles by air classification is simply not possible. Therefore, the horizontal fluidised bed, as shown in Figure 1.10, was designed to enable particle coating in continuous systems (Rümpfer and Jacob, 1998). In the horizontal bed, core particles are continuously fed at one side, while the coating product is discharged at the opposite end of the bed. An additional difficulty of continuous fluidised bed coaters, compared to batch fluidised beds, is that coating uniformity and coating layer thickness depend on the residence time of the particle in the bed. This problem is aggravated when dealing with polydisperse particles. In the bed, vertical baffles could be used to increase, or to improve control of, the particle residence time and thus to reduce the required length of the horizontal fluidised bed (Teunou and Poncelet, 2002).

Aside from the increased throughputs and the cost reduction, horizontal fluidised beds also offer the advantage of realising different fluidised bed unit operations into a single contained unit, such as drying, agglomeration and coating, or using multiple binders or coating materials.

![Figure 1.10. Continuous top-spray fluidised bed coating.](image)

1.3.5. Common problems encountered in fluidised bed coating

With respect to fluidised bed coating, a multitude of problems and side-effects are likely to be encountered. When wet particles collide, a liquid bridge is formed between them (Figure 1.4). Depending on the liquid bridge strength and the kinetic energy of the colliding particles, these bridges may persist beyond the point of solidification or drying and consequently, dry
agglomerates are formed (Smith and Nienow, 1983; Saleh et al., 1999). Depending on the process variables, particle growth due to agglomeration can accelerate beyond a point at which these larger agglomerates can no longer be fluidised. This phenomenon is termed defluidisation or bed quenching and should be avoided at all costs. To avoid this side-effect agglomeration, process controllers could increase the kinetic energy of the fluidised particles, decrease the liquid feed rate or increase the drying capacity of the supplied air. However, these measures decrease the overall energy efficiency and increase production time and hence production costs (Kage et al., 1998; Dewettinck and Huyghebaert, 1999).

Second, there is the issue of spray drying of the coating solution. The exhaust air of a fluidised bed coating process is usually not saturated and consequently, premature droplet evaporation is likely to occur before the coating solution droplet adheres onto the particle surface (Jones, 1985; Hemati et al., 2003). Depending on the size and the density of the produced fines, the spray-dried coating material could be entrained with the fluidising air and subsequently collected by the filter system. In case of heavier dry fines, the spray dried coating material remains in the fluidised bed and is either agglomerated or entrapped within the coating film, resulting in coating imperfections (Smith and Nienow, 1983). Furthermore, the droplets that successfully manage to impinge on the surface of the suspended particles – in case where large spray drying losses are present – will have increased viscosity at the moment of impingement due to the severe evaporation. As a result, droplet spreading and film-forming abilities are impaired, resulting in improper, or ‘orange peel’-like films characterised by a large number of pores (Eichler, 1989). Besides the reduced coating quality, spray drying losses increase production costs due to the loss in wall material and the increased processing times required to reach the same degree of coating (i.e. coating wall thickness) compared to a process where spray drying losses are absent (Gouin, 2005).

Premature droplet evaporation is the result of complex interactions between several factors including the evaporative capacity of the process air within the spraying region, the mean droplet travel distance and velocity, the droplet impingement efficiency and the droplet adhesion probability (Dewettinck and Huyghebaert, 1998; Heinrich et al., 2003a). One of the major factors in controlling the degree of spray drying losses is the reduction of the droplet path length, usually by positioning the nozzle at the surface of the fluidised bed in case of top-spray coating processes (Jones, 1994).

Spray drying losses and agglomeration are two side effects occurring at each other’s opposite end of the bed’s drying capacity range which implies that fluidised bed coating is often characterised by a narrow operational region as illustrated in Figure 1.11.
**Theoretical background**

### Figure 1.11

Schematic representation of the relationship between particle growth kinetics in fluidised bed processing and the spraying rate and fluidisation air flow rate process variables (after Guoin, 2005).

A third problem commonly encountered in fluidised bed coating is attrition. The combined fragmentation of particles and the crumbling of the coating by attrition is the result of inter-particle and particle-to-wall collisions. Elutriation of the resulting dust-like fragments gives rise to reduced process yields. Furthermore, dust-like particles easily agglomerate with the intact core particles, resulting in an end product with altered powder properties (Guignon *et al*., 2002). Attrition depends on the kinetic energy of the particles and on the friability of both the core material and the coating. For example, crystalline materials are more prone to attrition compared to amorphous solids (Liu and Litster, 1993b; Guignon *et al*., 2003).

Besides reducing waste product formation resulting from the aforementioned problems, product temperature and controlled coating film growth are important aspects of the coating process. When applying fluidised bed coating to heat sensitive products (core or coating), the primary aim should be to keep the product temperature below a certain threshold value in order to avoid unnecessary product degradation. Furthermore, by controlling both the product temperature and the product surface humidity, the drying rate could be kept constant which is essential in maintaining the coating film quality throughout the process (Larsen *et al*., 2003). Controlled growth of coating film and variance thereof could be a necessity in the preparation of coated particles with modified release properties. Narrow distributions of the coating film thickness are often required, because small deviations in the coating film thickness could alter the release properties (Watano *et al*., 1995; Abe *et al*., 1998).
1.4. Fluidisation

1.4.1. Fluidisation phenomenon

Fluidisation occurs when a fluid (gas, but applies to liquids as well) is drawn through a bed of particulate solids resulting in the solid material assuming liquid-like properties (Gupta and Sathiyamoorthy, 1999; Teunou and Poncelet, 2002). Depending on the gas flow rate, different fluidisation regimes occur, as shown in Figure 1.12 (Grace, 1986; Kunii and Levenspiel, 1991).

![Image of fluidisation regimes]

**Figure 1.12.** Different regimes occurring when a bed of particles is subjected to an increasing air flow rate, after Kunii and Levenspiel (1991).

Starting from a fixed bed, when increasing the gas velocity, a point will be reached where the drag force exerted by the gas upon the particles counteracts the weight of the bed: this is the onset of fluidisation and the corresponding gas velocity is called minimum fluidisation velocity, $v_{\text{mf}}$. A further increase in gas velocity results in instabilities, where part of the gas bypasses the rest of the bed in the form of bubbles. These bubbles tend to coalesce and grow during their ascent. The migration of these bubbles through the bed displaces the particles in a
highly agitated fashion resulting in a fluidised bed which appears as a boiling, frothing mixture.

Increasing the gas flow results in larger bubbles, up to a point where the bubble diameter is equal to the column (or reactor) diameter resulting in a regime, called slugging fluidisation. When the gas velocity is increased to approximately the terminal velocity of the particles, the fluidisation behaviour drastically changes into what is called turbulent fluidisation. Turbulent fluidisation is characterised by turbulent particle clusters and gas voids of varying shape and size, i.e. a distinct bubble phase is no longer present (Bi et al., 2000; Smolders and Baeyens, 2001; Harriott, 2003). Finally, operating the fluidised bed at gas velocities further beyond turbulent fluidisation, entrains the bed into a disperse or dilute phase which gives rise to pneumatic transport of the solids.

1.4.2. Minimum fluidisation velocity

Fluidisation is considered to begin at the gas velocity which exerts a drag force upon the particle bed, equal to the weight of the particle bed. If the drag force is expressed as the pressure drop across the bed, \( \Delta P_{\text{bed}} \), multiplied by the bed cross-sectional area, \( A_{\text{bed}} \), then fluidisation occurs when,

\[
\Delta P_{\text{bed}} A_{\text{bed}} = M_{\text{bed}} g = A_{\text{bed}} h_{\text{bed}} \left( \rho_{p} - \rho_{f} \right) g \left( 1 - \varepsilon_{\text{mf}} \right)
\]  

(1.12)

In Eq. (1.12) is \( A_{\text{bed}} \), the cross-sectional surface of the (circular) bed, \( \rho_{p} \) is the density of the particles (in \( \text{kg m}^{-3} \)), \( \rho_{f} \) is the density of the fluidising gas (or liquid, in \( \text{kg m}^{-3} \)) and \( \varepsilon_{\text{mf}} \) is the voidage of the bed at minimum fluidisation. The pressure drop across a static bed, \( \Delta P_{\text{bed}} \) (i.e. up to the point where fluidisation occurs) as a function of the gas velocity, \( v_{f} \), can be described by the (empirical) Ergun equation (Ergun, 1952; Rhodes, 1998):

\[
\frac{\Delta P_{\text{bed}}}{h_{\text{bed}}} = 150 \left( 1 - \varepsilon_{\text{bed}} \right)^{2} \frac{\mu_{f} v_{f}^{2}}{\left( \psi_{p} d_{p}^{2} \varepsilon_{\text{bed}} \right)^{2}} + 4 \frac{\left( 1 - \varepsilon_{\text{bed}} \right) \rho_{p} v_{f}^{2}}{\psi_{p} d_{p}^{2}}
\]  

(1.13)

For small particles (\( d_{p} < 10^{-3} \text{ m} \)), the second term in (1.13) is relatively small compared to the first term. Consequently, the relation between the pressure drop across the bed, \( \Delta P_{\text{bed}} \), and the gas velocity, \( v_{f} \), is nearly linear, as is shown in Figure 1.13.
In Eq. (1.13) is $\varepsilon_{\text{bed}}$, the bed voidage at the actual gas velocity, $v_g$ ($v_g \leq v_{mf}$) while $\mu_f$ represents the fluid viscosity. Particles are characterised by their volume-weighted average diameter, $d_p$, and the particle sphericity, $\psi_p$ in Eq. (1.13). Sphericity of an irregularly shaped particle is defined as the surface of a volume-equivalent sphere divided by the particle’s surface (Yang, 2003). Using the Ergun equation at minimum fluidisation conditions, Eq. (1.12) results in,

$$
\left(\rho_p - \rho_f\right)g = 150 \frac{\left(1-\varepsilon_{mf}\right)}{\varepsilon_{mf}^3} \frac{\mu_f v_{mf}}{(\psi_p d_p)^2} + \frac{7}{4} \frac{\rho_p v_{mf}^2}{\varepsilon_{mf}^3 \psi_p d_p} \quad (1.14)
$$

Equation (1.14) could further be written as,

$$
Ar_p = 150 \frac{\left(1-\varepsilon_{mf}\right)}{\varepsilon_{mf}^3 \psi_p^2} Re_{p,mf} + \frac{7}{4} \frac{Re_{p,mf}^2}{\varepsilon_{mf}^3 \psi_p} \quad (1.15)
$$

With, $Ar_p$ in Eq. (1.15), being the dimensionless Archimedes number and $Re_{p,mf}$, being the dimensionless particle Reynolds number at minimum fluidisation:

$$
Ar_p = \frac{\left(\rho_p - \rho_f\right)g d_p^3}{\mu_f} \quad (1.16)
$$

$$
Re_{p,mf} = \frac{\rho_p d_p v_{mf}}{\mu_f} \quad (1.17)
$$

One of the problems in solving the Ergun equation to predict the minimum fluidisation gas velocity, is that the bed voidage, $\varepsilon_{mf}$, is usually unknown. Therefore, several authors have proposed simpler empirical correlations to calculate $v_{mf}$, which are usually of the following form (Kunii and Levenspiel, 1991; Gupta and Sathiyamoorthi, 1999; Colters and Rivas, 2004):

**Figure 1.13.** Typical pressure drop profile across the particle bed, as a function of gas velocity (after Kunii and Levenspiel, 1991).
For instance, the well-known Wen and Yu correlation (1966), has the coefficients $a^* = 33.7$ and $b^* = 0.0408$. Another frequently used correlation is the Broadhurst and Becker equation to calculate $\varepsilon_{mf}$ as a function of fluidum and particle properties. Minimum fluidisation velocity, $v_{mf}$, could subsequently be retrieved by substituting $\varepsilon_{mf}$ in Eq. (1.15) (Rhodes, 1998):

\[
\varepsilon_{mf} = 0.586 \psi_p^{0.72} \left( \frac{\mu_f^2}{\rho_f g (\rho_p - \rho_f) d_p^3} \right)^{0.029} \left( \frac{\rho_f}{\rho_p} \right)^{0.021}
\] (1.19)

1.4.3. Powder classification

Whether a particle can be fluidised as well as the subsequent fluidisation behaviour largely depend on the hydrodynamic properties of both fluidum and particles. In order to group particles with similar fluidisation behaviour based on particle properties – such as size and density – several powder classification schemes have been proposed. These include the Geldart (1973), the Molerus (1982), Clark et al. (1988) and the dimensionless Geldart (Rietema, 1984) classification schemes.

Although it is one of the oldest classification schemes, the Geldart (1973) scheme has been well recognised and is still often referred to in literature. In this classification scheme, powders are divided into four different groups, according to particle size and density as shown in Figure 1.14:

- Group A is designated as ‘aeratable’ particles. These powders are characterised by small mean particle sizes ($d_p < 30 \mu m$) and/or low particle densities ($\rho_p < 1.4 \text{ g cm}^{-3}$). Group A particles are easily fluidised and exhibit a wide gas velocity interval in which smooth fluidisation (i.e. in the absence of bubbles) occurs. Once the gas velocity exceeds the minimum bubbling velocity, or $v_{mb}$, bubbles start to form.
- Group B, also called ‘sand-like’ particles. These particles usually have a density and/or size being greater than typical group A particles. More specifically, the particle density ranges between 1.4 and 4.0 g cm$^{-3}$, while the particle size varies between 150 and 500 $\mu m$. Similar to group A, group B particles are easy to fluidise, but there is no gas velocity interval in which smooth fluidisation occurs. In other words, once minimum fluidisation is reached, bubbles start to form ($v_{mf} = v_{mb}$). Glass beads and coarse sand are typical examples of group B powders.
- Group C materials are ‘cohesive’. These particles are very small ($d_p < 30 \mu m$) and/or have a low density (Figure 1.14). Because of their small size, interparticle (cohesive) forces are relatively large compared to the drag forces exerted by the fluidum. Consequently, these particles are rather hard to fluidise because of slugging and
channelling phenomena. However, additional energy can be supplied to the bed – for instance, through agitation or vibration – to improve or promote proper fluidisation of cohesive powders (see also Section 1.3.3.4).

- Group D powders are called ‘spoutable’. This group comprises powders with either a high density, or large particle diameter. Group D particles typically give rise to spouted fluidised beds, in which a jet is formed which blows material out of the bed.

![Diagram of Geldart (1973) powder classification scheme](image)

**Figure 1.14.** The Geldart (1973) powder classification scheme

The boundaries between the A, B and D groups can be defined numerically (Gupta and Sathiyamoorthi, 1999),

\[
\begin{align*}
A/B\text{-boundary: } \left( \rho_p - \rho_f \right) d_p &\leq 0.225 \text{ kg m}^{-2} \quad \text{for group A} \\
B/D\text{-boundary: } \left( \rho_p - \rho_f \right) d_p^2 &\geq 10^{-3} \text{ kg m}^{-1} \quad \text{for group B}
\end{align*}
\]

With \( \rho_p \) and \( \rho_f \), the density of the particles and the fluidum, respectively, in kg m\(^{-3} \) and \( d_p \), the particle diameter (in m). No specific numerical criterion exists to describe the boundary between the A and C groups.

### 1.4.4. Bubbling fluidised beds

#### 1.4.4.1. Introduction

Bubbling fluidisation occurs when type A or type B powders are fluidised at gas velocities above the minimum bubbling velocity, \( v_{mb} \). In case of type B (sand-like) powders, the minimum bubbling gas velocity is equal to the minimum fluidisation velocity, or \( v_{mb} = v_{mf} \). On the other hand, type A (aeratable) powders have an extended interval of smooth (absence of bubbles) fluidisation between \( v_{mf} \) and \( v_{mb} \).
The presence of bubbles is the driving force of the movement of particles, and consequently the mixing behaviour in the fluidised bed. Because of their good mixing properties, the majority of fluidised beds in coating or granulation processes are operated in a bubbling regime (Senadeera et al., 2000). Special attention will be paid to bubbles and their properties because of their importance to particle dispersion and mixing.

1.4.4.2. Two-phase description of the bubbling fluidised bed

One of the simplest descriptive models to predict bed expansion and gas flow during bubbling regime is the two-phase theory of fluidisation, proposed by Toomey and Johnstone (1952) and later expanded by Davidson and Harrison (1963) and Kunii and Levenspiel (1991). This theory divides the bubbling fluidised bed into two phases: the bubbling phase and the emulsion phase. The emulsion phase refers to the suspended solid material and the interstitial gas volume between the fluidized particles. The theory states that the gas flow through the emulsion phase is equal to the minimum fluidisation gas flow rate, as shown in Figure 1.15. Any gas in excess of the minimum fluidisation gas flow rate will bypass the emulsion phase in the form of bubbles, or – with \( A_b \) being the surface of the gas distributor,

\[
\dot{V}_a = \left( \text{gas flow through emulsion phase} \right) + \left( \text{gas flow through bubble phase} \right) = A_b v_{mf} + A_b (v_i - v_{mf})
\]

(1.22)

\[v_{mf} = v_{mf} A\]

\[\dot{V} > \dot{V}_{mf}\]

**Figure 1.15.** Gas flows inside the fluidised bed according to the two-phase theory of Toomey and Johnstone (1952).

Equation (1.22) could overestimate the gas flow rate in the bubble phase as a result of two prevailing effects in the gas flow pattern: (a) significant gas flow through the bubbles and, (b) larger interstitial gas flow in the emulsion phase, typical of Geldart type A powders (Clift and Grace, 1985; Fan and Zhu, 1998). To account for this reduction in gas flow through the bubble phase, Eq. (1.22) can be modified into (Peters et al., 1982):

\[
\dot{V}_a = \left( A_b v_{mf} + (1 - y_{bu}) A_b (v_i - v_{mf}) \right) + y_{bu} A_b (v_i - v_{mf})
\]

(1.23)
In Eq. (1.23) is $y_{bu}$ the variable expressing the reduction in gas flow through the bubble phase compared to the original two-phase model of Toomey and Johnstone. For instance, for low gas velocities in the bubbling regime, the gas flow reduction, $y_{bu}$, was empirically determined by Geldart (1986):

$$y_{bu} = 2.27 Ar_p^{0.21}$$

(1.24)

1.4.4.3. Bubble size

According to Davidson and Harrison (1963), the bubbles formed during fluidisation are not spherical, but rather hemispherical (see Figure 1.16). The flattened or pushed-in bottom contains a significant amount of particles and is called the wake. As the bubble travels through the fluidised bed, it drags the wake behind it, creating a mechanism for effective particle dispersion in bubbling beds. The bubble’s gas phase largely remains within the bubble – although diffusion occurs with the gas phase within the emulsion phase at the bubble’s surface – and part of the bubble’s gas phase penetrates into the surrounding emulsion phase, creating the so-called cloud around the bubble.

![Figure 1.16. Scheme of the bubble structure.](image)

In unbaffled fluidised beds having a rather small diameter ($h_{bed} > d_{bed}$), the diameter of the bubbles increases as they rise through the bed due to coalescence. To describe the relationship between bubble size, $d_{bu}$, and its position above the gas distributor, $h_{bu}$, the empirical Mori and Wen (1975) correlation is often used, which is valid for $(v_l - v_{mf}) < 0.48 \text{ m s}^{-1}$, $0.3 \text{ m} < d_{bed} < 1.3 \text{ m}$ and $60 \mu\text{m} < d_p < 450 \mu\text{m}$.

$$d_{bu} = d_{bu,max} - (d_{bu,max} - d_{bu,b}) e^{-0.3h_{bu}/d_{bed}}$$

(1.25)

In Eq. (1.25), $d_{bu,max}$ is the largest bubble diameter attainable, i.e. when all bubbles in the bed coalesce to form a single bubble, given a fluidised bed with sufficient height; $d_{bu,b}$ is the initial diameter of the bubbles as they are formed at the gas distributor. The maximum bubble size, $d_{bu,max}$, is given by Fan and Zhu (1998):
The initial bubble diameter, \( d_{bu,b} \), depends on the type of gas distributor used to fluidise the solid material. For perforated plate distributors, the initial bubble size depends on the number of perforations and their cross-sectional area compared to the total distributor plate surface:

\[
d_{bu} = \frac{1.38}{g^{0.2}} \left( \frac{\pi d_{bed}^2}{N_{or}} \right)^{0.4} \left( v_t - v_{mf} \right)^{0.4}
\]  

(1.27)

In Eq. (1.27), the term \( \pi d_{bed}^2 / N_{or} \) represents the distributor plate area per orifice, with \( N_{or} \) the total number of perforations (orifices) in the gas distributor. Eq. (1.27) assumes that a bubble is formed onto a single perforation of the gas distributor. However, for porous gas distributors, this assumption is no longer valid as the freshly formed gas bubbles are likely to span numerous pore openings of the gas distributor. Therefore, the following equation is used to predict the initial bubble size at porous gas distributors:

\[
d_{bu,b} = 0.376 \left( v_t - v_{mf} \right)^{0.4}
\]  

(1.28)

As an alternative to the Mori and Wen equations (1975), the bubble size may also be estimated based on the Darton correlation (Darton et al., 1977; Patil et al., 2005), which is only applicable to Geldart type B powders:

\[
d_{bu} = \frac{0.54}{g^{0.2}} \left( v_t - v_{mf} \right)^{0.4} \left( h_{bu} + 4 \left( \frac{\pi d_{bed}^2}{N_{or}} \right)^{0.5} \right)^{0.8}
\]  

(1.29)

Eq. (1.29) is only applicable to perforated gas distributors. According to Darton et al. (1977), the term \( 4 \left( \pi d_{bed}^2 / N_{or} \right)^{0.5} \approx 0.03 \) to correlate the initial bubble size when using porous gas distributors.

**1.4.4.4. Bubble rise velocity**

From experiments with single rising bubbles in a fluidised bed, Davidson and Harrison (1963) found that the rise velocity of a single bubble, in the absence of wall interaction (infinite bed), could be related to its size:

\[
v_{bu} = 0.711 \left( g d_{bu} \right)^{0.5}
\]  

(1.30)

When considering multiple rising bubbles in the bed, the drag exerted by the bed on the individual bubbles is reduced compared to a single rising bubble because the bubbles carry...
each other through the particle bed. To account for the multibubble effects, Eq. (1.30) is modified into

\[ v_{bu} = v_l - v_{mf} + 0.711(g \, d_{bu})^{0.5} \] (1.31)

**1.4.4.5. Bed expansion in bubbling fluidised beds**

The total voidage of a bubbling fluidised bed, \( \varepsilon_{\text{bed}} \), consists out of two parts: the voidage created by the bubble phase and the voidage in the emulsion phase. According to the two-phase fluidised bed model of Toomey and Johnstone (1952), the voidage in the emulsion phase is equal to the bed voidage at minimum fluidisation, or:

\[ \varepsilon_{\text{bed}} = \varepsilon_{bu} + (1 - \varepsilon_{bu}) \varepsilon_{mf} \] (1.32)

The bubble voidage, \( \varepsilon_{bu} \), is obtained from Kunii and Levenspiel (1991):

\[ \varepsilon_{bu} = \frac{\dot{V}_{bu}}{A_{\text{bed}} v_{bu}} = \frac{v_l - v_{mf}}{v_{bu}} \] (1.33)
1.5. Heat and mass transfer in fluidised beds

1.5.1. Introduction

As the fluidised bed coating process is, in essence, a complex heat and mass transfer process. Only the aspects of heat and mass transfer theory of direct interest to this study will be mentioned in this section. For a complete description of the heat transfer theory, the reader is referred to specialized literature, such as Janna (2000).

1.5.2. Basic principles of heat and mass transfer

1.5.2.1. Conductive heat transfer

In solid materials, heat is transferred through conduction which involves energy transfer from higher temperature regions to those having a lower temperature by means of molecular agitation, but without actual movement of the solid material. Conduction – by means of the heat flux, \( q \) in W m\(^{-2}\) – is described by Fourier’s law as follows (Incropera and Dewitt, 2002):

\[
\bar{q}_{\text{cond}} = -\lambda \nabla T = -\lambda \left( \bar{e}_x \frac{\partial T}{\partial x} + \bar{e}_y \frac{\partial T}{\partial y} + \bar{e}_z \frac{\partial T}{\partial z} \right)
\]

(1.34)

For one-dimensional heat transfer – for example, conduction through a flat plate – Fourier’s law, Eq. (1.34), becomes:

\[
q_{\text{cond}} = -\lambda \frac{dT}{dx}
\]

(1.35)

Eq. (1.35) expresses that the heat flux, \( q_{\text{cond}} \), is proportional to the temperature gradient, \( dT/dx \). This proportionality is the so-called thermal conductivity, \( \lambda \) (in W m\(^{-1}\)K\(^{-1}\)) which is a material property, but is temperature dependant as well.

1.5.2.2. Convective heat transfer

Introduction

Figure 1.17 illustrates the development of a boundary layer when a fluid is directed along a smooth flat surface. At great distances from the surface, the fluid may be considered inviscid. However, in the region closest to the surface, the viscous forces are dominant, creating a velocity gradient \( dv/dz \), normal to the surface. This region is called the boundary layer (Schlichting et al., 2004) and its thickness, \( \delta' \), is often defined as the distance where the local velocity is 99 % of the free-stream fluid velocity (Monteith and Unsworth, 1990).
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Figure 1.17. Laminar boundary layer for a forced laminar fluid flow, parallel to a solid surface

When a surface is immersed in a fluid, the heat losses through the laminar boundary layer of uniform thickness $\delta'$ can be written as

$$q = \frac{\lambda_f}{\delta'} (T_f - T_w)$$  \hspace{1cm} (1.36)

In Eq. (1.36) is $\lambda_f$, the thermal conductivity of the fluid in W m$^{-1}$K$^{-1}$, $T_f$ is the temperature of the bulk of the fluid, while $T_w$ is the surface temperature of the solid. Eq. (1.36) is only valid in case of forced laminar flow. For other types of flow, the equation can still be used, but $\delta'$ should be considered the thickness of an equivalent rather than the real laminar boundary layer. Because the thickness of the boundary layer cannot be retrieved directly, a more useful form of Eq. (1.36) can be derived by introducing the characteristic dimension of the body (surface), $d'$:

$$q = \left( \frac{d'}{\delta'} \right) \lambda_f \left( \frac{T_f - T_w}{d'} \right)$$  \hspace{1cm} (1.37)

The ratio $d'/\delta'$ is called the dimensionless Nusselt number, $Nu$, and provides a basis for comparing the rates of convective heat loss from geometrically similar bodies immersed in a moving fluid (Monteith and Unsworth, 1990). Rearranging Eq. (1.37) gives

$$Nu = \frac{q}{\lambda_f \left( T_f - T_w \right)}$$  \hspace{1cm} (1.38)

The Nusselt number can then be defined as the ratio of the actual heat flux to the flux that would occur in a layer of motionless gas with thickness $d'$, and with the same temperature difference across the gas layer, $T_f - T_w$. 

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Many different correlations to determine the dimensionless Nusselt number have been put forward for a variety of different geometries, flow characteristics and boundary conditions (Kreith et al., 1999; Sparrow et al., 2004). In the following paragraphs, some of the relevant correlations for both natural and forced convection will be given.

Natural or free convection
In natural convection, temperature differences in the fluid result in a density gradient, which causes buoyant forces to develop. Generally, the dimensionless Nusselt number can be described using the following relationship, which in turn allows calculating the heat transfer coefficient using Eq. (1.38):

\[ Nu = a^* \left( Gr \cdot Pr \right)^{m^*} \]  

(1.39)

Where \( a^* \) and \( m^* \) are constants depending on the geometry and the type of flow. For vertical plates and cylinders, these constants are given in Table 1.2. In Eq. (1.39) is \( Gr \) the dimensionless Grashof number while \( Pr \) is the dimensionless Prandtl number:

\[ Gr = \frac{\beta_f d' \rho_f^2 \left( T_i - T_w \right)}{\mu_f^2} \]  

(1.40)

\[ Pr = \frac{C_{p,f} \mu_f}{\lambda_f} \]  

(1.41)

Where, \( \beta_f \) is the thermal expansion coefficient of the fluid (in K\(^{-1}\)), \( d' \) the characteristic dimension, \( \rho_f \) the fluid density (in kg m\(^{-3}\)), \( \mu_f \) is the fluid viscosity (in Pa s) and \( C_{p,f} \) the thermal capacity of the fluid in J kg\(^{-1}\)K\(^{-1}\). All physical properties are evaluated at the film temperature, \( (T_i + T_w)/2 \), except for the thermal expansion coefficient of the fluid, \( \beta_f \), which is evaluated at the bulk fluid temperature, \( T_f \).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>( a^* )</th>
<th>( m^* )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( Ra = Gr \times Pr )</td>
<td>( Ra &lt; 10^4 ) (laminar)</td>
<td>1.36</td>
</tr>
<tr>
<td></td>
<td>( 10^4 &lt; Ra &lt; 10^9 ) (transition)</td>
<td>0.59</td>
</tr>
<tr>
<td></td>
<td>( Ra &gt; 10^9 ) (turbulent)</td>
<td>0.13</td>
</tr>
</tbody>
</table>

Forced convection
In forced convection, the fluid flow past an immersed surface is related to the dimensionless Prandtl and Reynolds (\( Re \)) numbers. The Reynolds number is the ratio of fluid’s inertial force to viscous forces and is defined as,
In which \( v_f \) is the linear velocity of the fluid flow past the immersed surface and \( d' \) is the characteristic dimension. Again, several authors have proposed empirical correlations between the dimensionless Nusselt number and the fluid properties, flow type and geometry, which are described using the \( Pr \) and \( Re \) dimensionless numbers. Some of the correlations relevant to this work include:

- Laminar flow (\( Re < 2100 \)) in circular pipes (either horizontal or vertical) with uniform surface temperature could be described by Sieder and Tate’s correlation (Kreith et al., 2000) – with \( \Delta x \), the length and \( d \), the inner diameter of the pipe:

\[
Nu = 1.86 \left( \frac{Re Pr}{\Delta x/d} \right)^{1/3} \left( \frac{\mu_t}{\mu_w} \right)^{0.14} \text{ valid over } \begin{cases} 
0.48 < Pr < 16700 \\
\Delta x/d < (Re Pr/8)(\mu_t/\mu_w)^{0.42} \\
0.0044 < (\mu_t/\mu_w) < 9.75 
\end{cases}
\] (1.43)

- Turbulent flow (\( Re > 10000 \)) in circular pipes with uniform surface temperature could be described by the Dittus-Boelter correlation (Winterton, 1998; Janna, 2000):

\[
Nu = 0.023 Re^{4/5} Pr^{2/5} \text{ valid over } \begin{cases} 
0.7 < Pr < 160 \\
Re > 10000 \\
\Delta x/d > 10 
\end{cases}
\] (1.44)

- Flow past a single sphere with constant surface temperature. The Whitaker equation can be used to relate the average Nusselt number to the pertinent flow variables (Sparrow et al., 2004):

\[
Nu = 2 + \left( 0.43 Re_p^{1/2} + 0.06 Re_p^{3/2} \right) Pr^{2/5} \left( \frac{\mu_t}{\mu_w} \right)^{1/4} \text{ valid over } \begin{cases} 
0.71 < Pr < 380 \\
3.5 < Re_p < 7.6 \times 10^4 \\
1.0 < (\mu_t/\mu_w) < 3.2 
\end{cases}
\] (1.45)

When dealing with small temperature differences between the surface, \( T_w \), and the bulk fluid, \( T_i \), the ratio between the viscosity of the bulk fluid to the viscosity of the fluid near the submerged surface – \( \mu_i/\mu_w \) in Eqs. (1.43), (1.44) and (1.45) – is practically equal to unity.
1.5.2.3. Radiative heat transfer

The third and final mode of heat transfer, radiation, is based on emission and absorption of electromagnetic radiation between two surfaces. Unlike convection or conduction, radiation requires no physical medium to propagate heat and only depends on the relative temperatures, the geometry and the surface structure of the materials that are emitting or absorbing heat (Earle, 1983). Radiative heat transfer can be described using the Stefan-Boltzmann law:

\[
\Phi_{\text{rad}} = \sigma A \epsilon' T^4
\]

(1.46)

Where \(\sigma\) is the Stefan-Boltzmann constant, \(\sigma = 5.669 \times 10^{-8} \text{ W m}^{-2} \text{K}^{-4}\), \(A\) is the area of the emissive surface and \(\epsilon'\) is the emissivity. The emissivity is used to describe the extent to which the surface behaves similar to a blackbody (\(\epsilon' = 1\)). A blackbody is an ideal surface which emits the maximum possible energy at a given wavelength and temperature.

In case of a single surface or body at temperature \(T_w\), the heat radiated into its surroundings – which is a uniform temperature, \(T_e\) – is given by the equation (Earle, 1983),

\[
\Phi_{\text{rad}} = \sigma A \epsilon' \left( T_w^4 - T_e^4 \right)
\]

(1.47)

1.5.2.4. Heat and mass transfer analogy

A close analogy exists between convective heat and convective mass transfer owing to the fact that conduction and diffusion in a fluid are governed by physical laws of identical form, that is, Fourier’s and Fick’s laws, respectively (Kreith et al., 2000). Similar to the Nusselt number for heat transfer (Eq. (1.38)), a dimensionless Sherwood number is defined as the ratio of actual mass flux, \(m'\) in kg m\(^{-2}\)s\(^{-1}\), to the rate that would occur in a layer of motionless gas with thickness \(d'\), and with the same concentration difference across the gas layer.

\[
Sh = \frac{m'}{D \left( C_i - C_w \right)} \frac{d'}{d'}
\]

(1.48)

With \(C_i\) being the volumetric concentration (in kg m\(^{-3}\)) of the component dissolved in the bulk fluid (e.g. water vapour in air), \(C_w\), the volumetric concentration of the component near the surface where mass transfer occurs and \(D\) being the molecular diffusion coefficient (in m\(^2\)s\(^{-1}\)).

While the Nusselt number for convection can be estimated using empirical correlations based on the Reynolds and Prandtl numbers – or based on the Grashof and Prandtl numbers in case of free or natural convection – the Sherwood number is likewise calculated using the same correlations, but based on the dimensionless Reynolds and Schmidt (Sc) numbers as shown in Eq. (1.49) (Monteith and Unsworth, 1990):
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1.5.3. Heat transfer in fluidised beds

1.5.3.1. Bed-to-gas heat transfer

In Section 1.5.2.2, the empirical correlation is given for the dimensionless Nusselt number to estimate heat transfer by forced convection around a spherical body (Whittaker equation). However, Kunii and Levenspiel (1991) reported that the whole-bed Nusselt number, $Nu_{\text{bed}}$, is usually higher than the individual particle Nusselt number, $Nu_p$, due to effects of voidage and

With the dimensionless Schmidt number, $Sc$, being the ratio of momentum diffusivity (or viscosity) to mass diffusivity, or:

$$Sc = \frac{\mu_f}{\rho_i D}$$

(1.50)

For example, the (forced) convective heat transfer across a sphere with uniform surface temperature is given by the Whitaker equation – assuming $\mu_f = \mu_w$:

$$Nu = 2 + \left(0.43Re_p^{1/2} + 0.06Re_p^{2/3}\right)Pr^{2/5}$$

(1.51)

The equivalent Whitaker equation for convective mass transfer (for example, evaporation at the sphere’s surface) is equal to,

$$Sh = 2 + \left(0.43Re_p^{1/2} + 0.06Re_p^{2/3}\right)Sc^{2/5}$$

(1.52)

Generally, the relationship between the Nusselt and the Sherwood number is given by dimensionless Lewis number, or $Le$. Knowledge of the Lewis and Nusselt number allows the derivation of the Sherwood number and vice versa (heat and mass transfer analogy), or:

$$\frac{Sh}{Nu} = \left(\frac{Sc}{Pr}\right)^m = Le^m$$

(1.53)

For water vapour in air at 20°C, the Lewis number is equal to 0.96, while the constant $m^*$ in Eq. (1.53) is equal to 0.25 for laminar, and 0.33 for turbulent flows (Monteith and Unsworth, 1990).
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turbulence in bubbling fluidised beds. The experimental whole-bed Nusselt numbers varied between (Figure 1.18):

\[ 2 + 0.6Re_p^{1/2}Pr^{1/3} < \text{Nu}_{\text{bed}} < 2 + 1.8Re_p^{1/2}Pr^{1/3} \]  \hspace{1cm} (1.54)

\[ \text{Nu}_\text{bed} = 2 + 0.6Re_p^{1/2}Pr^{1/3} \]

\[ \text{Nu}_\text{bed} = 2 + 1.8Re_p^{1/2}Pr^{1/3} \]

\[ \text{Nu}_\text{bed} = 0.03Re_p^{1.3} \]

\[ \text{Nu}_\text{bed} = 100 \]

\[ \text{Re}_p \]

\[ \text{Range of } \text{Nu}_{\text{bed}} \text{ from reported literature data} \]

\[ N_{\text{Re}} \]

\[ 1.0 \]

\[ 10 \]

\[ 100 \]

\[ 10^3 \]

\[ 10^4 \]

\[ 0.1 \]

\[ 1.0 \]

\[ 10 \]

\[ 100 \]

\[ 10^3 \]

\[ 10^4 \]

\[ \text{Figure 1.18. Whole bed dimensionless Nusselt numbers (bed-to-gas heat transfer) under various flow conditions, after Kunii and Levenspiel (1991).} \]

However, at low particle Reynolds numbers (\(Re_p < 100\)), Eq. (1.54) is no longer valid as the reported experimental whole-bed Nusselt numbers are significantly lower than predicted using the correlation in Eq. (1.54). This deviation can be attributed to the effects of boundary layer reduction due to particle collision and the generation of turbulence by bubble motion and particle collision (Brodkey et al., 1991). In this Reynolds number range, the following correlation is used instead (Figure 1.18):

\[ \text{Nu}_\text{bed} = 0.03Re_p^{1.3} \]  \hspace{1cm} (1.55)

1.5.3.2. Bed-to-(vertical) surface heat transfer

Introduction

When a vertical surface is submerged into a bubbling fluidised bed, some of the rising bubbles sweep past the surface, thereby washing away the particles located there and bringing fresh particles into direct contact with the surface. These groups of particles which are continuously being swept away along the vertical surface are denoted as emulsion packets. Thus, heat is transferred from the bed to vertical submerged surface by two transport
Theoretical background

Modelling heat and mass transfer in fluidised bed coating processes

mechanisms: convective heat transfer through the bubbles and, in between the bubbles, heat is transferred through the emulsion packets (Kunii and Levenspiel, 1991).

To take into account the coexistence of bubbles and particle emulsion packets in bubbling fluidised beds, the thermal resistance from the bed to the vertical and submerged surface in the bed (for example, the inner wall), $R$ in m$^2$K W$^{-1}$, is expressed as (Wang et al., 2005):

$$\frac{1}{R} = \frac{\epsilon_{bu,w}}{R_i} + \frac{1 - \epsilon_{bu,w}}{R_p}$$  \hspace{1cm} (1.56)

In Eq. (1.56) is $\epsilon_{bu,w}$ the bubble voidage at the submerged surface, $R_i$ and $R_p$ are the thermal resistances from the bubble and the emulsion phase, respectively, to the submerged surface (see Figure 1.19).

**Figure 1.19.** Conceptual representation of the different heat transfer modes occurring at a vertical surface submerged in a bubbling fluidised bed.

**Particles to inner wall heat transfer, $R_p$**

The heat transfer from the particles to the inner reactor wall, $R_p$ in Eq. (1.56), can be further divided into two serially connected heat transfers: heat transfer by the emulsion packets and heat transfer through the combination of the wall-particle contact point and the thin gas layer surrounding the wall-particle contact point. The latter is composed of conductive and radiative heat transfer, as shown in the next equation:

$$R_p = \left( \frac{1}{R_{pcd}} + \frac{1}{R_{rad}} \right)^{-1} + R_{packet}$$  \hspace{1cm} (1.57)
In Eq. (1.57), $R_{pcd}$ is the heat transfer resistance through the particle-vertical surface contact point and the surrounding thin gas layer, $R_{rad}$ is the radiative heat transfer and $R_{packet}$ is the heat transfer resistance due to the presence of emulsion packets. The radiative heat transfer can be neglected when the bed temperature is lower than 400°C (Kunii and Levenspiel, 1991). Considering the temperature range in which fluidised bed coaters operate in the food or pharmaceutical industries being well below this threshold temperature value (Dewettinck, 1997), particle radiation is not further taken account. As a result, Eq. (1.57) can be simplified into:

$$R_p = R_{pcd} + R_{packet}$$  \hspace{1cm} (1.58)

Heat transfer through the contact point between the particle and the vertical surface can be calculated according to Yagi and Kunii (1960):

$$\frac{1}{R_{pcd}} = \frac{2\lambda_{rfw}}{d_p} + \kappa_{w}C_{p,f} \rho_{f} v_{f}$$  \hspace{1cm} (1.59)

In Eq. (1.59), $\kappa_{w}$ is the wall (or surface) mixing constant and is generally assumed to be 0.05 (Kunii and Levenspiel, 1991). The thermal conductivity for stagnant gas in the vicinity of the submerged vertical surface, $\lambda_{rfw}^{\circ}$, can be calculated according to Kunii and Smith (1960):

$$\lambda_{rfw}^{\circ} = \varepsilon_{mf, w} \lambda_{f} + \frac{(1-\varepsilon_{mf, w}) \lambda_{p}}{\tau_{w} (\lambda_{p}/\lambda_{f})^{1/3}}$$  \hspace{1cm} (1.60)

where $\tau_{w}$ represents the equivalent thickness – compared to the particle diameter – of the gas film around the surface-particle contact point. Since $\tau_{w}$ depends on variables such as particle size, particle geometry and bed voidage, it is difficult to estimate this parameter. Therefore, the thermal conductivity $\lambda_{rfw}^{\circ}$ can be calculated using a simplified expression of Eq. (1.60):

$$\lambda_{rfw}^{\circ} = \varepsilon_{mf, w} \lambda_{f} + \left(1-\varepsilon_{mf, w}\right) \lambda_{p}$$  \hspace{1cm} (1.61)

In Eqs. (1.60) and (1.61), the variable $\varepsilon_{mf, w}$ is the bed voidage at the submerged vertical surface and at minimum fluidisation gas velocity and could be assumed to be equal to the overall bed voidage at minimum fluidisation (Kunii and Levenspiel, 1991):

$$\varepsilon_{mf, w} = \varepsilon_{mf}$$  \hspace{1cm} (1.62)

Considering thermal diffusion through an emulsion packet and assuming that the properties of the emulsion are equal to those at minimum fluidisation (two phase fluidised bed model), $R_{packet}$ in Eq. (1.57) was calculated according to Mickley et al. (1961):
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\[
\frac{1}{R_{\text{packet}}} = \frac{2}{\sqrt{\pi}} \left( \frac{\lambda_i \rho_p (1 - \varepsilon_{mf}) C_{p,p} f_{bu,w}}{1 - \varepsilon_{bu,w}} \right)
\]

(1.63)

Where \( f_{bu,w} \) is the bubble frequency at the vertical surface. Generally, it is assumed to be equal to the overall bubble frequency in the bed, \( f_{bu} \) (Kunii and Levenspiel, 1991). Bubble frequency can be estimated based on the volumetric flow rate of the fluidisation gas and the empirical correlation for bubble size, as detailed in Section 1.4.4.3. Similarly, the bubble voidage at the vertical surface – \( \varepsilon_{bu,w} \) in Eq. (1.63) – is assumed to be equal to bubble voidage in the bulk of the bed, \( \varepsilon_{bu} \), and its calculation has already been given in Section 1.4.4.5. Finally, in Eq. (1.63), is \( \lambda_i^* \) the thermal conductivity for stagnant gas in the bed and is calculated as:

\[
\lambda_i^* = \varepsilon_{mf} \lambda_i + \frac{(1 - \varepsilon_{mf}) \lambda_p}{\tau (\lambda_p / \lambda_i) + 2/3}
\]

(1.64)

Similar to Eq. (1.60), the parameter \( \tau \) represents the equivalent thickness – compared to the particle diameter – of the gas film around the particle-particle contact point. Again, this parameter is difficult to assess, since it depends on variables such as particle size, particle geometry and bed packing and voidage. Consequently, the thermal conductivity \( \lambda_i^* \) was calculated using a simplified expression of Eq. (1.64), similar to Eq. (1.61):

\[
\lambda_i^* = \varepsilon_{mf} \lambda_i + (1 - \varepsilon_{mf}) \lambda_p
\]

(1.65)

Bubbles to inner wall heat transfer

In Eq. (1.56), the term \( \varepsilon_{bu,w}/R \) describes the heat transfer resistance between the gas bubbles and the inner reactor wall. The heat transfer through convection at a submerged surface in the fluidised bed depends on the geometry of the heat transfer surface. However, Baskakov et al. (1974) suggested an empirical correlation to approximate the Nusselt number, regardless of the specific surface geometry:

\[
Nu_i = 0.009 \ Ar_p^{1/2} Pr^{1/3} \quad \text{valid for} \quad 160 \mu m < d_p < 4 \text{ mm}
\]

(1.66)
2. Modelling the fluidised bed coating process: state of the art

2.1. The purpose of modelling in fluidised bed coating processes

In essence, fluidised bed coating is a complex heat and mass transfer process that involves many different microprocesses, such as droplet production, evaporation, heat transfer, droplet impingement and particle behaviour in a fluidised bed (Heinrich et al., 2003). However, little is known about the interactions between these microprocesses and how they are related to process yields, coating mass distributions and coating uniformity (Dewettinck, 1997).

In Section 1.3.2.4, an overview was given of the different particle growth mechanisms when liquid binders or wall materials are sprayed into a fluidised bed, while a summary of potential side-effects was given in Section 1.3.5. Achieving layered growth with minimal spray-drying and with suppression of side-effect agglomeration requires a careful balancing of the different thermodynamic process variables involved, such as the properties related to the supply of fluidisation air and the addition of liquid binder to the bed (Dewettinck and Huyghebaert, 1998; Guignon et al., 2003; Hemati et al., 2003; Ronsse et al., 2004; Gouin, 2005). Currently, the design and optimisation of fluidised bed coating processes are not possible without extensive trial-and-error testing due to the large amount of variables involved (Link and Schlünder, 1997; Teneu and Poncelet, 2002). Process models prove to be useful tools in understanding or clarifying the impact of the different input variables on process efficiency and could therefore substantially reduce the research and design work required for the successful coating of new products in a fluidised bed (Dewettinck et al., 1999).

Models also prove useful in process control. Process controllers could be implemented as feedback, feedforward or as predictive controllers (Haley and Mulvaney, 1995). Feedforward control is particularly interesting as it includes measuring unanticipated disturbances in process inputs and taking corrective action before these disturbances could affect the process. However, in order to be successful, feedforward control requires an accurate model of how these disturbances affect the actual coating process. An example of model-based predictive control is given by Larsen et al. (2003), where the spraying rate of the coating solution was maximised based on the in-process calculation of the degree of utilisation of the bed’s potential evaporation energy.

Finally, models could assist in the scale-up of the coating process or to evaluate new reactor designs. The scale-up of the fluidised bed coating process needs to address many issues including air flow, product uniformity, air dew-point control, distributor plate design, attrition, atomising air pressure, binder feed rate, nozzle location, coating time and batch charge (Turton and Cheng, 2005). However, models suitable for scale-up and reactor design studies are probably the most difficult to conceive, as the extent to which models can be extrapolated (e.g. to larger geometries) is limited (Guignon et al., 2002).
2.2. Modelling approach

2.2.1. Introduction

There exist several approaches to modelling process systems (Cameron et al., 2005), such as the fluidised bed coating process. At one extreme is the group of modelling methodologies which try to incorporate the fundamental physics of heat and mass transfer of multi-phase systems into the model. These kinds of models are also called ‘white-box’ models. At the other end of the spectrum are the empirical models in which the reactor (or coater) is considered a black box and arbitrary functions are used to fit the input-output data. Black-box models are quite restricted in their use: they cannot be generalised or extrapolated to other working conditions (Guignon et al., 2002). In between the black and white-box models, are the so-called ‘grey-box’ models which implement a certain level of mechanistic process description, combined with empirical functions. The majority of fluidised bed coating models, including population balance models, belong to this last category.

2.2.2. Black-box modelling

Early models for fluidised bed coating were black-box and consisted of a single heat and/or mass balance to describe the liquid sprayed fluidised bed, of which an example is given in Figure 2.1. Ebey (1987) presented one of the first thermodynamic models suitable for aqueous film coating and was capable of predicting the steady state outlet air temperature and relative humidity. However, the model did not include heat losses, nor the effect of the addition of atomisation air. Later, Dewettinck et al. (1999) developed a black-box thermodynamic model to predict the steady state thermodynamic operation point during fluidised bed coating, including the vaporisation efficiency. As opposed to Ebey (1987), both the heat loss and the effect of atomisation air release were included (Figure 2.1).

![Figure 2.1. The conceptual representation of the TOPSIM black-box model for batch fluidised bed coating processes, developed by Dewettinck (1997).](image)

More recently, Larsen et al. (2003) incorporated a dynamic model into a process control strategy for aqueous film coating of pellets in a fluidised bed, while Gouin (2005)
demonstrated a similar model, for both aqueous and non-aqueous (e.g. ethanolic) film coating processes. As these black-box models do not consider fluidised bed behaviour, models originally developed for pan coating could also be applied as well. Among these, are the models proposed by Reiland et al. (1983), Stetsko et al. (1983), Rodriguez et al. (1996) and am Ende and Berchielli (2005).

The black-box modelling approach is very useful if no significant insight is needed into the underlying physical principles of fluidised bed coating and consequently, the resulting black-box models are relatively easy to implement into process control. However, they have certain disadvantages. First, black-box models do not take fluidised bed behaviour into account as the gas and solid phases are assumed to be perfectly mixed. Consequently, these models are less suited to predict dynamic behaviour of the bed and therefore, they are commonly applied to predict the steady state thermodynamic operation point. Second, the solid phase is characterised by a single set of variables (i.e. temperature, moisture content) which is in contradiction with the large experimental temperature and humidity gradients reported in liquid-sprayed fluidised beds (Smith and Nienow, 1982; Maronga and Wnukowski, 1997a; 1998). Finally, side-effect agglomeration, premature spray drying and other yield-reducing or quality-degrading phenomena, as discussed in Section 1.3.5, cannot be easily integrated in simple black-box models.

2.2.3. Population balance modelling

In population balance modelling, the properties of the solid phase (particles) – such as particle temperature, moisture content, coating layer thickness,... – are given as statistical distributions. The population balance is a statement of continuity that describes how the distribution of one or more particle-related variables changes with time and space. It describes how the rate of variation of the number of particles in a given interval of one or more particle-related property variables can be related to the rate at which particles enter and leave that interval by the different phenomena occurring such as particle removal from or introduction to the system, coating, agglomeration, breakage ... (Saleh et al., 2003). The general form of the population balance for a continuous particulate system can be written as (Hounslow et al., 1988; Verkoeijen et al., 2002):

\[
\frac{\partial p(x_1,\ldots,x_n,t)}{\partial t} = -\sum_{i=1}^{n} \nabla \cdot \left( \frac{dx_i}{dt} p(x_1,\ldots,x_n,t) \right) + B' - D' \tag{2.1}
\]

In Eq. (2.1) is \( p(x_1,\ldots,x_n,t) \) the so-called population density function which gives the distribution of the population as a function of the different population property variables \( x_i \) and time, defined such that \( p(x_1,\ldots,x_n,t)dx_1dx_2\ldots dx_n \) is the fraction of the population between \( x_i + dx_i, x_j + dx_j, \ldots, x_n + dx_n \) at time \( t \). The population property variables \( (x_i) \) are typically classified into internal and external coordinates. The external coordinates are the
variables used to specify the location of the particles within the system, while internal coordinates refer to intrinsic properties of the particles, such as diameter, coating layer thickness, moisture content or even residence time (Burgschweiger and Tsotsas, 2004; Cameron et al., 2005).

The term $\frac{dx_i}{dt}$ in Eq. (2.1) represents the change over time of each variable $x_i$, such as the growth rate of the coating layer. It is important to note that $\frac{dx_i}{dt}$ could be time-dependant or even depend on the other population properties $x_1$, $x_2$, … $x_n$ (Immanuel and Doyle, 2005). Finally, the terms $B'$ and $D'$ represent the so-called birth and death rates. These are the source (for instance particle solidification, particle input) and sink terms (i.e. particle removal) in the population.

In fluidised bed processing, population balances have been extensively applied for granulation and agglomeration processes using aqueous binders (Waldie et al., 1987; Watano et al., 1996, Cryer, 1999; Heinrich and Mörl, 1999; Adetayo and Ennis, 2000; Saleh et al., 2003; Heinrich et al., 2002a, b; 2003a, b; 2004; Drechsler et al., 2005). More recently, hot melt granulation processes have also been modelled using population balances (Tan et al., 2004; 2005; 2006a, b). One of the reasons for the high number of research reports on fluidised bed granulation and/or agglomeration is the fact that these unit operations are increasingly being applied as continuous processes (Vervaet and Remon, 2005; Gerstlauer et al., 2006). By means of air classification, particles having reached a desired size are continuously removed (Heinrich et al., 2002b). Hence the need for particle growth models to control the granulation or agglomeration processes.

Because granulation and agglomeration are basically size enlargement processes, the population balance equation is usually based on a single internal coordinate, namely the particle diameter. Population balance-based models for aqueous film coating have been reported less frequently, but include modelling of coating of pharmaceutical tablets (Denis et al., 2003) and seeds (Liu and Litster, 1993a; Duarte et al., 2004). However, granulation models are closely related to coating models, as some authors include particle growth by layering next to growth by agglomeration (Heinrich et al., 2002b; Saleh et al., 2003)

2.2.4. Lumped-region models

Smith and Nienow (1982) obtained the temperature profiles around a nozzle which sprayed methanol on top of a fluidised bed of glass beads or aluminium oxide particles. Later, Maronga and Wnukowski (1997a; 1998) obtained both temperature and humidity profiles using a single probe, which scanned one half of a diametrical plane inside the reactor during steady coating regime. Their studies showed the existence of four different zones in the fluidised bed, delineated parallel to the air distributor plate according to their coating function (Figure 2.2): a spraying zone, which is the area closest to the nozzle and marked with high
humidity and low temperature. The drying zone, situated below the spraying zone, is characterised by high fluctuations of both temperature and humidity. The layer above the air distributor plate is the heat transfer zone, marked by a rapid decrease in temperature but with a constant humidity. And finally, between the heat transfer zone and drying zone is the so-called non-active zone, with constant temperature and constant humidity. Also, cold and humid pockets, not adjoined with the spraying zone, could be identified.

![Diagram of liquid-sprayed fluidised bed](image)

**Figure 2.2.** Distinguishing the liquid-sprayed fluidised bed into different zones, after Maronga and Wnukowski (1998).

Whereas the majority of the models mentioned in Sections 2.2.2 and 2.2.3 treat the fluidised bed as a single ‘box’, lumped-region models divide the bed into a number of sections and perfect mixing is assumed within each section (Cameron *et al*., 2005). Based on the observation of different zones (spraying, inert, drying and heat transfer zone) in a liquid sprayed fluidised bed, several lumped region models have been proposed. Early models only distinguished spraying and non-spraying zones (Wnukowski and Setterwall, 1989; Becher and Schlünder, 1997; Nakamura *et al*., 1998; Cheng and Turton, 2000a, b). Later, Maronga and Wnukowski (1998) developed a three-zone model which took into account the spraying, drying and heat transfer zone. In each zone, a population balance was constructed describing exchange of particles with neighbouring zones and particle growth due to coating mass deposition, without considering the thermodynamic aspects of fluidised bed coating.
2.2.5. Computational fluid dynamic (CFD) models

To further increase the level of detail in modelling compared to lumped region models, computational fluid dynamic (CFD) models can be used. Computational fluid dynamics is the science of predicting fluid flow coupled with heat and mass transfer and related phenomena by solving the mathematical equations that govern the conservation of mass, momentum and energy (Xia and Sun, 2002).

Basically, two approaches exist for the modelling of multiphase (e.g. gas-solid-liquid) flows (Goldschmidt et al., 2004): Eulerian (or continuum) and Lagrangian (or discrete element) models. In Eulerian models, both the solid and gas phases are considered a continuum and empirical equations have to be applied to introduce fluid-particle drag into the model (Gidaspow et al., 2004). Moreover, with regard to modelling granulation and/or coating, constitutive relations for particle–particle collisions, droplet–particle coalescence and granulation kinetics are required (Goldschmidt et al., 2003).

Discrete particle models, on the other hand, do not require additional closure equations for the suspended particulate phase since they compute the motion of every individual particle, taking collisions and external forces acting on the particles directly into account (Goldschmidt et al., 2003). Since discrete element models describe particulate motion in more detail, it is expected that these models show superior resemblance with experimental results. However, in order to be representative of (industrial) fluidised bed coating and/or granulation processes, Lagrangian models require a minimum number of particles in the simulation, often resulting in numerically intensive simulations (Graham and Moyeed, 2002; Goldschmidt et al., 2003).

Other approaches in modelling liquid-sprayed fluidised bed include combining population balance models with Eulerian models, as demonstrated by Tan et al. (2004)
3. Combined thermodynamic and population balance of the fluidised bed coating process

3.1. Introduction

In the previous chapter, an overview of the different existing approaches to the modelling of the fluidised bed coating process was given. In this chapter, a new model will be presented that should provide perspectives in optimising the coating process. The process optimisation could be quantified using different performance criteria such as (Teunou and Poncelet, 2002):

- Coating efficiency, which relates the amount of coating material, deposited on the particle surface, to the total amount of coating material introduced during the process (Dewettinck and Huyghebaert, 1999; Donida and Rocha, 2002; Vieira et al., 2004; Donida et al., 2005).
- Core material efficiency, which relates to the amount of core material which is lost or is considered out-of-specification due to the occurrence of attrition or agglomeration throughout the coating process.
- Energy efficiency, expressing the degree to which the drying capacity and the thermal energy of the heated inlet air are consumed during the coating process (Kage et al., 1996; Larsen et al., 2003).
- Coating morphology and quality; although coating morphology is difficult to quantify, variables such as film thickness uniformity, homogeneity of film density and the fraction of uncoated particle surface could be used as performance criteria.
- Throughput, expressing the batch size of core material that can be handled per unit of process time.

In order for the proposed model to be useful in process optimisation, the following requirements were put forward:

- The model has to predict the coating process dynamically and not just under steady state conditions, allowing the model to be applied for process optimisation during transitional regimes (i.e. start-up).
- The model has to be capable of predicting at least one of the aforementioned performance criteria.
- Prediction of a single reactor temperature and/or air humidity is insufficient, especially when the model is to be used in studying the effect of alternative reactor geometries on process efficiency or in scale-up studies. Consequently, the prediction of the spatial distribution of the thermodynamic variables (such as air temperature, air humidity, particle temperature and particle surface moisture content) is a required capability of the model.
• Besides predicting the thermodynamic properties of the gas phase (air) and the core particles, the model should also be capable of calculating the dynamic coating mass distributions (with the coating mass being the amount of coating material each individual particle receives during the coating process).

In this chapter the development of a model, fulfilling all of these 4 requirements to a certain extent, will be discussed. Next, the model will be validated using experimental thermodynamic data from a lab-scale pilot plant. Once validated, the model will be used to evaluate the effects of operational parameters on relevant thermodynamic variables, such as outlet air temperature and outlet air humidity as well as the effects on particle-related properties, including the particle temperature, moisture content and coating mass distributions.
3.2. Model description

To develop the model, the fluidised bed was horizontally partitioned into a finite number, \( n \), horizontal sections or control volumes, \( S_i \), each having a constant volume \( V_{\text{bed}}/n \) – implying a diminishing height per control volume, \( h_{S,i} \), in case of partitioning a fluidised bed having a truncated cone geometry, as shown in Figure 3.1. As the fluidised bed reactor for top-spray coating usually has the geometry of an inverted truncated cone, the control volumes will also have the shape of a flat, truncated cone. Figure 3.1 shows the discretisation of the bed into different control volumes. It should also be noted that the modelled geometry is not limited to tapered fluidised beds, as shown in Figure 3.1, but different geometries, such as cylindrical fluidised beds could also be included.

![Figure 3.1. Discretisation of the fluidised bed into different control volumes.](image)

In each control volume, both the gas (air) phase and the solid (particles) phase were modelled. Thermodynamic properties of the gas phase within each control volume were considered to be uniform, the gas phase was characterised by a single temperature, \( T_{a,i} \), and a single relative humidity, \( \varphi_{a,i} \), as demonstrated in Figure 3.3. In a first instance, the solid phase within each control volume was considered uniform and the model’s heat and mass balance equations were based on the local averaged particle temperature, \( T_{p,i} \), the local averaged particle moisture content, \( W_{p,i} \), and the local averaged particle coating mass content, \( Y_{p,i} \). In a later stage (see Section 3.4), the solid phase within each control volume was modelled as a population and consequently, the particle-related variables, were treated as statistic distributions within each control volume.

To complete the modelling of the fluidized bed coating process additional assumptions were made:
1. Particles and air in each control volume are perfectly mixed.
2. All particles have the same diameter $d_p$.
3. Both the size of each control volume and the mass of particles contained within each control volume are constant. Assuming a monodisperse particle population implies a constant number of particles in each control volume.
4. Each control volume continuously exchanges particles with its neighbouring control volumes, as shown in Figure 3.2. The rate at which particles are transferred from control volume $S_i$ towards control volume $S_{i+1}$ is equal to the particle transfer rate from $S_{i+1}$ towards $S_i$. The rate at which particles are exchanged is expressed by $r_i$, as the fraction of the particle population exchanged between control volumes $S_i$ and $S_{i+1}$, per time unit.
5. Particles are mechanically inert; there is neither attrition nor agglomeration.
6. The droplets containing the coating solution have only a limited penetration depth into the bed and consequently, particle-droplet contact takes place in a limited volume of the fluidised bed (Smith and Nienow, 1982; Maronga and Wnukowski, 1997a). Therefore, distinction has to be made between coating – where both droplets and particles coexist – and non-coating control volumes. As the model was developed for top-spray fluidised bed coating, the upper $c$ out of a total number of $n$ control volumes were chosen as coating control volumes as demonstrated in Figure 3.2. Furthermore, it was assumed that all spraying liquid is collected on the particles without premature droplet evaporation. The spraying liquid is uniformly deposited on all particles in each coating control volume.
7. The weight of the coating mass added to the particle is small compared to the weight of the particle itself. Consequently, the weight of each individual particle was assumed constant throughout the process. Similarly, the thickness of the deposited coating film is small compared to the particle diameter, therefore the particle diameter was assumed to be constant.
8. The mass flow of dry air is constant and is the same for all control volumes.
9. The air exhaust is at atmospheric pressure. The pressure drop across the fluidised bed is small compared to the overall atmospheric pressure. Consequently, the drying process was assumed to take place at constant atmospheric pressure.
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Figure 3.2. Scheme of the overall model including the mass flows of gas (air), liquid and solid (particles) phase.

3.3. Heat and mass balances

3.3.1. Gas and particle-related balances

As already stated in assumption (6) in Section 3.2 of this chapter, a distinction was made between coating and non-coating control volumes. A detailed overview of the variables involved within a single coating and in a non-coating control volume is given in Figure 3.3. Furthermore, it was assumed that both the coating solution and the atomisation air were homogeneously distributed within all the coating control volumes. Thus, each coating control volume receives coating solution and atomisation air at a rate of \( \dot{M}_{\text{sol}}/c \) and \( \dot{G}_{\text{at}}/c \), respectively. By taking the distinction between a coating and a non-coating control volumes into account, the following equations were obtained for the dynamic heat and mass balances for dry air, moisture, particles and coating material:
Figure 3.3. Detail of the two types of control volumes used in the model: (a) the non-coating control volume $S_i$, $1 \leq i \leq n - c$ and (b), the coating control volume $S_i$, $n - c < i \leq n$.

Particles mass balance in a single control volume (solid phase continuity equation). The mass balance for the core particles in a control volume $S_i$ accounts for the change in particle mass contained in $S_i$ due to outboard particle transfer from $S_i$ towards $S_{i-1}$ or $S_{i+1}$ and inbound particle transfer from $S_{i-1}$ or $S_{i+1}$ towards $S_i$. Considering assumptions (3) and (4) in Section 3.2, the particle mass balance could be written as

$$\frac{dM_{pp,i}}{dt} = \left( r_{i-1} M_{pp,bed} + r_i M_{pp,bed} \right) - \left( r_{i-1} M_{pp,bed} + r_i M_{pp,bed} \right) = 0$$  \hspace{1cm} (3.1)$$

In Equation (3.1) is $M_{pp,i}$ the total mass of particles in control volume $S_i$, while $M_{pp,bed}$ represents the total mass of core particles in the fluidised bed.

Air mass balance in a single control volume (gas phase continuity equation). The only additional factor to take into consideration in the air mass balance is the release of atomisation air in the coating control volumes:

Modelling heat and mass transfer in fluidised bed coating processes
Moisture balance of the particles. In each non-coating control volume the balance of moisture on the surface of the core particles \( W_{p,i} \) is governed by the amount of water introduced by the particles entering \( S_i \) from \( S_{i-1} \) or \( S_{i+1} \), the amount of water removed by the particles leaving \( S_i \) to \( S_{i-1} \) or \( S_{i+1} \) and the water evaporated from or condensed on the particle surface:

\[
M_{pp,i} \frac{dW_{p,i}}{dt} = r_{s,i} M_{pp,bed} W_{p,i} + r_i M_{pp,bed} W_{p,i+1} - W_{p,i} M_{pp,bed} (r_{i-1} + r_i) - r_{D,i} M_{pp,i} \quad 1 \leq i \leq n - c
\]

In Equation (3.4) is \( r_{D,i} \) the drying rate of the coating solution deposited onto the particle surface; it is expressed as mass unit of evaporated water per time unit and per core particle mass unit.

In a coating control volume, the application of coating solution onto the fluidised particles needs to be taken into account. Assuming no droplet evaporation prior to droplet-particle contact and uniform distribution of the coating solution over the upper \( c \) coating control volumes, the particle surface moisture balance for a coating control volume becomes:

\[
M_{pp,i} \frac{dW_{p,i}}{dt} = r_{s,i} M_{pp,bed} W_{p,i} + r_i M_{pp,bed} W_{p,i+1} - W_{p,i} M_{pp,bed} (r_{i-1} + r_i) - r_{D,i} M_{pp,i} + \frac{1}{c} (1 - DM_{sol}) M_{sol}
\]

Gas phase (air) moisture balance. The change in air moisture in a non-coating control volume \( S_i \) is determined by the incoming moisture in the process air from \( S_{i-1} \), the moisture in the process air leaving towards \( S_{i+1} \) and the amount of water evaporated on the particle surface. For a coating control volume, the contribution of moisture from the atomisation air had to be added to the moisture balance, resulting in:

\[
M_{a,i} \frac{dX_{a,i}}{dt} = G_{a,i} X_{a,i-1} - G_{a,i} X_{a,i} + r_{D,i} M_{pp,i} \quad 1 \leq i \leq n - c
\]

\[
M_{a,i} \frac{dX_{a,i}}{dt} = G_{a,i} X_{a,i-1} - G_{a,i} X_{a,i} + \frac{1}{c} G_{at} X_{at} + r_{D,i} M_{pp,i} \quad n - c < i \leq n
\]

In Eqs. (3.6) and (3.7) is \( M_{a,i} \) the mass of dry air present in the control volume \( S_i \), while \( X_{a,i} \) is the absolute air humidity in \( S_i \).

Coating mass balance of the particles. According to Bodrov and Minaev (1987), the amount of coating liquid that settles on a fluidised particle is assumed to be proportional to the
particle surface and its residence time inside the spraying region. In the model, the spherical particles were assumed to have the same diameter and surface. Consequently, all particles located inside a single coating control volume were assumed to receive an equal amount of coating mass. Hence, the equation for the coating mass balance is

\[ M_{pp,i} \frac{dY_{p,i}}{dt} = r_{i,i-1}M_{pp,bed}Y_{p,i-1} + r_iM_{pp,bed}Y_{p,i+1} - r_1 M_{pp,bed} \left(r_{i+1} + r_i\right) \]

\[ 1 \leq i \leq n - c \quad (3.8) \]

\[ M_{pp,i} \frac{dY_{p,i}}{dt} = r_{i,i-1}M_{pp,bed}Y_{p,i-1} + r_iM_{pp,bed}Y_{p,i+1} - r_1 M_{pp,bed} \left(r_{i+1} + r_i\right) \]

\[ n - c < i \leq n \quad (3.9) \]

In Eqs. (3.8) and (3.9) is \( Y_{p,i} \) the average coating mass (dry matter), expressed as unit mass of coating material per unit mass of core particles.

**Particle heat balance.** Prior to constructing the particle heat balance, the dimensionless Biot numbers were calculated to verify whether internal temperature gradients in the particles play a significant role in the heat transfer between the gas and the solid (particles) phase. The dimensionless Biot number compares the internal heat transfer resistance due to conduction \( (\frac{d_p}{\lambda_p}) \) with the heat transfer resistance due to convection and radiation at the particle surface \( (\alpha_{p,i}) \), or (Kreith et al., 2000):

\[ Bi_{p,i} = \frac{\alpha_{p,i} \frac{d_p}{\lambda_p}}{\lambda_p} \]

(3.10)

When the Biot number is small \( (Bi_{p,i} << 0.1) \), then the internal heat transfer resistance is negligible compared to the heat transfer resistance at the particle surface and consequently, the particle can be considered to be isothermal (Janna, 2000, Collier et al., 2004).

An example of the Biot number of glass beads \( (\lambda_p = 1.3 \text{ W/m.K}) \) is given in Figure 3.4. The Biot number was calculated for different particle diameters, which typically range between 50 and 1000 µm in fluidised bed coating, and for 3 different fluidising air velocities. For a detailed calculation procedure of the heat transfer coefficient at the particle surface, \( \alpha_{p,i} \), the reader is referred to Section 3.3.3. As can be seen in Figure 3.4, the Biot numbers are typically small \( (Bi_{p,i} << 0.1) \) and consequently, particles were considered isothermal and were consequently represented by a single temperature in the heat balance. However, when dealing with larger particles, i.e. larger than 1000 µm, which is typical for pharmaceutical pellet and tablet coating (Cole et al., 1995), the internal temperature gradient has to be included in modelling heat transfer, rendering the model significantly more complex.
Radiative heat transfer within fluidised beds is negligible compared to convective heat transfer when operating at temperatures below 400 K (Kunii and Levenspiel, 1991). Considering the temperature range at which fluidised bed coaters generally operate being much lower than this threshold value, only convection was considered as the particle/gas heat transfer mechanism. More details the calculation of heat transfer between the gas phase and the solid (particle) phase is given in Section 3.3.3.

The enthalpy balance of the particles within any control volume $S_i$ depends on the enthalpy of the particles entering and leaving $S_i$, the convective heat transfer between the air and the particles, the latent heat of evaporation of water on the particle surface and the heat losses through the shell of the reactor. Since the diameter of the sprayed droplets is generally between 10 and 40 µm (Lefebvre, 1988), it was assumed that the droplets, travelling from the nozzle towards the particle surface reached wet bulb temperature ($T_{wb}$) before colliding upon the particle surface.

$$
\frac{dT_{p,i}}{dt} = \left( r_{i-1} - r_i \right) C_{p,p} M_{pp,bed} T_{p,i-1} - \left( r_i + r_{i+1} \right) C_{p,p} M_{pp,bed} T_{i+1} - r_{i+1} M_{pp,ed} T_{p,i+1} - \left( r_{i+1} + r_i \right) C_{p,p} M_{pp,bed} T_{i+1} - r_{i+1} M_{pp,ed} T_{p,i+1} - \Phi_{exo,p,i}
$$

Figure 3.4. The dimensionless Biot number as a function of particle diameter and fluidisation air velocity.
Eq. (3.11) only applies to the coating control volumes. In a non-coating control volume, the droplet-related term \( c^{-1}M_{\text{sol}}C_{\text{p,sol}}(T_{w,i} - T_{p,i}) \) is equal to zero. In Eq. (3.11) is \( A_{\text{pp,i}} \) the total surface of the solid (particle) phase in \( S_i \) and \( Q_{\text{lat,i}} \) is the latent heat of vaporisation of pure water and is given by the following equation (Iguaz et al., 2003):

\[
Q_{\text{lat,i}} = 10^3 \left( 2500.6 - 2.364356 \left( T_{p,i} - 273.15 \right) \right)
\]  

(3.12)

In a fluidised bed, besides convective heat transfer between the fluidising medium (air) and the inner reactor wall, the particles also transfer heat towards the inner reactor wall through particle-wall collisions (Kunii and Levenspiel, 1991). Consequently, the total heat loss through the wall of the control volume (\( \Phi_{\text{loss,i}} \)) is split into two parts: particles-to-environment heat losses (\( \Phi_{\text{loss,p,i}} \) in Eq. (3.11)) and fluidising air-to-environment heat losses (\( \Phi_{\text{loss,a,i}} \)).

**Gas phase (air) heat balance.** The equation for the enthalpy balance in the gas phase (air) within every coating control volume is given by the enthalpy of the air entering \( S_i \) from \( S_{i-1} \), the enthalpy of the air leaving form \( S_i \) to \( S_{i+1} \), the enthalpy of the supplied atomisation air, the heat transfer between air and particles, the heat required to heat vapour, originating from evaporated solvent (water) on the particles, to air temperature and the heat losses from the fluidising medium towards the environment:

\[
M_{\text{a,i}}C_{\text{p,a,i}} \frac{dT_{\text{a,i}}}{dt} = G_{\text{a,i}}C_{\text{p,a,i}}T_{\text{a,i-1}} - G_{\text{a,i}}C_{\text{p,a,i}}T_{\text{a,i}} + \frac{1}{c} G_{\text{at}}C_{\text{p,at}}T_{\text{at}} - a_{\text{p,i}}A_{\text{pp,i}}(T_{\text{p,i}} - T_{\text{a,i}}) - r_{\text{D,i}}M_{\text{pp,i}}C_{\text{p,v,i}}(T_{\text{a,i}} - T_{\text{p,i}}) - \Phi_{\text{loss,a,i}}
\]

(3.13)

In a non-coating control volume, the droplet-related terms are zero and consequently, the enthalpy balance equation reduces to

\[
M_{\text{a,i}}C_{\text{p,a,i}} \frac{dT_{\text{a,i}}}{dt} = G_{\text{a,i}}C_{\text{p,a,i}}T_{\text{a,i-1}} - G_{\text{a,i}}C_{\text{p,a,i}}T_{\text{a,i}} - a_{\text{p,i}}A_{\text{pp,i}}(T_{\text{a,i}} - T_{\text{p,i}}) + r_{\text{D,i}}M_{\text{pp,i}}C_{\text{p,v,i}}(T_{\text{a,i}} - T_{\text{p,i}}) - \Phi_{\text{loss,a,i}}
\]

(3.14)

In Eqs. (3.13) and (3.14) is \( C_{\text{p,a,i}} \) the specific heat of moist air in \( S_i \). It was calculated using the following equation (Becker and Isaacson, 1970):

\[
C_{\text{p,a,i}} = C_{\text{p}} + C_{\text{p,v}}X_{\text{a,i}}
\]

(3.15)

Where \( C_{\text{p}} \) is the specific heat of dry air (\( C_{\text{p}} = 10^3 \) J kg\(^{-1}\)K\(^{-1}\)) and \( C_{\text{p,v}} \) is the specific heat of water vapour (\( C_{\text{p,v}} = 1850 \) J kg\(^{-1}\)K\(^{-1}\)).
3.3.2. Heat transfer at the reactor wall

To quantify the overall heat losses in each control volume $S_i$, the reactor shell was modelled into different elements or control volumes as shown in Figure 3.5. The number of wall elements is equal to the number of fluidised bed control volumes. In each wall element, a heat balance is constructed to calculate the wall element temperature $T_{w,i}$:

$$
C_{p,w} M_{w,i} \frac{dT_{w,i}}{dt} = \left[ \text{heat inputs from bed and adjacent wall elements} \right] - \left[ \text{heat output to environment and adjacent wall elements} \right]
$$

(3.16)

Figure 3.5. The reactor wall control volume.

With the mass of the reactor wall element, $M_{w,i}$, being calculated as:

$$
M_{w,i} = \pi d_{S,i} d_w h_{S,i} \rho_w
$$

(3.17)

In Eq. (3.17) is $d_{S,i}$ the inner (bottom) diameter, $d_w$ the reactor wall thickness, $h_{S,i}$ the height of the wall element and $\rho_w$, the specific density of the reactor wall material. In modelling the wall element’s heat balance, the following interconnected heat in- and output terms were considered, an overview is given in Figure 3.6:

**Bed to inner wall heat transfer.** An overview of the heat transfer mechanisms occurring between the fluidised bed and a vertical submerged surface (such as the reactor wall) has been given in Section 1.5.3.2. To calculate the inner heat transfer resistance, $R_i$, Eqs. (1.56) to (1.66) were used. Radiative heat transfer between the bed and the inner reactor wall is neglected because of the relatively low particle temperatures ($T_{p,i} < 400$ K) and the low temperature differences between the inner wall and the particle bed. The calculations for bed to inner wall also required knowledge of the bubble frequency and bubble voidage – calculated according to the formulas in Section 1.4.4 – and, minimum fluidisation velocity and bed voidage at minimum fluidisation – which is calculated using the Ergun equation in (1.14) and the Broadhurst and Becker equation in (1.19), respectively.
Heat transfer through the reactor wall. Because of the relatively small heat transfer resistance of the reactor wall (stainless steel) compared to the heat transfer resistances from the bed towards the wall and from the wall towards the environment, the wall element was considered to have a single temperature, $T_{w,i}$. Due to the geometric nature of the wall element ($h_{S,i} \gg d_w$), vertical heat conduction to or from adjacent wall elements had to be taken into account and consequently,

$$R_{\text{cond},i} = \frac{h_{S,i} + h_{S,i+1}}{2 \lambda_w}$$  \hspace{1cm} (3.18)

Heat transfer from the wall towards the environment. At the outer reactor wall, heat is transferred to the environment by means of radiation and natural convection. The convective heat transfer was approximated by calculating the Nusselt number of a vertical cylinder which has been given in Eq. (1.39). The radiative heat loss is calculated according to Eq. (1.47).

Total heat transfer towards the environment. By combining all described heat transfers, the total heat balance, as given in Eq. (3.16), for each wall element could now be written as, with $A_{w,i}$ being the inner reactor wall surface area in the $i^{th}$ wall element, while $A'_{w,i}$ is the outer reactor wall surface area:

$$C_p w A_{w,i} d_w \frac{dT_{w,i}}{dt} = \frac{A_{w,i} \varepsilon_{bs,w} (T_{a,i} - T_{w,i})}{R_{\text{conv},i}} + \frac{A_{w,i} (1 - \varepsilon_{bs,w}) (T_{p,i} - T_{w,i})}{R_{p,i}} - \sigma \varepsilon_{w} A'_{w,i} (T_{w,i} - T_e)$$

$$\quad - \frac{A'_{w,i}}{R'_{\text{conv}}} (T_{w,i} - T_e) + \frac{\pi d_{s,i} d_w}{R_{\text{cond},i-1}} (T_{w,i-1} - T_{w,i}) - \frac{\pi d_{s,iw+1} d_w}{R_{\text{cond},i}} (T_{w,i} - T_{w,i+1})$$  \hspace{1cm} (3.19)

Figure 3.6. Overview of the different heat transfers at a single wall control volume.
In Eqs. (3.11), (3.13) and (3.14), the heat transfer from the fluidised bed towards the inner reactor wall was divided into two parts: particles-to-wall heat transfer ($\Phi_{loss,p,i}$) and gas-to-wall heat transfer ($\Phi_{loss,a,i}$). These two heat transfers correspond to the two first terms in Eq. (3.19), or:

$$\Phi_{loss,a,i} = A_{w,i} \varepsilon_{bu,w} \left( T_{a,i} - T_{w,i} \right) / R_{conv,i}$$  \hspace{1cm} (3.20)

$$\Phi_{loss,p,i} = A_{w,i} \left( 1 - \varepsilon_{bu,w} \right) \left( T_{p,i} - T_{w,i} \right) / R_{p,i}$$  \hspace{1cm} (3.21)

### 3.3.3. Estimating heat and transfer rates at particle/gas interface

In the heat and mass balances described in Section 3.3.1, both the heat transfer rate, $\alpha_{p,i}$ and the mass transfer rate (as drying rate), $r_{D,i}$ between the fluidising air and the particles within each control volume $S_i$ are still unknown. To estimate these heat and mass transfers, the following equations were used:

**Particle/gas heat transfer rate.** To estimate the convective heat transfer coefficient between the gas phase and the particles, $\alpha_{p,i}$, the Nusselt number for forced convection around a sphere was used and approximated using the Whitaker equation, Eq. (1.45). Because of the relatively small difference between the air temperature $T_{a,i}$ and the particle temperature $T_{p,i}$, the term $\mu_{a,i} / \mu_{a,i}' \approx 1$ in Eq. (1.45).

The thermal conductivity of the air $\lambda_{a,i}$ (W m$^{-1}$K$^{-1}$), required to calculate the Nusselt number, also depends on the air temperature, $T_{a,i}$. The following equation, obtained by regression analysis of thermal conductivity data from Janna (2000), was used – with $T_{a,i}$ expressed in Kelvin:

$$\lambda_{a,i} (T_{a,i}) = 1.5207 \cdot 10^{-11} T_{a,i}^3 - 4.8574 \cdot 10^{-8} T_{a,i}^2 + 1.0184 \cdot 10^{-4} T_{a,i} - 3.9333 \cdot 10^{-4}$$  \hspace{1cm} (3.22)

Finally, the air viscosity – $\mu_{a,i}$ in Eqs. (1.41) and (1.42)– was calculated based on the Sutherland-model (1895), with $T_{a,i}$ in Kelvin:

$$\mu_{a,i} (T_{a,i}) = 10^{-6} \frac{1.458 T_{a,i}^{\frac{3}{2}}}{T_{a,i} + 110.4}$$  \hspace{1cm} (3.23)

**Particle/gas mass transfer rate.** When considering the drying rate of real (porous, irregularly shaped) particulate solids, there is a rather complex relationship between the drying rate and the moisture content. Figure 3.7 illustrates a typical drying rate curve of wet solids with air of constant temperature and humidity.

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On a typical drying rate curve, the following regimes can be distinguished (Cole et al, 1995):

- Section AB or the initial transitional regime. This regime corresponds to the cooling of the surface of the drying solid material to the wet bulb temperature of the drying medium (air).
- Section BC or the constant drying rate regime. After the steady state temperature has been reached in the surface of the drying solid material, a constant drying rate is obtained. In this regime, the only limiting factor is the moisture gradient in the drying medium (air) surrounding the drying solids; moisture concentration at the surface of the solids is not limiting.
- Sections CD and DE or the first falling rate and second falling rate period, respectively. Once a critical moisture concentration has been reached, the surface of the drying solids is no longer saturated and diffusion of moisture from the bulk of the solids towards their surface becomes the limiting factor.

![Figure 3.7. Typical drying rate curve, drying rate as a function of moisture content in the solids.](image)

In modelling the drying process of the deposited coating solution on the core particles, the particles were assumed to be inert, non-porous spheres. Consequently, surface moisture concentration was assumed not to be limitative, corresponding to section ACE’ in the drying rate curve in Figure 3.7. The only limiting factor that was taken into consideration was the moisture concentration gradient between the air at the particle surface and the bulk of the fluidising air. Consequently, the drying (or condensation) rate $r_{D,i}$ was calculated as:
In Eq. (3.24) the term \( \left( T_{a,i} + T_{p,i} \right)/2 \) corresponds to the average film temperature (Campbell, 1977). The mass transfer coefficient \( \alpha'_{p,i} \) was calculated through an approximation by means of the dimensionless Sherwood number using the Whitaker equation, in Eq. (1.52).

The molecular diffusivity for water vapor in air, \( D_{v,i} \) – which is required to calculate the Schmidt number – was calculated according to Campbell (1977) as follows:

\[
D_{v,i} = 24.2 \cdot 10^{-6} \left( \frac{T_{a,i}}{293.15} \right)^{1.75} \left( \frac{10^5}{P_i} \right) \tag{3.25}
\]

in which the control volume’s pressure \( P_i \) can be approximated with \( P_i \approx P_{atm} = 101325 \) Pa as stated in the model assumptions in Section 3.2. In the calculation of the dimensionless Reynolds and Schmidt numbers, the density of the moist air, \( \rho_{a,i} \), was calculated using the ideal gas law:

\[
\rho_{a,i} = \frac{P_i \left( 1 + X_{a,i} \right)}{T_{a,i} R \left( \frac{1}{MW_a} + \frac{X_{a,i}}{MW_v} \right)} \tag{3.26}
\]

The main driving factor for drying is the moisture concentration gradient between the air at the particle surface and the bulk of the fluidising air. This moisture concentration gradient is expressed as the vapour pressure gradient \( \left( P_{v,p,i} - P_{v,a,i} \right) \) in Eq. (3.24). The vapour pressure in the fluidising air, \( P_{v,a,i} \), was calculated as

\[
P_{v,a,i} = \varphi_{a,i} P_{v,sat} \left( T_{a,i} \right) = \frac{P_i X_{a,i}}{\left[ \frac{MW_v}{MW_a} + X_{a,i} \right]} \tag{3.27}
\]

In Eq. (3.24) is \( P_{v,p,i} \) the vapour pressure at the particle surface. Assuming that the boundary gas layer at the surface of a wetted particle is saturated,

\[
P_{v,p,i} = P_{v,sat} \left( T_{p,i} \right) \tag{3.28}
\]

In Eqs. (3.27) and (3.28), is \( P_{v,sat} \) the saturated vapour pressure and can be approximated through the following equation (Campbell, 1977):
\[ P_{v,\text{sat}}(T_a) = 10^3 e^{\left(52.57633 - 6790.4985 T_a^{-1} - 5.02808 \ln(T_a)\right)} \] (3.29)

The vapour pressure at curved surfaces, including the surface of the core particles, also depends on the particle diameter and was calculated using the Kelvin-Laplace equation (Scherer, 1998):

\[
\frac{2}{(d_p/2)^2} y_{\text{water/air}} = \ln\left(\frac{P_{v,p,\text{sat}}}{P_{v,a,\text{sat}}}\right) \frac{R^2 T_p P_{\text{water}}}{MW_{\text{water}}} 
\] (3.30)

However, considering that the particle diameter typically ranges between 50 and 1000 µm, the change in particle surface vapour pressure was negligible, \( P_{v,p,\text{sat}}(T_p) / P_{v,a,\text{sat}}(T_p) << 0.01 \).
3.4. Population balance modelling

3.4.1. Introduction

In the heat and mass balance equations in Section 3.2, the solid phase or particles were attributed a single set of properties in each control volume $S_i$. These particle-related properties included the average particle temperature ($T_{p,i}$), average particle moisture content ($W_{p,i}$) and average particle coating mass ($Y_{p,i}$) in each control volume $S_i$. However, with regard to improving the control of the coating process and the suppression of yield-reducing side-reactions, the statistical distribution of these properties in the particle phase provides more relevant information than just the averaged particle-related properties as predicted by the described model thus far. Therefore, the model described in Section 3.2 was turned into a population balance model of which a brief description has been given in Section 2.2.3.

3.4.2. The deterministic population balance model

In Section 3.3, the different heat and mass transfer balances were constructed. In these balance equations, four different particle-related variables were considered:

1. The particle location or the control volume a particle resides in at time $t$
2. Particle temperature, $T_{p,i}$
3. Moisture content at the surface of the particle, $W_{p,i}$
4. Coating mass deposited onto the particle, $Y_{p,i}$

For the development of the population balance model, a balance equation analogous to Eq. (2.1) was constructed for each control volume. The particle-related variables $T_{p,i}$, $W_{p,i}$ and $Y_{p,i}$ were selected as internal coordinates. Similar to Eqs. (3.4) and (3.11), the terms describing the in- and outbound particle transfer, expressed by means of the particle exchange rate $r_i$, were included in the population balance equation for $S_i$. Thus, the following population balance equation was formed for each control volume $S_i$:

$$
\frac{\partial p_i(T_p, W_p, Y_p, t)}{\partial t} = -\frac{\partial}{\partial T_p} \left( p_i(T_p, W_p, Y_p, t) \frac{dT_p}{dt} \right) - \frac{\partial}{\partial W_p} \left( p_i(T_p, W_p, Y_p, t) \frac{dW_p}{dt} \right) - \frac{\partial}{\partial Y_p} \left( p_i(T_p, W_p, Y_p, t) \frac{dY_p}{dt} \right)$$

$$- \sum_{i=1}^{n} \frac{r_i}{n} p_{i+1}(T_p, W_p, Y_p, t) + \frac{r}{n} p_{i+1}(T_p, W_p, Y_p, t)$$

$$= \frac{(r_i + r_{i+1})}{n} p_i(T_p, W_p, Y_p, t)$$

(3.31)

In Eq. (3.31) the terms $dT_p/dt$, $dW_p/dt$ and $dY_p/dt$ represent the rate at which the internal coordinates (particle temperature, moisture content and coating mass content) of the
population change over time. These three terms provide the necessary mechanism to link the control volume’s population balance equation with the heat and mass balances of the gas (air) phase, as can be seen in the derivation of these three rate terms:

1. The particle temperature change per unit time could be retrieved from Eq (3.11):

\[
C_{p,p}M_p \frac{dT_p}{dt} = \alpha_p A_p \left(T_{a,i} - T_p\right) - r_D, iM_p Q_{\text{lat}} - \frac{\Phi_{\text{loss},p,i} M_p}{M_{pp,i}} \quad 1 \leq i \leq n - c \tag{3.32}
\]

\[
C_{p,p}M_p \frac{dT_p}{dt} = \alpha_p A_p \left(T_{a,i} - T_p\right) - r_D, iM_p Q_{\text{lat}} - \frac{1}{c} \frac{M_p M_{\text{sol}}}{M_{pp,i}} \left(T_{p,i} - T_{wb,i}\right) - \frac{\Phi_{\text{loss},p,i} M_p}{M_{pp,i}} \quad n - c < i \leq n \tag{3.33}
\]

Note that the heat balance in this equation is based on the mass \((M_p)\) and the surface \((A_p)\) of an individual particle instead of using the mass \((M_{pp,i})\) and surface \((A_{pp,i})\) of the entire solids phase present in the control volume \(S_i\), as expressed in Eq. (3.11). In fact, Eq. (3.32) is nothing more than the heat balance of a single individual particle in the control volume \(S_i\). The terms expressing heat loss and contribution of spraying liquid (at wet bulb temperature, \(T_{wb}\)) are scaled down from the control volume level in Eq. (3.11) to the individual particle level by multiplying with the factor \(M_p / M_{pp,i}\) in Eq. (3.32). Also, the heat and mass transfer rates, \(\alpha_p\) and \(r_D\), and the latent heat of vaporisation on the particle surface, \(Q_{\text{lat}}\), are based on the individual particle properties instead of the local averaged properties within each control volume \(S_i\).

2. The growth rate of the coating mass on the particles, \(dY_p/dt\), could be retrieved from Eqs. (3.8) and (3.9). Again, the equations represent the coating mass balance of a single individual particle in the control volume \(S_i\):

\[
\frac{dY_p}{dt} = 0 \quad 1 \leq i \leq n - c \tag{3.34}
\]

\[
\frac{dY_p}{dt} = \frac{1}{c} \frac{DM_{\text{sol}}}{M_{pp,i}} \quad n - c < i \leq n \tag{3.35}
\]

3. Finally, the change in moisture content at the particle surface of each individual particle \(dW_p/dt\) is given by the following equation, which is based on Eqs. (3.4) and (3.5):

\[
\frac{dW_p}{dt} = -r_D \quad 1 \leq i \leq n - c \tag{3.36}
\]

\[
\frac{dW_p}{dt} = \frac{1}{c} \frac{(1 - DM_{\text{sol}}) M_{\text{sol}}}{M_{pp,i}} - r_D \quad n - c < i \leq n \tag{3.37}
\]
Combining the population balance equation (3.31) with the rate equations in Eqs. (3.32) to (3.37) enables the solution of each control volume’s population balance equation. However, as stated in Section 3.3, the air-related heat and mass balances were also based on the local averaged particle properties within each control volume. These particle-related properties cannot be treated any longer as single variables, but have to be considered statistical distributions. Therefore, the heat and mass transfer balances of the gas phase had to be modified to couple them with the population balances:

**Gas phase (air) moisture balance.** Modifying equations (3.6) and (3.7) results in the following equations for both non-coating (Eq. (3.38)), and coating control volumes (Eq. (3.39)):

\[
\frac{dX_{a,i}}{dt} = G_{a,1}X_{a,1} - G_{a,i}X_{a,i} + M_{pp,i} \int \int \int p_i \left( T_p, W_p, Y_p, t \right) r_D dT_p dW_p dY_p
\]  
(3.38)

\[
\frac{dX_{a,i}}{dt} = G_{a,1}X_{a,1} - G_{a,i}X_{a,i} + \frac{1}{c} G_{at} X_{at} + M_{pp,i} \int \int \int p_i \left( T_p, W_p, Y_p, t \right) r_D dT_p dW_p dY_p
\]  
(3.39)

The integration of \( p_i \times r_D \) yields the cumulative drying rate of the particle population \( p_i \) at time \( t \) in the control volume \( S_i \). It is important to notice that the drying rate, \( r_D \), is not constant within each control volume, but depends on the particle temperature as well. Therefore, the \( r_D \) variable could not be placed outside of the integrated part in Eqs. (3.38) and (3.39).

**Gas phase (air) heat balance.** Modifying equations (3.13) and (3.14) results in the following equations for both non-coating (Eq. (3.40)) and coating control volumes (Eq. (3.41)):

\[
M_{a,i} C_{p,a,i} \frac{dT_{a,i}}{dt} = G_{a,1} C_{p,1} T_{a,1} - G_{a,i} C_{p,i} T_{a,i} + M_{pp,i} C_{p,v,i} \int \int \int \left( T_{a,i} - T_p \right) p_i \left( T_p, W_p, Y_p, t \right) r_D dT_p dW_p dY_p
\]  
(3.40)

\[
M_{a,i} C_{p,a,i} \frac{dT_{a,i}}{dt} = G_{a,1} C_{p,1} T_{a,1} - G_{a,i} C_{p,i} T_{a,i} + \frac{1}{c} G_{at} C_{p,at} T_{at}
\]

\[
+ M_{pp,i} C_{p,v,i} \int \int \int \left( T_{a,i} - T_p \right) p_i \left( T_p, W_p, Y_p, t \right) r_D dT_p dW_p dY_p
\]

\[
- A_{pp,i} \int \int \int \left( T_{a,i} - T_p \right) p_i \left( T_p, W_p, Y_p, t \right) \frac{dW_p}{dY_p} dT_p dW_p dY_p
\]

(3.41)

In Eqs. (3.40) and (3.41), the integration terms yield the cumulative heat transfer of the particle population \( p_i \) at time \( t \) in the control volume \( S_i \). These integration terms close the final gap between the population balances, describing the particle-related properties, and the gas phase (air) related heat and mass balances. A schematic overview of how the different balance
equations are linked together to form a mathematically solvable model is illustrated in Figure 3.8.

\[
\frac{\partial p_i}{\partial t} = -\frac{\partial}{\partial T_p} \left( p_i \frac{dT_p}{dt} \right) - \frac{\partial}{\partial W_p} \left( p_i \frac{dW_p}{dt} \right) - \frac{\partial}{\partial Y_p} \left( p_i \frac{dY_p}{dt} \right) + \frac{r_{i1}}{n} p_{i1} + \frac{r_{i1}}{n} p_{i1} - \frac{(r_i + r_{i1})}{n} p_i
\]

**Figure 3.8.** Coupling the population balance equation with the air-related heat and mass balance equations.

### 3.4.3. Towards a stochastic population balance model

#### 3.4.3.1. Problem description

The most common methods in solving population balance models are based on the discretisation of the population balance equation, which is usually in the form of a partial differential equation, such as Eq. (3.56). An extensive overview of the different discretisation schemes can be found in Ramkrishna (2000). However, many of these methods have been developed specifically for growth (agglomeration or coagulation) and break-up phenomena in particulate systems. These models typically deal with internal population coordinates which are usually one-dimensional, such as the particle diameter. By comparison, the model presented in this work is multivariate (a three-dimensional internal coordinate, consisting of \( T_p \), \( W_p \), and \( Y_p \)). Furthermore, the model consists of a set of \( n \) interconnected population balance equations. Finally, the balance equations contain terms which are connected with a set of \( n \) gas-related heat and mass balance equations with integro-differential character; some of these terms, such as the heat losses, are non-linear. As a result of the apparent model complexity, the application of a specific discretisation scheme to the population balance equations in their partial differential form is problematic. Another problem with this common solution method is, whenever the model equations are modified, it requires complete basic remodelling and reworking of the discretisation of the population balance equations.
One way to circumvent these difficulties is through the use of Monte Carlo methods. In the past, various authors have successfully made use of Monte Carlo-based techniques, especially when multivariate population balance equations have to be solved (Gooch and Hounslow, 1996; Smith and Matsoukas, 1998; Tandon and Rosner, 1999; Lee and Matsoukas, 2000; Mishra, 2000; Lin et al., 2002). Some authors even state that the Monte Carlo implementation of the stochastic approach might be the only means of exactly predicting the complete time evolution of multivariate population balances (Laurenzi et al., 2002). Monte Carlo techniques for population balance modelling utilise stochastic tools to sample a finite subset of a system in order to infer its properties. Due to the discrete nature of Monte Carlo techniques, they are especially useful in the simulation of processes that are inherently discrete such as agglomeration of particles, particle-droplet contact in coating process, etc… (Smith and Matsoukas, 1998). The main benefits of Monte Carlo-based techniques include easy implementation and modification, because discretisation problems that hinder the direct integration of the partial differential equations are not an issue (Lin et al., 2002).

However, these methods also have some serious drawbacks: First of all, the sample size of the finite subset of the system being simulated has to be large enough to be representative of the simulated population. This problem is aggravated by the fact that Monte Carlo-based techniques are numerically intensive and require long calculation times to find a solution (Zhao et al., 2005). Consequently, finding the correct sample size for the Monte Carlo is usually a trade-off between tolerable statistical error on the model-predicted results and calculation time.

3.4.3.2. Monte Carlo model and solution method

In the Monte Carlo model, a finite population of $N_{\text{sim}}$ particles was defined, representative of the entire particle population. The modelled particles were uniformly distributed over the $n$ control volumes, so each control volume contained $N_{\text{sim}}/n$ particles. The number of particles, $N_{\text{sim}}$, therefore had to be an integer multiple of $n$. Each modelled particle was described by the variables used as internal coordinates in the continuous population balance equations in Eq. (3.31), namely particle temperature ($T_p$), moisture content ($W_p$) and coating mass content ($Y_p$). Particle diameter was assumed constant, as stated in the assumptions under 3.2. The time variable was discretised into multiple fixed–length time steps $\Delta t_{\text{sim}}$ and during each time step the model fell apart into two steps: first, particle exchange between $S_i$ and its neighbouring control volumes was modelled. Next, the heat and mass transfer between the particles and the fluidising air were calculated, followed by solving the heat and mass balances for the gas phase in each control volume.

As explained in Section 3.2, the particle exchange phenomenon is described by means of the so-called particle exchange rate variable, $r_i$, which expresses the fraction of the population exchanged from control volume $S_i$ towards $S_{i+1}$ per unit time. Assuming equal particle
Combined thermodynamic and population balance of the fluidised bed coating process

diameter, the probability for a single particle to be transported to a neighbouring control volume between \( t \) and \( t + \Delta t \) is uniform. Thus, by random selection of particles and subsequent reassigning to vacant locations in the neighbouring control volumes, the particle exchange (or mixing) in the fluidised bed is modelled as shown in Figure 3.9. In this respect, this procedure integrates the population balance equation of each control volume and the solution obtained in this manner is directly comparable to that obtained from standard integration techniques.

The number of particles in \( S_i \) that have to be reassigned to the neighbouring control volumes and the number of particles in \( S_{i+1} \) and \( S_{i-1} \) that have to be reassigned to \( S_i \) is equal to

\[
N_{i \rightarrow i+1} = N_{i+1 \rightarrow i} = \text{int} \left( \Delta t_{\text{sim}} r_i N_{\text{sim}} \right) \tag{3.42}
\]

\[
N_{i \rightarrow i-1} = N_{i-1 \rightarrow i} = \text{int} \left( \Delta t_{\text{sim}} r_i N_{\text{sim}} \right) \tag{3.43}
\]

In Eqs. (3.42) and (3.43) the function \( \text{int}() \) indicates that result between brackets has to be rounded to an integer number. It is important to note that the validity of the continuity equation, more specifically the discretised form of Eq. (3.1), has to be verified:

\[
(N_{i \rightarrow i-1} + N_{i \rightarrow i+1}) - (N_{i-1 \rightarrow i} - N_{i+1 \rightarrow i}) = 0 \tag{3.44}
\]

It is important to avoid incorrect rounding of Eqs. (3.42) and (3.43), as the model will not abide any longer to the (integer) continuity equation in (3.44) and ultimately results in particle depletion or overloading in the control volume where the continuity equation is invalid.

![Figure 3.9. Simulation of particle exchange between control volumes \( S_i \) and \( S_{i+1} \).](image)

Modelling heat and mass transfer in fluidised bed coating processes
The second part of the stochastic model consists of solving all the heat and mass balance equations of the particles and the gas phase in each control volume at time \( t \). The thermodynamic calculations were split up into two sections: a particle-related and a gas-related section.

The first section dealt with all thermodynamic calculations on the particle level. First, the heat \((\alpha_p)\) and mass transfer rates \((r_D)\) for each individual particle were calculated as described in Section 3.3.3. Then, particle temperature, particle moisture content and coating mass content were recalculated for each individual particle using the balance equations in (3.32) for \( T_{p,j} \), (3.35) for \( Y_{p,j} \), (3.36) and (3.37) for \( W_{p,j} \). Note that the index \( j \) in \( T_{p,j} \) refers to the unique number, between 1 and \( N_{\text{sim}} \), each simulated particle has been given. Solving the individual particle heat balance required the knowledge of the heat loss in each control volume where the particle resides. These heat loss terms were calculated using the control volume temperatures at time \( t - \Delta t \) as the air temperature \((T_{a,i})\) was not yet known for time \( t \) in the first part of the thermodynamic calculation.

The second part dealt with the remaining thermodynamic calculations on the gas phase level. These included calculating the air temperature \((T_{a,i})\) and air relative humidity \((\phi_{a,i})\) in each control volume based on the balance equations given in Eqs. (3.38) to (3.41). However, to account for the discrete (hence, non-continuous) model, these balance equations needed to be modified, more specifically in the terms describing the local cumulated gas-particle heat and mass transfers.

The modified mass balance equations for the air moisture in the discrete model yield:

\[
M_{a,i} \frac{dX_{a,i}}{dt} = G_{a,i-1} X_{a,i-1} - G_{a,i} X_{a,i} + \frac{N_{\text{bed}}}{N_{\text{sim}}} \sum_{k=1}^{N_{\text{sim}}} M_p r_{D,E(k)} (3.45)
\]

\[
M_{a,i} \frac{dX_{a,i}}{dt} = G_{a,i-1} X_{a,i-1} - G_{a,i} X_{a,i} + \frac{1}{c} G_{a,i} X_{a,i} + \frac{N_{\text{bed}}}{N_{\text{sim}}} \sum_{k=1}^{N_{\text{sim}}} M_p r_{D,E(k)} (3.46)
\]

In Eqs. (3.45) and (3.46) is \( N_{\text{sim}} \) the total number of simulated particles, \( N_{\text{sim},i} \) the number of simulated particles in each control volume. Assuming equally sized control volumes, \( N_{\text{sim},i} = N_{\text{sim}} / h \). The operator \( E( ) \) in Eqs. (3.45) and (3.46) is the so-called enumerator operator which returns the global index number of a particle (between 1 and \( N_{\text{sim}} \)) using the local index number of a particle in \( S_i \) (between 1 and \( N_{\text{sim},i} \)). For the discrete model, the heat balance equations for the gas phase are modified into:
Combined thermodynamic and population balance of the fluidised bed coating process

Due to computational limitations, the number of simulated particles \( N_{\text{sim}} \) is necessarily smaller than the number of particles involved in the physical system being simulated \( N_{\text{bed}} \). As the cumulative particle-related transfer terms are calculated based on the number of simulated particles \( N_{\text{sim}} \) and, the gas volume (or \( M_a \)) and gas flow rates (\( G_a \)) in the gas phase-related balances are exactly dimensioned as in the physical system being simulated, the particle-related transfer terms have to be multiplied with the scaling factor \( N_{\text{bed}}/N_{\text{sim}} \) in Eqs. (3.45) to (3.48) to connect them with gas phase (air) heat and mass balances.

3.4.3.3. Random number generator selection

Stochastic models, used in large-scale Monte Carlo simulations, rely on the use of algorithms which produce random numbers, the so-called random number generator or RNG. In particular, the stochastic model (shown in Section 3.4.3.2) uses large quantities of random numbers for the simulation of particle mixing behaviour. For instance, consider an example simulation with \( N_{\text{sim}} = 10000 \) particles, \( n = 24 \) control volumes and with a particle transfer rate per control volume, \( r_i = 2 \) Hz, then simulating a coating process of 1 hour requires approximately \( 3.5 \times 10^9 \) random numbers.

However, by definition computers are deterministic in nature and can therefore never generate ‘true random’ numbers. Random number generators are nothing more than algorithms that produce long periodic sequences of numbers with certain pre-defined distribution criteria (Hellekalek, 1998), hence these generator algorithms are often denoted as ‘pseudo-random’ number generators (Knuth, 1997; Klimasauskas, 2002). The periodicity and the inevitable presence of correlations between the sequentially generated random numbers produced by a deterministic pseudo-random number generator could interfere, or even fail in certain Monte-Carlo simulations (Ferrenberg et al., 1992; Vattulainen et al., 1995; Hellekalek, 1997). Many of these RNG algorithms were conceived when computational power was a fraction of what is available today. With the ever increasing processing speed of computers, there is a tendency for large-scale Monte Carlo simulations which consume the entire period of these older RNGs in just a few seconds of calculation time resulting in possible artefacts in simulation results (Srinivasan et al., 2003).
In order to avoid aberrations or artefacts in the simulation results, different statistical tests were developed for the RNGs. These tests compare the RNG-based output of some calculation or processing with what would have been obtained with a truly random independent identically distributed number sequence (Srinivasan et al., 2003). Some of these tests even include fully standardised Monte Carlo simulations. However, passing one of these tests does not prove that the RNG is suited to be used in a specific Monte Carlo simulation. But, the more tests a proposed RNG passes, the more confidence that the RNG is suited to be used for a specific Monte Carlo simulation, although absolute certainty can never be guaranteed (L’Ecuyer, 2001). An overview of the different types of commonly used pseudo-random number generators, along with the description of some of the predominant statistical tests is given in Appendix A.

According to Klimasauskas (2002), the standard built-in RNG in Visual Basic does fail in some of these tests – listed in Appendix A – and given its short period of \(2^{24}\), this algorithm has to be discarded for use in Monte Carlo simulations. On the other hand, the Mersenne Twister has the colossal period of \(2^{19937} - 1\) and passes all tests of the Diehard testing suite (Matsumoto and Nishimura, 1998). Furthermore, the execution speed of the Mersenne Twister is comparable to the ANSI-C \texttt{RAND} generator and faster than VB’s \texttt{RND} generator, so there is no performance hit in large-scale Monte Carlo simulations when substituting these built-in generators with the Mersenne Twister. Therefore, the Mersenne Twister was selected as the RNG algorithm to provide the pseudo-random numbers in the simulation of the model presented in this Chapter.

3.4.4. Practical implementation and simulation procedure

The combined thermodynamic and population balance model, as detailed in Sections 3.2 to 3.4 and its transformation into a stochastic model as detailed in Section 3.4.3 were integrated into a tailor-made computer simulation program. For a detailed description of the different parts and procedures of the simulation program, the reader is referred to Appendix B.
3.5. Statistical issues with stochastic, discrete models: assessing model performance

3.5.1. Introduction

Prior to the analysis of the simulation results, model sensitivity studies were performed to investigate the effect of the number of control volumes \((n)\) and the number of simulated particles \((N_{\text{sim}})\) on model performance. The discrete nature of the proposed stochastic model, in the form of a limited number of control volumes and a limited number of simulated particles, combined with the use of random numbers, results in error propagation throughout the model. It is obvious that decreasing the number of control volumes \((n)\) or the number of simulated particles \((N_{\text{sim}})\) increases the variance of the model-predicted output variables. Therefore, the critical number of control volumes and the critical size of the simulated particle population needs to be assessed, which results in an acceptable error on the model-predicted output variables.

3.5.2. Methods and calculations

In analysing the impact of the sample size (number of simulated particles, \(N_{\text{sim}}\)) and the discretisation of the fluidised bed into \(n\) control volumes, distinction has to be made between two groups of modelled output variables: the control volume-based variables – including air and heat loss variables – and the particle-based variables, which are treated as statistical distributions. The predicted outlet air temperature \((T_{\text{a,out}})\) and relative humidity \((\varphi_{\text{a,out}})\) at steady state were selected to be representative of the group of variables at control volume level, while the simulated particle temperature \((T_{\text{p}})\) at steady state conditions and the coating mass distribution \((Y_{\text{p}})\) at the end of the simulated process were chosen to represent the variables at particle level. Two series of simulations were performed: the first series with a number of control volumes ranging between \(n = 8\) and \(n = 64\) while using a constant number of simulated particles, \(N_{\text{sim}} = 10000\). In all cases, the ratio of coating control volumes to the total number of control volumes \((c/n)\) was kept constant. A second series consisted of simulations with a varying number of simulated particles between \(N_{\text{sim}} = 100\) and \(N_{\text{sim}} = 15000\), while the number of control volumes was kept constant, \(n = 24\).

Each simulated process consisted of a coating process of inert glass spheres from start-up to stationary conditions using reactor and bed dimensions of the Glatt GPCG-1 (Binzen, Germany) fluidised bed unit, of which the dimensions have been specified in Table 3.1. The simulated process consisted of two distinct phases: in the heating phase, no coating solution was sprayed \((M_{\text{sol}} = 0)\). Next, after 600 seconds of simulated time – which corresponds to the time required to reach steady state conditions in the absence of spraying – a step input was given for the spraying rate \((M_{\text{sol}} = 10 \text{ g min}^{-1})\). An overview of all parameters and variables used for each simulation is listed in Table 3.1.
Some variables such as the particle exchange rate \((r)\) and the fraction of coating volumes \((c/n)\) were unknown; in Section 3.6 more details concerning the estimation of these variables are given. Other variables, such as bubble frequency \((f_{bu})\) and bed voidage at minimum fluidisation gas velocity \((\varepsilon_{mf})\) were computed prior to the simulation using the empirical correlations found in literature, which are given in Chapter 1. The results of these calculations are included in Table 3.1.

| Table 3.1. Model and operational parameters of the simulation. |
|-----------------------------|-----------------|------|
| **Variable**               | **Symbol** | **Unit** | **Value** |
| **Model parameters**       |             |       |           |
| Control volumes            | \(n\)      |       | 24*       |
| Coating control volumes    | \(c\)      |       | 3*        |
| Number of simulated particles | \(N_{sim}\) |       | 10008**   |
| Relative coating volume size, % of bed volume | \(c/n\) |       | 12.5      |
| Particle exchange rate      | \(r\)      | Hz    | 1.6       |
| **Fluidisation air properties** |       |       |           |
| Inlet air temperature      | \(T_{a,\text{in}}\) | °C  | 70        |
| Inlet air volumetric flow rate | \(G_{a,\text{in}}\) | kg s\(^{-1}\) | 1.34 \times 10\(^{-2}\) |
| Inlet air absolute humidity | \(X_{a,\text{in}}\) | kg kg\(^{-1}\) | 7.26 \times 10\(^{-3}\) |
| **Bed dimensions**         |             |       |           |
| Bed height                 | \(h_{\text{bed}}\) | m    | 0.092     |
| Reactor height             | \(h_{r}\)  | m    | 0.56      |
| Reactor bottom diameter    | \(d_{b}\)  | m    | 0.15      |
| Reactor top diameter       | \(d_{t}\)  | m    | 0.30      |
| **Bed material (glass beads)** |       |       |           |
| Overall mass               | \(M_{\text{bed}}\) | kg  | 1.5       |
| Particle diameter          | \(d_{p}\)  | µm   | 365       |
| Particle sphericity        | \(\psi_{p}\) |       | 1         |
| Specific density           | \(\rho_{p}\) | kg m\(^{-3}\) | 2600      |
| Specific heat              | \(C_{p,p}\) | J kg\(^{-1}\)K\(^{-1}\) | 837       |
| Thermal conductivity       | \(\lambda_{p}\) | Wm\(^{-1}\)K\(^{-1}\) | 0.8       |
| **Liquid spraying**        |             |       |           |
| Spraying rate              | \(M_{\text{sol}}\) | kg s\(^{-1}\) | 1.67 \times 10\(^{-4}\) |
| Dry matter content         | \(DM_{\text{sol}}\) | kg kg\(^{-1}\) | 0.10    |
| Solution temperature       | \(T_{\text{sol}}\) | °C  | 20        |
| Atomisation air mass flow rate | \(G_{at}\) | kg s\(^{-1}\) | 2.04 \times 10\(^{-3}\) |
| Atomisation air temperature | \(T_{at}\)  | °C  | 20        |
| Atomisation air pressure   | \(P_{at}\)  | bar  | 2.5       |
| Atomisation relative air humidity | \(\phi_{at}\) |       | 0.30      |
Table 3.1. Model and operational parameters of the simulation (continued)

<table>
<thead>
<tr>
<th>Variable</th>
<th>Symbol</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>External air (atmospheric) conditions</strong></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>External air temperature</td>
<td>$T_e$</td>
<td>°C</td>
<td>20</td>
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<tr>
<td>External air relative humidity</td>
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<tr>
<td>Atmospheric pressure</td>
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<td><strong>Other parameters</strong></td>
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<td>Reactor wall thickness</td>
<td>$d_w$</td>
<td>m</td>
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</tr>
<tr>
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<td>Reactor wall emittance for far-infrared radiation</td>
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<tr>
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<tr>
<td>Bed voidage at minimum fluidisation velocity</td>
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<tr>
<td>Bubble frequency</td>
<td>$f_{bu}$</td>
<td>Hz</td>
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<tr>
<td><strong>Simulation parameters</strong></td>
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<tr>
<td>Simulated time</td>
<td>$t_{sim}$</td>
<td>s</td>
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</tr>
<tr>
<td>Of which,</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>• Heating stage</td>
<td>s</td>
<td>600</td>
<td></td>
</tr>
<tr>
<td>• Spraying (coating) stage</td>
<td>s</td>
<td>900</td>
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</tr>
<tr>
<td>Simulation time step</td>
<td>$\Delta t_{sim}$</td>
<td>s</td>
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</tr>
<tr>
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<td></td>
</tr>
<tr>
<td>* The number of control volumes varied in simulation series 1, the c/n ratio remained constant</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>** The sample size ($N_{sim}$) was varied in simulation series 2</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

3.5.3. Results and discussion

3.5.3.1. Number of simulated particles

a) Control volume-based model-predicted variables

Figure 3.10 presents the average simulated steady state outlet air temperature, $\bar{T}_{a,\text{out}}$, and standard deviation, $s(T_{a,\text{out}})$ as a function of the number of simulated particles, while Figure 3.11 shows the effect on the average outlet air relative humidity, $\bar{\phi}_{a,\text{out}}$, and standard deviation, $s(\phi_{a,\text{out}})$. The number of simulated particles strongly influenced the outlet air temperature and relative humidity distribution. The effect is minimal when considering the average temperature or relative humidity, but there is a strong inverse relationship between the standard deviation on the modelled outlet air thermodynamic properties and the number of particles considered in the simulation which reached an asymptotic value of $s(T_{a,\text{out}}) = 0.044^\circ$C and $s(\phi_{a,\text{out}}) = 7 \times 10^{-4}$ at approximately $N_{sim} = 10000$. 

Modelling heat and mass transfer in fluidised bed coating processes
Using the outlet air thermodynamic properties to assess the minimum sample size, $N_{\text{sim}}$, is arguable, as varying the sample size might not alter the modelled outlet air properties, but could seriously affect the model-predicted axial profiles for air temperature and humidity. Therefore, the effect of the number of simulated particles, $N_{\text{sim}}$, on the axial temperature and humidity profiles of the fluidisation air was studied, and the results are shown in Figure 3.12 for the modelled average air temperature, $\bar{T}_{a,i}$, Figure 3.13 for the modelled standard deviation on the air temperature, $s(T_{a,i})$, Figure 3.14 for the modelled average air relative humidity, $\bar{\phi}_{a,i}$, and Figure 3.15 for the modelled standard deviation on the air relative humidity, $s(\phi_{a,i})$. Each point in all of these profile curves corresponds to a single control volume; in every simulation a number of 24 control volumes were used.

Considering the axial thermodynamic profiles ($T_{a,i}$ and $\phi_{a,i}$), there seems to be little influence, except when dealing with very small sample sizes ($N_{\text{sim}} < 2500$) when considering average air temperature and relative humidity. Again, the standard deviation ($s(T_{a,i})$ and $s(\phi_{a,i})$) is strongly correlated with the sample size and appears to be stabilising at around $N_{\text{sim}} = 10000$. Also, $s(T_{a,i})$ appears to be constant in Figure 3.13, while $s(\phi_{a,i})$ has significantly higher values for the higher positioned control volumes in Figure 3.15. This could be explained by the fact that heat transfer occurs in every part of the bed, while mass transfer (evaporation) is mainly concentrated in the upper part of the fluidised bed.

![Figure 3.10](image_url). Influence of the number of simulated particles, $N_{\text{sim}}$, on the steady state outlet air temperature, $\bar{T}_{a,\text{out}}$ (○) and standard deviation, $s(T_{a,\text{out}})$ (●).
Figure 3.11. Influence of the number of simulated particles, \( N_{\text{sim}} \), on the steady state outlet air relative humidity, \( \varphi_{a,\text{out}} \) (---) and standard deviation, \( s(\varphi_{a,\text{out}}) \) (--).
Figure 3.13. Influence of the number of simulated particles, $N_{\text{sim}}$, on the axial profile for air temperature standard deviation, $s(T_{a,i})$.

Figure 3.14. Influence of the number of simulated particles, $N_{\text{sim}}$, on the axial average air relative humidity profile, $\bar{\varphi}_{a,i}$.
b) Particle population-based model-predicted variables

Next, the effect of sample size on the model-predicted particle-related variables was studied. Figures 3.16 and 3.17 show the effect of sample size on the simulated coating mass distribution \(Y_p\) after \(t = 900\) seconds of spraying, while Figures 3.18 and 3.19 show its effect on the global particle temperature distribution after reaching steady state conditions. When dealing with particle-related properties, the variance has a different meaning than in case of control volume-based variables: here, the model-predicted variance could be considered a statistical estimator for the variance of the continuous (infinite) population of which \(N_{\text{sim}}\) particles were sampled.
Figure 3.16. Influence of the number of simulated particles, $N_{\text{sim}}$, on the average coating mass $\bar{Y}_p$, in g coating/kg core after $t = 900$ s of spraying, and standard deviation, $s(Y_p)$.

Figure 3.17. Influence of the number of simulated particles, $N_{\text{sim}}$, on the coating mass distribution $Y_p$, in g coating/kg core after $t = 900$ s of spraying.
Figure 3.18. The influence of the number of simulated particles, $N_{\text{sim}}$, on the average particle temperature at steady state, $\bar{T}_p$ (○) and standard deviation, $s(T_p)$ (●).

Figure 3.19. The influence of the number of particle, $N_{\text{sim}}$, on the particle temperature distribution, $T_p$.

Considering the coating mass and particle temperature distributions, no significant influence of the sample size could be found beyond $N_{\text{sim}} = 7500$. Combined with the results of control volume-based variables, it was concluded that for future simulations, $N_{\text{sim}} = 10000$ resulted in sufficiently low standard deviation for all output variables. A further increase of $N_{\text{sim}}$ only marginally reduced the variance of the control volume-based output variables and further
improved the particle-related property distributions, but comes with a calculation time penalty. Calculation time scaled linearly with the number of particles used in the simulation. A simulated coating process of 900 seconds using 10000 particles required a calculation time of approximately 3 hours. For every 4000 additional particles, the calculation time increased with +/- 1 hour on an Intel Pentium® IV (3.6 GHz) equipped computer.

3.5.3.2. Number of simulated control volumes

a) Control volume-based model-predicted variables

Figure 3.20 shows the influence of the number of control volumes on the simulated steady state outlet air temperature, $T_{a,\text{out}}$, while Figure 3.21 demonstrates the effect on steady state outlet air relative humidity, $\varphi_{a,\text{out}}$. There appears to be little influence concerning the number of control volumes, $n$, on the modelled outlet air thermodynamic properties. Increasing $n$ resulted in increasing the standard deviations, $s(T_{a,\text{out}})$ and $s(\varphi_{a,\text{out}})$, until a plateau was reached at about $n = 32$.

![Figure 3.20](image.png)

**Figure 3.20.** Influence of the number of control volumes, $n$, on the steady state outlet air temperature, $T_{a,\text{out}}$ (○-) and standard deviation, $s(T_{a,\text{out}})$ (●-).
Figure 3.21. Influence of the number of control volumes, \( n \) on the steady state outlet air relative humidity, \( \bar{\varphi}_{a,\text{out}} \) (○) and standard deviation, \( s(\varphi_{a,\text{out}}) \) (●).

Figure 3.22. Influence of the number of simulated control volumes, \( n \), on the axial average air temperature profile, \( T_{a,i} \).

More interesting is what happens to the axial temperature and humidity profiles with varying number of simulated control volumes. Figures 3.22 and 3.23 illustrate the simulated axial profiles of average air temperature, \( T_{a,i} \), and standard deviation, \( s(T_{a,i}) \), while Figures 3.24 and 3.25 present the axial air humidity profiles. The axial air temperature and humidity profiles were significantly biased when the number of control volumes used in the simulation was
lower than 24. Also, a low number of control volumes, artificially reduced the standard deviation on the axial thermodynamic profiles. For instance, the higher standard deviation in air humidity, $s(\phi_{a,i})$, in the upper control volumes due to evaporation is simply not visible when using a lower number of control volumes.

**Figure 3.23.** Influence of the number of simulated control volumes, $n$, on the axial profile of the standard deviation on air temperature, $s(T_{a,i})$.

**Figure 3.24.** Influence of the number of simulated control volumes, $n$, on the axial average air relative humidity profile, $\bar{\phi}_{a,i}$.
Figure 3.25. Influence of the number of simulated control volumes, n on the axial profile of the standard deviation on air relative humidity, \( s(\varphi_{a,i}) \).

b) Particle population-based model-predicted variables

Figures 3.26 and 3.27 present the relationship between the simulated coating mass distribution \( (Y_p) \) after \( t = 900 \) s of spraying and the number of modelled control volumes, \( n \). The effect of \( n \) on the modelled particle temperature distribution is given in Figures 3.28 and 3.29. Both particle temperature distribution, \( T_p \), and particle coating mass distribution, \( Y_p \), stabilised at about \( n = 24 \) to \( n = 32 \) control volumes. For instance, the asymmetry in the particle temperature due to the evaporative cooling in the upper part of the bed, only became clearly visible when \( n \geq 24 \). A further increase of the number of modelled control volumes above \( n = 24 \) or 32 had little effect on the output variables. The choice of \( n \) will then be determined by the required resolution of the model-predicted one-dimensional axial thermodynamic profiles and the available computational resources. The increase in calculation time due to a higher number of control volumes scaled linearly. A simulated coating process of 900 seconds using 24 control volumes required a calculation time of approximately 3 hours, while increasing the number of control volumes to 48 or 64 increased the calculation time to 3.5 and 4 hours, respectively. In all simulations presented in this study, a number of 24 control volumes were used.
Figure 3.26. Influence of the number of modelled control volumes, $n$, on the average coating mass, $\bar{Y}_p$ (−−), in g coating/kg core after $t = 900$ s of spraying, and standard deviation, $s(Y_p)$ (−−).

Figure 3.27. Influence of the number of modelled control volumes, $n$, on the coating mass distribution $Y_p$, in g coating/kg core after $t = 900$ s of spraying.
Figure 3.28. Influence of the number of modelled control volumes, $n$, on the average particle temperature, $\bar{T}_p$ (–) and standard deviation, $s(T_p)$ (●).

Figure 3.29. The influence of the number of modelled control volumes, $n$, on the particle temperature distribution, $T_p$.

3.5.4. Conclusions

Based on the results presented in Section 3.5.3.1 concerning the effects of sample size ($N_{\text{sim}}$), and in Section 3.5.3.2 considering the effects of the number of modelled control volumes ($n$), all future simulations in this chapter will use $N_{\text{sim}} = 10000$ and $n = 24$. However, the minimum
number for both $n$ and $N_{\text{sim}}$ should be re-evaluated when a totally different set of parameters and variables is used in the simulation. More specifically, different values for the bed dimensions ($d_b$, $d_t$, $h_t$ and $h_{\text{bed}}$), bed weight ($M_{\text{bed}}$) and particle diameter ($d_p$) could necessitate the re-evaluation of the minimum values for $n$ and $N_{\text{sim}}$. 
3.6. Model validation

3.6.1. Introduction

To validate the proposed model, experimental steady state coating data, generated by Dewettinck (1997), have been used to validate the model in this study. In these experiments, the process variables of interest were varied and their effects on the steady state thermodynamic properties of the outlet air were experimentally measured. The model-predicted results were compared against these experimental results. Also, a comparison is made between the new model and the Topsim model, which was proposed by Dewettinck (1997); Dewettinck et al., (1999). Topsim (which stands for thermodynamic operation point simulation) is a fully black-box thermodynamic model of the fluidised bed in which the bed is approached as a single homogenous volume and is restricted to the calculation of steady state conditions only (cfr. Section 2.2.2).

3.6.2. Materials and methods

3.6.2.1. Fluidised bed setup

In the validation experiments, the equipment used was a Glatt GPCG-1 lab-scale fluidised bed unit (Glatt GmbH, D). An overview of the unit is given in Figure 3.30. The principal component of this fluidised bed unit is the tapered reactor or expansion chamber. Different insertable reactor types are available, but in all of these validation experiments, the top-spray insert was selected. The dimensions of this reactor type are listed in Table 3.1.

On top of the reactor insert is the filter housing, including two bag filters which are shaken alternatively in order to reintroduce smaller particles, collected onto the filter fabric, back into the reactor. Fluidisation air is drawn through the expansion chamber by means of a suction fan located behind the bag filters. Consequently, the reactor operates in underpressure to avoid contamination of its surroundings in case of reactor leaks. The reactor unit operates using external air which is only heated prior to entry into the reactor. No conditioning of the fluidisation air to alter its humidity takes place and consequently the atmospheric (external) temperature \( T_e \) and relative humidity \( \varphi_e \) will influence the drying rate in the fluidised bed to a certain extent, an effect dubbed ‘the weather effect’ by Dewettinck (1997). The heated air enters the bed through the air distributor, which consisted of a woven metallic wire mesh. The air distributor’s aim is to retain the fluidised product in the reactor chamber and to homogenise the inlet air to ensure good (i.e. no dead spots) fluidisation. The fluidised bed’s air consumption is controlled by means of a flap situated in front of the air distributor. Inlet air flow rate at ambient temperature is measured by means of a Pitot-tube, positioned in front of the air heating coils, connected to a differential pressure gauge.
The bed or core material in this study were glass beads with a volume weighted average diameter of 365 µm (Sovitec Micropearl®, B) of which 0.75 kg were fluidised in the Glatt GPCG-1 fluidised bed coater. For thermodynamic analysis, distilled water at ambient temperature was used as the spraying liquid. The spraying liquid was atomised into the bed by means of a pneumatic nozzle (Schlick Model 970 S0 using lengthened liquid insert D). A pneumatic nozzle uses compressed air to induce high shear forces at the liquid/gas interface to create very small droplets, with diameters typically ranging between 10 and 40 µm (Lefebvre, 1988). The tip of the spraying nozzle was located approximately 0.12 m above the air distributor. The pneumatic nozzle also introduces additional air into the fluidised bed which affects the overall thermodynamic properties of the bed such as air humidity and drying rate. The atomisation air mass flow rate $G_{at}$, was obtained through measuring the linear air velocity with a Testovent 4000 anemometer (Testo, B) while blowing compressed air through the nozzle at different pressures – between 0.5 and 3.5 bar – in a cylindrical pipe with a diameter of 0.03 m. Regression analysis resulted in the following equation (Dewettinck et al., 1999):

$$\frac{dV_{at}}{dt} = \pi \left( \frac{0.03}{2} \right)^2 (-0.151 P_{at}^2 + 1.324 P_{at} + 0.127) \quad \text{with } R^2 = 0.9859$$

(3.49)

The dry air mass flow rate $G_{at}$ could then be calculated as

$$G_{at} = \frac{P_{at} \left( 1 + X_{at} \right)}{T_{at} R^\prime \left( \frac{1}{MW_a} + \frac{X_{at}}{MW_v} \right)} \frac{dV_{at}}{dt}$$

(3.50)
Finally, reactor outlet air temperature was measured at the top of the reactor by means of a stainless steel sheathed T-type thermocouple (Thermocoax, F). Steady state conditions were achieved after 15 min of spraying. In order to take a representative value for the outlet air temperature, the average value of measurements after 20, 25 and 30 min of spraying was taken. Ambient temperature and humidity were recorded using a Testo 610 (Testo, B) instrument.

### 3.6.2.2. Calibration

As already stated under Section 3.5, there are two remaining essential variables that need to be characterised in order to complete the modelling of the fluidised bed. These unknown variables are:

- The size of the spraying region, \( c/n \) (being the fraction of coating control volumes compared to the total number of control volumes)
- The particle exchange rate, \( r_i \)

Based on the conclusions with regard to the size of the actual spraying region in fluidised bed coating drawn by Maronga and Wnukowski (1997a), a coating volume with a relative size of \( c/n = 12.5\% \) was assumed, which corresponded to 3 coating control volumes out of the total of 24 modelled control volumes. The values for the particle exchange rates, \( r_i \), were based on measured axial dispersion coefficients \( (D_{p,ax}) \) found in literature. Both the particle exchange rate and the axial dispersion coefficient describe the axial mixing of particles in a fluidised bed (Fyhr and Kemp, 1999). Their relation is given by

\[
D_{p,ax} = r_i h_{bed}^2 \tag{3.51}
\]

Mostoufi and Chaouki (2001) demonstrated that the axial dispersion coefficients for sand particles with \( d_p = 385 \mu m \) and with superficial gas velocities \( (v_a - v_{mf}) \) varying from 0.5 to 2.8 m s\(^{-1}\), ranged between \( 3.3 \times 10^{-3} \) and \( 5.6 \times 10^{-2} \) m\(^2\) s\(^{-1}\), which is close to the glass beads with \( d_p = 365 \mu m \) and superficial gas velocities ranging from 0.65 to 1.06 ms\(^{-1}\) used in this study’s experimental validation. Based on their experiments, Mostoufi and Chaouki (2001) found a linear correlation between the surplus gas velocity \( (v_a - v_{mf}) \) and the axial dispersion coefficient, \( D_{p,ax} \), which could be expressed by means of the dimensionless Peclet number. For sand particles, the axial Peclet number was calculated to be

\[
P_{e,ax} = \frac{(v_a - v_{mf})d_p}{D_{p,ax}} = 0.02 \tag{3.52}
\]

Using Eqs. (3.51) and (3.52), the particle exchange rates were calculated to range between \( r = 0.81 \) s\(^{-1}\) and 1.82 s\(^{-1}\) for superficial gas velocities between 0.65 and 1.06 m s\(^{-1}\) (these values correspond to the inlet air velocities between 3 and 5 m s\(^{-1}\) in the inlet duct of the Glatt
Alternatively, the exchange rate could also be calculated based on the average circulation time, $t_{\text{circ}}$, for which an equation has been proposed by Rowe (1973) for particles in a bubbling fluidised bed:

$$r = \frac{1}{t_{\text{circ}}} = \frac{0.6(v_a - v_{mf})\left(1 - \frac{(v_a - v_{mf})}{v_{bu}}\right)}{h_{mf}}$$  \hspace{1cm} (3.53)

In Equation (3.53) is $h_{mf}$ the bed height at minimum fluidisation velocity. Using Rowe’s correlation in Eq. (3.53), the particle exchange rate, $r$ was calculated to be equal to 1.78 Hz for a superficial gas velocity of 0.86 m s$^{-1}$. Although Rowe’s correlation yields higher values for the particle exchange rate in comparison when using the Peclet number, it has to be emphasized that the Peclet number was experimentally derived (Mostoufi and Chaouki, 2001), while Rowe’s correlation is purely theoretical.

### 3.6.3. Validation results

The process variables, having the largest impact on the thermodynamic operation point were varied and the experimentally measured steady state outlet air temperatures were compared with the model-predicted outlet air temperatures. The process conditions along with the experimentally measured ($T_{a,\text{out,exp}}$) and the model-predicted values ($T_{a,\text{out,sim}}$) for the outlet air temperature during steady state coating regime are given in Table 3.2. The remaining fixed operational parameters along with the model parameters are listed in Table 3.1. The total simulated time does not reflect the true length of the fluidised bed coating process. To save calculation time, the simulation was terminated after a thermodynamic steady state was reached, which was approximately 900 s after initiating spraying. Again, prior to spraying, the heating of the bed was simulated until steady state was reached, which was after approximately 600 s of simulated time. Consequently, in total, 1500 s of combined heating and spraying were simulated during each run.

The correlation between the experimentally measured outlet air temperatures and the model-predicted values is presented in Figure 3.31. The linear regression coefficients and the sum of squared residuals ($SSR$), based on the outlet air temperatures, are shown in Table 3.3. The $SSR$ is a measure of the model’s deviation from the experimental values and is calculated as

$$SSR = \sum (T_{a,\text{out,exp}} - T_{a,\text{out,sim}})^2$$  \hspace{1cm} (3.54)

The values are compared to those of the model (Topsim, model II, also described in Section 2.2.2), described by Dewettinck et al. (1999). In this model, the reactor was considered a black-box taking neither coating deposition nor particle dispersion (mixing behaviour) into account.
Table 3.2. Process conditions and experimental vs. model-predicted values of outlet temperature.

<table>
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<tr>
<th>No.</th>
<th>$T_e$ (°C)</th>
<th>$\varphi_e$</th>
<th>$T_{a,in}$ (°C)</th>
<th>$M_{sol}$ (g min$^{-1}$)</th>
<th>$P_{at}$ (bar)</th>
<th>$G_{a,in}$ (kg s$^{-1}$)</th>
<th>$T_{a,exp}$ (°C)</th>
<th>$T_{a,sim}$ (°C)</th>
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<td>3.23</td>
<td>2.5</td>
<td>$1.25 \times 10^{2}$</td>
<td>40.3 ± 0.6</td>
<td>39.19 ± 0.05</td>
</tr>
<tr>
<td>27</td>
<td>20.4</td>
<td>0.43</td>
<td>50</td>
<td>3.20</td>
<td>2.5</td>
<td>$1.41 \times 10^{2}$</td>
<td>41.9 ± 0.6</td>
<td>40.00 ± 0.05</td>
</tr>
<tr>
<td>28</td>
<td>21.6</td>
<td>0.39</td>
<td>50</td>
<td>3.22</td>
<td>2.5</td>
<td>$1.56 \times 10^{2}$</td>
<td>43.1 ± 0.6</td>
<td>41.12 ± 0.05</td>
</tr>
</tbody>
</table>
**Figure 3.31.** Correlation between experimental and simulated outlet air temperature, using the new model (■) and the model Topsim-II (○) described by Dewettinck *et al.* (1999).

**Table 3.3.** Regression analysis between model-predicted and experimental outlet air temperature, including the sum of squared residuals and the 95%-confidence intervals.

<table>
<thead>
<tr>
<th></th>
<th>$R^2$</th>
<th>SSR</th>
<th>Slope, $a$</th>
<th>95% confidence interval of slope</th>
<th>Intercept, $b$</th>
<th>95% confidence interval of intercept</th>
</tr>
</thead>
<tbody>
<tr>
<td>Proposed model</td>
<td>0.9592</td>
<td>63.9</td>
<td>0.996</td>
<td>[0.913; 1.079]</td>
<td>0.433</td>
<td>[-3.145; 4.011]</td>
</tr>
<tr>
<td>Topsim model II</td>
<td>0.9786</td>
<td>119.5</td>
<td>0.954</td>
<td>[0.898; 1.011]</td>
<td>0.301</td>
<td>[-2.273; 2.874]</td>
</tr>
</tbody>
</table>

* Dewettinck *et al.*, 1999

From the values of the slope and intercept of the regression line and their 95%-confidence intervals, it can be seen that the predicted outlet air temperatures, produced by the newly built model approached very closely the experimentally measured values. In contrast to the model described by Dewettinck *et al.* (1999), the newly built model generally predicted lower values of the outlet air temperature (Figure 3.31). Also, the values predicted by the model tend to deviate less from the experimental ones, hence the lower SSR.
3.6.4. Conclusions

The model has proven to be reliable in terms of predicting the overall thermodynamic behaviour of the fluidised bed in coating processes. However, only the steady state outlet air thermodynamic properties were used to validate the model which could be considered insufficient as the model also predicts axial temperature and humidity profiles of both the gas and the solid (particles) phase. This kind of validation, based on measuring the distribution of prevalent thermodynamic variables inside the bed, will be introduced in Chapter 4.

When focussing on the model-predicted outlet air temperature and comparing its results against existing models (e.g. Topsim proposed by Dewettinck et al., 1999), the new model provides slightly better accuracy. However, the main benefit of the new model in the study at hand is not its improved accuracy, but its more detailed description of the fluidised bed coating process in terms of:

- Whereas current models (e.g. Topsim) are limited to the calculation of the steady-state operation point, the proposed model is capable of performing dynamic simulation and for example, it allows to study the behaviour of the coating process when sudden changes in the input variables (e.g. spraying rate) are applied.
- Whereas the Topsim model treats the fluidised bed as being perfectly mixed and being represented by a single set of variables (e.g. air temperature, humidity,…), the proposed model takes the one-dimensional (axial) distribution of the thermodynamic properties of both the gas (air) and solid (particles) phases into account.
- The proposed model integrates a multivariate population balance model to describe particle properties including thermodynamic variables and coating mass content distributions.

However, the added complexity of the model also implies some drawbacks. First of all, the model requires a larger amount of calibration data including particle mixing behaviour (expressed by the dispersion coefficients or exchange rates) and fluidised bed behaviour (including bed height, voidage fraction). Secondly, the time required to simulate a coating process using the proposed model is significantly longer (up to 12 hours for a coating process with a duration of 1 hour) compared to existing models, such as Topsim, which could be solved instantaneously using contemporary hardware.
3.7. Sensitivity analysis

3.7.1. Introduction

Now that the model has been validated, it can be used to assess the impact of the different input variables (process conditions, material properties, …) on the coating process dynamics. In a sensitivity analysis, the variation of a single variable in the set of input variables and its resulting impact on the model-predicted outputs is studied. The main interest in this sensitivity study went to the process variables that are commonly used as control variables in the fluidised bed coating process (e.g. inlet air temperature, $T_{a,in}$ or inlet air volumetric flow rate, $V_{a,in}$). Other variables and parameters such as reactor dimensions and, to a lesser extent, material properties are usually fixed in practical applications, and consequently they will be discussed only briefly in this sensitivity study.

3.7.2. Methods and calculations

3.7.2.1. Reference case simulation

In the sensitivity analysis, a reference case (i.e., a reference set of input variables) has to be defined. Again, the same set of input variables was used as in Section 3.5 for the analysis of the critical number of control volumes and the minimum sample size and is described in detail in Table 3.1. The determination or estimation of unknown variables such as the relative size of the coating region ($c/n$) and average particle exchange rate ($r_i$) have already been discussed in Section 3.6.2.2. The reference case simulation consisted of a coating process of inert glass spheres which was simulated from start-up to stationary conditions using reactor and bed dimensions of the Glatt GPCG-1 fluidised bed unit. The simulated process consisted of two distinct phases: in the heating phase, no coating solution was sprayed ($M_{sol} = 0 \text{ g min}^{-1}$). Next, after 500 seconds of simulated time, a step input was given for the spraying rate ($M_{sol} = 10 \text{ g min}^{-1}$).

3.7.2.2. Translation factor

To describe the sensitivity of the model, each parameter was varied individually and the translation factor, or also called normalised sensitivity coefficient (NSC) was calculated using the equation (He et al., 2000):

$$
\frac{1}{K_{\omega}} = \frac{\partial \Omega}{\Omega} \cdot \frac{\partial \omega}{\partial \ln \omega} = \frac{\partial \ln \Omega}{\partial \ln \omega}
$$

(3.55)

The translation factor describes the relative change of an output variable $\Omega$ as a result of a relative change, or perturbation, of the input variable $\omega$. A translation factor higher than 1 indicates an amplifying effect of the change in variable $\omega$; a value lower than 1 implies an...
attenuating effect of the change in variable \( \omega \). In practice, model sensitivity is usually described by the local sensitivity coefficient, which is the absolute change of \( \Omega \) as a result of the absolute change in \( \omega \), or \( \partial \Omega / \partial \omega \). However, as the different input variables have different units and ranges, the comparison of model sensitivity by means of \( \partial \Omega / \partial \omega \) (i.e. the so-called local sensitivity coefficient) is often less meaningful than using the translation factors or normalised sensitivity coefficients. The latter are dimensionless and allow for the direct comparison between each other, even though their counterpart local sensitivity coefficients may be orders of a magnitude apart (He et al., 2000).

Practically, the translation factor could be calculated, using the simulation results from two distinct simulations using varied inputs, in two different ways. The first method is the one-sided difference approach,

\[
\frac{1}{K_{\omega}} \approx \frac{\omega}{\Omega(\omega)} \cdot \frac{\Omega(\omega + \Delta \omega) - \Omega(\omega)}{\Delta \omega} \quad (3.56)
\]

Alternatively, the central difference approach may be used

\[
\frac{1}{K_{\omega}} \approx \frac{\omega}{\Omega(\omega)} \cdot \frac{\Omega(\omega + \Delta \omega) - \Omega(\omega - \Delta \omega)}{2\Delta \omega} \quad (3.57)
\]

It could be easily demonstrated that the central difference approach is equal to the average of two one-sided differences, using both negative (\( \Delta \omega < 0 \)) and positive (\( \Delta \omega > 0 \)) perturbations. Comparing the one-sided translation factors from both negative (\( \Delta \omega < 0 \)) and positive perturbations (\( \Delta \omega > 0 \)) when the perturbation is not infinitely small could potentially result in non-equal or asymmetric one-sided translation factors which indicate a non-linear relationship between \( \Omega \) and \( \omega \). In this chapter’s sensitivity analysis, the one-sided difference approach was used.

It is also important to note that the accuracy of this practical method is difficult to assess. The smaller the perturbation \( \Delta \omega \), the lower the truncation error resulting from the omission of higher-order terms in Taylor series, but the higher the loss-of-significance error resulting from subtracting two almost equal numbers (He et al., 2000).

### 3.7.2.3. Selection of the input variables

The main focus went to the process variables which are commonly used in process control. However, some material-related variables were also included in this study. Table 3.4 gives a complete overview of all input variables used in this sensitivity analysis. In all cases, the relative size of the perturbation, \( \Delta \omega / \omega \), is 10 %. It is important to note that all temperature variables are expressed in Kelvin rather than degrees Celsius for the calculation of the translation factors since the use of referenced units is meaningless in sensitivity studies.
Table 3.4. The different input variables used in the sensitivity analysis.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Symbol</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fluidisation air properties</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Linear fluidisation air velocity (inlet duct)*</td>
<td>$v_{a,\text{in}}$</td>
<td>m s$^{-1}$</td>
</tr>
<tr>
<td>Inlet air temperature</td>
<td>$T_{a,\text{in}}$</td>
<td>K</td>
</tr>
<tr>
<td>Liquid spraying</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Atomisation air pressure **</td>
<td>$P_{\text{at}}$</td>
<td>bar</td>
</tr>
<tr>
<td>Atomisation air relative humidity</td>
<td>$\Phi_{\text{at}}$</td>
<td>-</td>
</tr>
<tr>
<td>Atomisation air temperature</td>
<td>$T_{\text{at}}$</td>
<td>K</td>
</tr>
<tr>
<td>Spraying rate</td>
<td>$M_{\text{sol}}$</td>
<td>g min$^{-1}$</td>
</tr>
<tr>
<td>Spraying solution temperature</td>
<td>$T_{\text{sol}}$</td>
<td>K</td>
</tr>
<tr>
<td>Spraying solution dry matter content</td>
<td>$DM_{\text{sol}}$</td>
<td>kg kg$^{-1}$</td>
</tr>
<tr>
<td>Relative size of the coating volume</td>
<td>$c/n$</td>
<td>-</td>
</tr>
<tr>
<td>External (atmospheric) properties</td>
<td></td>
<td></td>
</tr>
<tr>
<td>External air temperature</td>
<td>$T_{e}$</td>
<td>K</td>
</tr>
<tr>
<td>External air relative humidity</td>
<td>$\Phi_{e}$</td>
<td>-</td>
</tr>
<tr>
<td>Bed material properties</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Batch size</td>
<td>$M_{\text{bed}}$</td>
<td>kg</td>
</tr>
<tr>
<td>Particle specific density</td>
<td>$p_{p}$</td>
<td>kg m$^{-3}$</td>
</tr>
<tr>
<td>Particle thermal conductivity</td>
<td>$\lambda_{p}$</td>
<td>W m$^{-1}$K$^{-1}$</td>
</tr>
<tr>
<td>Particle specific heat</td>
<td>$C_{p,p}$</td>
<td>J kg$^{-1}$K$^{-1}$</td>
</tr>
<tr>
<td>Particle diameter</td>
<td>$d_{p}$</td>
<td>µm</td>
</tr>
<tr>
<td>Particle exchange rate</td>
<td>$r_{i}$</td>
<td>Hz</td>
</tr>
</tbody>
</table>

Remarks

* The linear air velocity at the inlet duct corresponds to the air velocity as measured by the pitot probe in the Glatt GPCG-1 (see also Figure 3.30). To recalculate to volumetric air flow rate, the linear air flow rate has to be multiplied with the surface area of the cross-section through the inlet air duct, being $3.744 \times 10^{-3}$ m$^2$.

** The atomisation pressure is recalculated to the atomisation air mass flow rate using Eqs. (3.49) and (3.50).

3.7.2.4. Selection and definition of the output variables

The quantity of model generated data was quite large, thus a selection of output variables or derived output variables was made based on their relevance in real-life applications. The selection of output variables consisted of:
• Steady state outlet air temperature ($T_{a,\text{out}}$) and air humidity ($X_{a,\text{out}}$ or $\varphi_{a,\text{out}}$). In practical applications, these variables are easy to measure and consequently, are highly relevant in process control applications (Eichler, 1989).

• Steady state particle temperature ($T_p$) and particle moisture content ($W_p$) distributions. Particle moisture content distribution is particularly useful, as it can be correlated with the agglomeration tendency of the particles (Smith and Nienow, 1983).

• The coating mass distribution ($Y_p$) which gives an indication as to how uniformly the coating material is distributed among the particle population. Based on the coating mass distribution, a coating growth rate ($\delta_c$ in m s$^{-1}$) could be derived:

$$\delta_c = \frac{M_p}{\pi \rho_c d_p^2} \frac{dY_p}{dt} \quad \text{(3.58)}$$

In Eq. (3.58) is $\rho_c$ the density of the coating material. The growth rate is a theoretical value and expresses the change in thickness of the coating layer, assuming homogenous distribution of the coating over the (perfectly, non-porous) spherical particle.

• The thermodynamic efficiency of the process is characterised using three different variables: the steady state heat loss ($\Phi_{\text{loss}}$), the vaporisation efficiency ($\eta_{\text{vap}}$) and the thermal efficiency ($\eta_{\text{th}}$). The vaporisation efficiency expresses the degree to which the evaporative capacity of the supplied air is consumed in the drying process, or:

$$\eta_{\text{vap}} = \frac{X_{a,\text{out}} - X_{a,\text{in}}}{X_{a,\text{sat}} - X_{a,\text{in}}} \quad \text{(3.59)}$$

In Eq. (3.59) is $X_{a,\text{sat}}$ the absolute humidity of the outlet air at saturation (or $\varphi_{a,\text{out}} = 1$). It is clear that the higher the vaporisation efficiency, the more saturated the fluidisation air inside the bed and consequently, the higher the average particle moisture content ($W_p$) and the lower the drying rate $r_D$, thus increasing the risk of agglomeration. Therefore, fluidised bed coating processes never use the full evaporative capacity.

• Finally, the thermal efficiency or $\eta_{\text{th}}$ compares the energy used for evaporation to the total thermal energy supplied to the bed:

$$\eta_{\text{th}} = \frac{Q_{\text{vap}}}{G_{a,\text{in}} C_{p,a,\text{in}} (T_{a,\text{in}} - T_{a,\text{out}}) + M_{\text{sol}} C_{p,\text{sol}} (T_{\text{sol}} - T_{a,\text{out}}) + G_{a,\text{at}} C_{p,\text{at}} (T_{a,\text{at}} - T_{a,\text{out}})} \quad \text{(3.60)}$$

In Eq. (3.60), the outlet air temperature is used as reference. $Q_{\text{vap}}$ is the latent heat of vaporisation and is calculated according to Eq. (3.12). In case of a discrete model using $N_{\text{sim}}$ particles, the $Q_{\text{vap}}$ variable is calculated as
\[ Q_{\text{vap}} = 10^3 \frac{N_{\text{bed}}}{N_{\text{sim}}} \sum_{k=1}^{N_{\text{p}}} M_p r_{D,k} \left( 2.5 \times 10^3 - 2.364 T_{a,E(k)} \right) \]  

(3.61)

3.7.3. Results and discussion

3.7.3.1. Reference case simulation

The simulated air temperature and air relative humidity evolutions during the first 1500 seconds – the time after which steady state condition is reached during the coating (or spraying) stage – of a reference coating process are graphically presented in Figure 3.32. Air temperature and air relative humidity profiles along the reactor \( z \)-axis – being the bed’s central axis normal to the air distributor – are shown in Figure 3.33.

![Figure 3.32](image)

**Figure 3.32.** Simulated air temperature and air relative humidity profiles of the lowest positioned non-coating control volume, \( S_1 \) (\( T_a : \bullet, \varphi_a : \infty \)) and the highest positioned non-coating control volume, \( S_2 \) (\( T_a : \triangle, \varphi_a : \triangle \)) and the average of the coating control volumes, \( S_2 \) to \( S_4 \) (\( T_a : \bullet, \varphi : \infty \)). Spraying was initiated after \( t = 500 \) s.

It can be seen in Figure 3.32 that the loss of moisture in the coating solution sprayed onto the particle surface was very fast in such a way that only significantly higher air moisture contents are predicted in the volume of the bed where coating takes place (\( S_2 \) to \( S_4 \)). Due to evaporative cooling in the coating region and due to the release of compressed air at ambient temperature through the binary nozzle, air temperature was significantly lower in the coating control volumes (\( S_2 \) to \( S_4 \)) than the rest of the reactor. Also, a larger temperature difference
between the particles and the process air was created in the lower three control volumes situated closest to the air distributor ($S_1$ to $S_3$). Compared to the relative volume of $S_1$, $S_2$ and $S_3$ only being 12.5% of the bed, the particle-fluidum heat transfer occurring in $S_1$, $S_2$ and $S_3$ accounted for 48.7% of all heat transferred between the particles and the fluidising air. This temperature difference in the lower positioned control volumes could be explained by the fact that fluidised particles mainly absorb heat in the region closest to the air distributor; once the particles have migrated towards the upper bed regions, energy is transferred from the particles to the air. The simulated heat transfer from the fluidised bed towards the environment along with the average wall and bed air temperatures is presented in Figure 3.34. The average bed air ($T_a$) and average wall temperature ($T_w$) in Figure 3.34 were calculated as,

$$T_a = \frac{\sum_{i=1}^{n} M_{a,i} T_{a,i}}{\sum_{i=1}^{n} M_{a,i}}$$

(3.62)

$$T_w = \frac{\sum_{i=1}^{n} h_{S,i} d_{S,i} T_{a,i}}{\sum_{i=1}^{n} h_{S,i} d_{S,i}}$$

(3.63)

**Figure 3.33.** Simulated air temperature ($T_{a,i}$: ■), air relative humidity ($\phi_{a,i}$: ○) and average particle temperature ($T_{p,i}$: ▲) profiles along the reactor $z$-axis during steady-state coating.

The results in Figure 3.34 clearly show relatively high heat transfers from the bed towards the vertical reactor wall. Considering the low temperatures at which these beds operate and the low density of the fluidised bed, local heat transfer coefficients up to 134 W m$^{-2}$K$^{-1}$ can be
attained during steady spraying regime. However, the overall heat loss coefficient was limited by the combined convective and radiative heat loss from the wall towards the environment, of which the heat transfer coefficients did not surpass 6.5 W m$^{-2}$K$^{-1}$.

Finally, the model was also capable of predicting the evolution of the coating mass distribution during fluidised bed coating. Figure 3.35 presents the evolution of the coating mass distribution, expressed as unit mass of coating material per unit mass of core material, while Figure 3.36 shows the coating mass distributions, in detail, taken at different stages during the simulation.

**Figure 3.34.** Predicted evolution of the average fluidised bed air temperature (–––) and average reactor wall temperature (–●–) and total heat transfers from the fluidised bed towards the wall (–○–) and from the wall towards the environment (–○–). Spraying was initiated after $t = 500$s.
Figure 3.35. Predicted evolution of the coating mass distribution ($Y_p$), expressed as kg coating per kg core material.

Due to the constant spraying rate and under the assumption that neither spray drying nor attrition occurs, the relationship between average coating mass and process time is linear. Initially, the simulated coating mass distribution was asymmetric, but evolved to a normal...
distribution. A linear relationship \( R^2 = 0.9995 \) was found between the predicted coating mass standard deviation and the square root of the process time. This linear relationship could be easily proven considering that the coating mass each particle receives is directly proportional to the time spent in the coating control volumes throughout the process (Nakamura et al., 1998). Consequently, the probability for a particle to spend \( I_c \) cycles (or timesteps) in the coating control volumes during \( I \) cycles, assuming the particle mixing behaviour in the bed is unaffected by the addition of the relatively small mass of coating material, is given by the binomial distribution:

\[
P(I_c | I) = \binom{I}{I_c} \left( \frac{c}{n} \right)^{I_c} \left( \frac{n-c}{n} \right)^{I-I_c}
\]

The binomial distribution in Eq. (3.64) has a standard deviation which is proportional to the square root of the number of cycles, \( I \), which in turn is linearly proportional to the process time:

\[
s = \sqrt{\frac{I c(n-c)}{n^2}} \tag{3.65}
\]

However, the simplification of the particle behaviour, expressed as the binomial distribution in Eq. (3.64), assumes that particle exchange is not limiting. More specifically, the particles are assumed to be perfectly mixed in the whole of the coating control volumes \( c \) and in the whole of the non-coating control volumes \( n - c \). If the limitative effect of particle exchange is taken into account in calculating the standard deviation on the coating mass distribution, Eq. (3.65) results in:

\[
s = \frac{1}{r} \sqrt{\frac{I c(n-c)}{n^2}} \tag{3.66}
\]

Eq. (3.66) proves that, using a simple mathematical model, the coating mass distribution width is directly proportional to the relative size of the coating volume \( c/n \) and inversely proportional to the particle exchange rate \( r_i \).

### 3.7.3.2. Overview of the calculated translation factors

The results of the translation factor calculation are summarised in Table 3.5 for the exhaust air properties and the thermodynamic efficiencies as output variables and in Table 3.6 for the particle-related model-predicted output variables. The calculated translation factors from these tables have been represented graphically in Figures 3.37a to 3.37l. In these graphs, both the single-sided translation factors for a negative input variable variation \( \Delta \omega < 0 \) as well as for a positive variation \( \Delta \omega > 0 \) are shown. In some cases, there was a significant asymmetry.
between the translation factor for $\Delta \omega < 0$ and for $\Delta \omega > 0$, indicating a non-linear effect of the input variable on the model-predicted output.

**Table 3.5.** Translation factors (central difference, calculated as the average from both single-sided differences) for the exhaust air properties and the thermodynamic efficiency of the process.

<table>
<thead>
<tr>
<th>Input variable ($\omega$)</th>
<th>$\bar{T}_{\text{a,out}}$</th>
<th>$\bar{\theta}_{\text{a,out}}$</th>
<th>$\bar{X}_{\text{a,out}}$</th>
<th>$\bar{n}_{\text{cap}}$</th>
<th>$\bar{n}_{\text{th}}$</th>
<th>$\Phi_{\text{loss}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P_{\text{at}}$ (bar)</td>
<td>$-2.09 \times 10^{-3}$</td>
<td>$1.95 \times 10^{-2}$</td>
<td>$-4.70 \times 10^{-2}$</td>
<td>$-5.66 \times 10^{-3}$</td>
<td>$-1.91 \times 10^{-2}$</td>
<td>$-2.97 \times 10^{-2}$</td>
</tr>
<tr>
<td>$r_{\text{f}}$ (Hz)</td>
<td>$9.59 \times 10^{-3}$</td>
<td>$-1.78 \times 10^{-3}$</td>
<td>$9.12 \times 10^{-4}$</td>
<td>$-2.11 \times 10^{-3}$</td>
<td>$-5.54 \times 10^{-5}$</td>
<td>$-2.36 \times 10^{-4}$</td>
</tr>
<tr>
<td>$\dot{M}_{\text{sol}}$ (g min$^{-1}$)</td>
<td>$-4.08 \times 10^{-2}$</td>
<td>$1.82$</td>
<td>$5.49 \times 10^{-1}$</td>
<td>$2.70$</td>
<td>$1.01$</td>
<td>$-7.77 \times 10^{-1}$</td>
</tr>
<tr>
<td>$T_{\text{a}}$ (K)</td>
<td>$5.71 \times 10^{-2}$</td>
<td>$-9.62 \times 10^{-1}$</td>
<td>$8.49 \times 10^{-3}$</td>
<td>$-6.78 \times 10^{-4}$</td>
<td>$-5.72 \times 10^{-1}$</td>
<td>$8.37 \times 10^{-2}$</td>
</tr>
<tr>
<td>$T_{\text{a,in}}$ (K)</td>
<td>$4.52 \times 10^{-4}$</td>
<td>$-13.8$</td>
<td>$4.81 \times 10^{-1}$</td>
<td>$-1.75$</td>
<td>$-4.03$</td>
<td>$2.52$</td>
</tr>
<tr>
<td>$v_{\text{a,in}}$ (m s$^{-1}$)</td>
<td>$3.77 \times 10^{-2}$</td>
<td>$-1.64$</td>
<td>$-4.72 \times 10^{-3}$</td>
<td>$-2.42$</td>
<td>$-1.01$</td>
<td>$1.82$</td>
</tr>
</tbody>
</table>

$\Phi_{\text{loss}}$
Table 3.6. Translation factors (central difference, calculated as the average from both single-sided differences) for the particle-related output variables.

<table>
<thead>
<tr>
<th>Output variable (Ω)</th>
<th>$\bar{T}_p$</th>
<th>$s(T_p)$</th>
<th>$\bar{W}_p$</th>
<th>$s(W_p)$</th>
<th>$\bar{\delta}_c$</th>
<th>$s(\delta_c)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P_{at}$ (bar)</td>
<td>-1.40 × 10^{-3}</td>
<td>3.36 × 10^{-2}</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>-1.07 × 10^{-2}</td>
</tr>
<tr>
<td>$r_i$ (Hz)</td>
<td>-7.61 × 10^{-3}</td>
<td>-7.29 × 10^{-1}</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>-5.84 × 10^{-1}</td>
</tr>
<tr>
<td>$\dot{M}_{sol}$ (g min^{-1})</td>
<td>-2.98 × 10^{-2}</td>
<td>8.06 × 10^{-1}</td>
<td>1.00</td>
<td>1.00</td>
<td>1.00</td>
<td>9.91 × 10^{-1}</td>
</tr>
<tr>
<td>$T_{at}$ (K)</td>
<td>3.76 × 10^{-2}</td>
<td>-1.03</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>-6.26 × 10^{-2}</td>
</tr>
<tr>
<td>$T_{a,in}$ (K)</td>
<td>4.75 × 10^{-3}</td>
<td>1.69</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>5.80 × 10^{-2}</td>
</tr>
<tr>
<td>$v_{a,in}$ (m s^{-1})</td>
<td>3.84 × 10^{-2}</td>
<td>-3.30 × 10^{-1}</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>-3.35 × 10^{-2}</td>
</tr>
<tr>
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Figure 3.37. Translation factors describing the predicted effects of variation on (a) the outlet air temperature, $T_{a,\text{out}}$, (b) the outlet air relative humidity, $\bar{\varphi}_{a,\text{out}}$, and (c) the outlet air absolute humidity, $X_{a,\text{out}}$ (single-sided difference, $\Delta\omega < 0$, $\Delta\omega > 0$).
Combined thermodynamic and population balance of the fluidised bed coating process

(d) Output variable: Vaporisation efficiency

(e) Output variable: Thermal efficiency

(f) Output variable: Heat losses (W)

Figure 3.37. Translation factors describing the predicted effects of variation on (d) the vaporisation efficiency, $\eta_{vap}$, (e) the thermal efficiency, $\eta_{th}$ and (f) the steady state heat loss, $\Phi_{loss}$ (single-sided difference, $\Delta \omega < 0$, $\Delta \omega > 0$).

Modelling heat and mass transfer in fluidised bed coating processes
Figure 3.37. Translation factors describing the predicted effects of variation on (g) the average steady state particle temperature, $\bar{T}_p$, (h) its standard deviation, $s(T_p)$ and the average steady state particle moisture content, $\bar{W}_p$ (i) (single-sided difference, $\Delta \omega < 0$, $\Delta \omega > 0$).
Figure 3.37. Translation factors describing the predicted effects of variation on (j) the standard deviation of the particle moisture content during steady state, \( s(W_p) \), (k) the average coating growth rate \( \delta_c \), and (l) its standard deviation \( s(\delta_c) \) (single-sided difference, \( \Delta \omega < 0 \), \( \Delta \omega > 0 \)).
In the following sections, the effect of each input variable is discussed:

### 3.7.3.3. Inlet air temperature

While the inlet air temperature ($T_{a,in}$) had the most pronounced effect on the outlet air temperature (Figure 3.37a), it had virtually no effect on the absolute outlet air humidity (Figure 3.37c). This effect can be explained by the drying kinetics of the sprayed liquid on the particle surface. Due to the small diameter of the fluidised particles and the low operational air relative humidity, mass (water) transfer is not limiting. As long as the system is not operating near saturation conditions, inlet air temperature has no significant effect on absolute air humidity distribution inside the bed. Also, the particle moisture content is unaffected by the inlet air temperature because the drying rate is not limiting in the range of conditions examined.

To better understand the effect of inlet air temperature on the bed thermodynamics, a series of simulations was performed with an inlet air temperature ranging between $T_{a,in} = 50 \, ^\circ C$ and $T_{a,in} = 90 \, ^\circ C$. All other variables were left unaltered. Figures 3.38a to 3.38e illustrate the different axial temperature and humidity profiles for both the gas and solid phase. As can be seen from Figures 3.38a and 3.38c, both the particle and air temperature profiles increase linearly with $T_{a,in}$, while maintaining a constant temperature gradient. Figure 3.38e shows that only the particles in the coating volume (upper $c$ control volumes) have been wetted and the moisture content $W_p$ is independent of $T_{a,i}$. This, however, is only valid when the process does not operate near its saturation point (as will be shown later). From Figure 3.38d it can be seen that the standard deviation on the particle temperature, $s(T_p)$, also increases linearly with inlet air temperature. The highest $s(T_p)$ is predicted in the non-coating control volumes positioned just below the coating control volumes. This phenomenon could be explained by the return of particles from the upper $c$ coating control volumes, which have been cooled evaporatively, mixing with the particles coming from lower positioned control volumes, resulting in a wide local particle temperature distribution.

Due to the high sensitivity of the thermodynamic properties of the fluidised bed and of the energetic efficiency (both thermal and vaporisation efficiency, see Figure 3.37d to 3.37f), the inlet air temperature should be the variable of choice in process control applications. However, the rather slow responsiveness of the fluidised bed to changes in inlet air temperature, compared to other process variables, more specifically the spraying rate, renders the air temperature less suited as control variable.
3.7.3.4. Inlet air flow rate

From Figure 3.37 it can be concluded that inlet air flow rate, $G_{a,in}$, has the same effect, albeit to a lesser extent, as the inlet air temperature. The exception is the particle temperature distribution, where increasing the inlet air flow rate results in a narrower particle temperature
distribution, whereas an increased inlet air temperature yields a widened particle temperature distribution. The explanation for this behaviour is that the inlet air flow rate – as opposed to inlet air temperature – is also related to the fluidisation behaviour of the bed and to the particle exchange rate \( r \). Increasing the inlet air flow rate results in more rapid particle exchange, resulting in narrowing the particle temperature and coating mass distribution, as demonstrated in Section 3.7.3.1. The effect of the inlet air flow rate on the bed height, calculated using the equations from Section 1.4.4.5, and the effect on the particle exchange rate, calculated using the Peclet number in Eq. (3.52) is illustrated in Figure 3.39.

![Figure 3.39](image)

**Figure 3.39.** The model-predicted particle exchange rate, \( r \) (○) and bed height, \( h_{\text{bed}} \) (■) as a function of the volumetric air flow rate, \( \dot{V}_{a,\text{in}} \).

More details concerning the axial temperature and humidity profiles for the gas and solid phase are given in Figure 3.40 for a volumetric air flow rate ranging between \( \dot{V}_{a,\text{in}} = 7.5 \times 10^{-3} \) and \( \dot{V}_{a,\text{in}} = 2.2 \times 10^{-2} \text{ m}^3\text{s}^{-1} \), which corresponds to a mass flow rate between \( G_{a,\text{in}} = 8.9 \times 10^{-3} \text{ kg s}^{-1} \) and \( G_{a,\text{in}} = 2.7 \times 10^{-2} \text{ kg s}^{-1} \) (This range also corresponds to Pitot-tube readings between \( v_{a,\text{in}} = 2.0 \text{ m s}^{-1} \) and \( v_{a,\text{in}} = 6.0 \text{ m s}^{-1} \) on the Glatt GPCG-1). Because of the varying bed height, the axial profiles were expressed by means of a dimensionless height, \( h_i/h_{\text{bed}} \). The lowest two values for the inlet air flow rate (\( \dot{V}_{a,\text{in}} = 7.5 \times 10^{-3} \) and \( \dot{V}_{a,\text{in}} = 9.4 \times 10^{-3} \text{ m}^3\text{s}^{-1} \)) resulted in moisture saturation of the bed’s air (Figure 3.40b). Saturation is also accompanied with a narrowing of the particle temperature distribution, as seen in Figure 3.40d and moisture accumulation on the particles (Figure 3.40e).
Figure 3.40. Model-predicted axial thermodynamic profiles during steady state conditions resulting from varying inlet air flow rate: (a) air temperature profile, $\bar{T}_{a,i}$, (b) air relative humidity profile, $\bar{\phi}_{a,i}$, (c) average particle temperature profile, $\bar{T}_p$, (d) standard deviation of particle temperature, $s(T_p)$ and (e) average particle moisture content, $\bar{W}_p$ in mg kg$^{-1}$.

3.7.3.5. Atomisation air properties

When comparing the three variables characterising the atomisation air (pressure, $P_{at}$, relative air humidity, $\phi_{at}$, and air temperature, $T_{at}$), it appears that only the temperature of the atomisation air had a pronounced effect on the bed thermodynamics. However, it was
expected that the mixing of the process air with the colder ($T_{at} = 20 \, ^\circ C$) and drier ($\phi_{at} = 0.3$) atomisation air in the upper control volumes at a mass ratio of about 9:4 should largely alter the bed’s thermodynamic operation point, including the bed’s air humidity. Comparing the translation factors for output variables with different units and ranges proved to be problematic.

To clarify the impact of the atomisation air, three series of simulations were performed: In the first series $P_{at}$ was varied between 0.5 and 5.0 bar, then, $\phi_{at}$ was varied between 0 and 1, and finally, the $T_{at}$ variable was varied between 5 and 50 $^\circ C$. Results are presented in Figure 3.41. The effect on the particle moisture content was found to be negligible and was therefore not included in Figure 3.41. From these figures it is obvious that expressing $T_{at}$ in Kelvin results in a very narrow interval for $\Delta \omega/\omega$ in Eq. (3.55), thus artificially increasing the translation factor. Also, the effect of altering the atomisation air pressure (or atomisation air flow rate $G_{at}$ using the relation in Eqs. (3.49) and (3.50)) is clearly in contradiction to its negligible translation factor (see Tables 3.5 and 3.6). It can be noticed from Figures 3.41a and 3.41c that the atomisation air pressure had a significant decreasing effect on both the outlet air temperature and the average particle temperature, while the particle temperature distribution was widened as can be seen in Figure 3.41d.

**Figure 3.41.** Model-predicted outlet air temperature, $\bar{T}_{a,\text{out}}$ (a), outlet air relative humidity, $\bar{\phi}_{\text{a,\text{out}}}$ (b), average particle temperature, $\bar{T}_{p}$ (c) and standard deviation on particle temperature, $s(T_{p})$ (d) when varying the atomisation air pressure (■), the atomisation air temperature (○) and the atomisation air relative humidity (■).
To conclude, the atomisation air pressure is a non-negligible variable affecting the bed thermodynamics, especially in small reactor configurations which have a much smaller difference between atomisation air flow rate and fluidisation air flow rate. Furthermore, in real coating applications, atomisation air temperature and air relative humidity are fixed parameters as opposed to atomisation air pressure which is varied in accordance to the desired droplet size and the rheological properties of the coating solution.

3.7.3.6. The spraying rate and coating solution properties

Higher spraying rates resulted in lower outlet air temperatures and higher outlet air humidities, and consequently in higher thermal and vaporisation efficiencies, and reduced heat losses. The spraying rate variable, $\dot{M}_{\text{sol}}$, affected every modelled output variable: increasing the spraying rate also resulted in widened particle temperature and moisture content distributions. However, this effect was found to be inverted when the bed is operating near its saturation point as can be observed in Figure 3.42d. The axial temperature and humidity profiles for a spraying rate varying between $\dot{M}_{\text{sol}} = 1$ and $\dot{M}_{\text{sol}} = 21 \text{ g min}^{-1}$ are illustrated in Figure 3.42. The modelled fluidised bed reached partial moisture saturation at 16 g min$^{-1}$ and became fully saturated above 18 g min$^{-1}$ (Figures 3.42b and e).
Figure 3.42. Model-predicted axial thermodynamic profiles during steady state conditions resulting from varying the spraying rate: (a) air temperature profile, $T_{a,i}$, (b) air relative humidity profile, $\varphi_{a,i}$, (c) average particle temperature profile, $\bar{T}_p$, (d) standard deviation of particle temperature, $s(T_p)$ and (e) average particle moisture content, $\bar{W}_p$, in mg kg$^{-1}$.

Also noteworthy is what happens during the transition of the bed towards its point of saturation when the spraying rate $\dot{M}_{\text{sol}}$ is increased. Figure 3.43 represents the simulated air relative humidity for different control volumes in case of spraying at $\dot{M}_{\text{sol}} = 18$ g min$^{-1}$. The arrows in Figure 3.43 indicate inflection points on the relative humidity profiles. The physical explanation behind the occurrence of these inflection points is that wetted particles, no longer being dried due to saturation, migrate towards lower positioned control volumes and start transferring mass (water) until the process air in that control volume is also depleted in its evaporative capacity. In Figure 3.43, this downward moving front of overwetted particles is clearly visible by the indicated arrows.

Changing the dry matter content of the coating solution, $DM_{\text{sol}}$, showed an opposite and less pronounced effect compared to the spraying rate, as the dry matter content is related to the amount of water introduced into the fluidised bed. Details concerning the effect of dry matter content are given in Figure 3.44, where the results are presented when the dry matter content was varied between 0.01 and 0.25. It is important to note that the model makes no link whatsoever between the coating solution’s dry matter content and its effect on coating liquid distribution throughout the bed.
Figure 3.43. Simulated air relative humidity profiles, $\theta_{\text{air},i}$, of the lowest positioned non-coating control volume, $S_1$ (•), the middle positioned non-coating control volume $S_{11}$ (○), the highest positioned non-coating control volume, $S_{21}$ (▲) and the average of the coating control volumes, $S_{22}$ to $S_{24}$ (○). Spraying was initiated after $t = 500$ s with $M_{\text{sol}} = 18$ g min$^{-1}$. Inflection points are indicated by the arrows.

(a) Simulated air relative humidity profiles. (b) Simulated air temperature and dry matter content of the coating solution. (c) Simulated average particle temperature and dry matter content of the coating solution. (d) Simulated standard deviation on particle temperature and dry matter content of the coating solution.
Increasing the dry matter content increases coating solution viscosity, which in turn increases the droplet size and as a result, the volume over which the coating liquid is distributed (or coating volume, \(c/n\)) is increased. This effect however, was not simulated in the model. To overcome this shortcoming in the proposed model, a method will be presented in the next chapter for modelling a more accurate liquid distribution throughout the bed during the coating process.

Finally, the effect of the the coating solution temperature, \(T_{\text{sol}}\), (see Tables 3.5 and 3.6) on the bed thermodynamics is minimal as the amount of sensible heat carried by the coating solution is negligible compared to the sensible heat of the supplied air, or the latent heat that is absorbed during evaporation of the coating solution’s solvent.

3.7.3.7. The relative size of the coating volume

Because of its relevance in the coating process, the relative size of the coating volume or \(c/n\) is being dealt with separately.

Eq. (3.66) predicts that the fraction of coating volumes (\(c/n\)) influences the coating mass distribution width or the coating growth rate, \(\delta_c\), as defined in Eq. (3.58). In order to study the effect of the fraction of the coating volumes, a series of simulations was performed using the reference conditions of Table 3.1, but with varying relative coating volume (\(c/n\)). In Figure 3.45, the effect on the thermodynamic behaviour is presented. The relative size of the coating volume did not have any significant influence on the outlet air thermodynamic properties as the global heat and mass balances of the bed do not change, but a pronounced effect on the...
bed’s temperature and humidity gradients was observed. More specifically both the air and particle temperature gradients across the bed decrease when the relative size of the coating volume (or c/n) is increased. Also, the average particle temperature decreased with increasing c/n as seen in Figures 3.45c and 3.45d.

Figure 3.46 shows the effect of the fraction of the coating volumes (c/n) on the cumulative coating mass distribution after \( t = 1500 \) s. Increasing the \( c/n \) – ratio resulted in a more narrow coating distribution, which would theoretically become infinitely narrow when the fraction of coating volumes reached 100%. Similar conclusions regarding the effect of the coating region size on the coating mass distribution were drawn by Maronga and Wnukowski (1997) and by Nakamura et al. (1998). Although increasing the coating volume’s relative size (c/n) would be beneficial to the coating mass (or thickness) distribution in micro-encapsulation applications, in reality, the larger fraction of wetted particles is likely to increase agglomeration phenomena due to liquid bridge formation (Smith and Nienow, 1983) – an effect which was not taken into consideration in the model. Consequently, the relative size of the coating volume is to be restricted to allow sufficient drying on the wetted particles before they re-enter the coating region; a value of \( c/n = 10\% \) is recommended (Maronga and Wnukowski, 1997).
Figure 3.45. Model-predicted axial thermodynamic profiles during steady state conditions resulting from varying the relative size of the coating volume ($c/n$): (a) air temperature profile, $\overline{T_{ai}}$, (b) air relative humidity profile, $\overline{\varphi_{ai}}$, (c) average particle temperature profile, $\overline{T_p}$, (d) standard deviation of particle temperature, $s(T_p)$ and (e) average particle moisture content, $\overline{W_p}$ in mg kg$^{-1}$ (for clarity, the direction of the axes has been reversed in plots b and d).

Figure 3.46. Simulated effect of the relative size of the coating volume ($c/n$) on the cumulative coating mass distributions ($Y_p$).

3.7.3.8. Particle exchange rate

Eq. (3.66) also predicts that the particle exchange rate ($r_i$), which is related to the axial dispersion coefficient ($D_{p,ax}$), also influences the coating mass distribution width. To study the effect of the particle exchange rate, a series of simulations were performed using the reference conditions from Table 3.1, but with particle exchange rates ranging between $r_i = 0.5$ Hz and
The model-predicted effect of the particle exchange rate on the bed’s axial temperature and humidity profiles is presented in Figures 3.47a and 3.47e, while the effect on the coating mass distributions after $t = 1500$ seconds is given in Figure 3.48.

The same conclusions as for increasing the $c/n$-ratio can be drawn: The particle exchange rate had no significant effect on the outlet air thermodynamic properties as opposed to the bed’s temperature and humidity gradients. Both the particle temperature distribution and the coating mass distribution narrow when the particle exchange rate is increased. Consequently, increasing the particle exchange rate could be beneficial in controlling the coating mass distribution and coating uniformity. In this respect, the Wurster or bottom-spray coating process is a typical example of a coating process where improved and less random particle circulation is achieved through the use of the central draft tube and the air distributor providing controlled airflow.

Particle exchange rates are mainly determined by the excess gas velocity $v_a - v_{mf}$ (Stein et al., 2000; Mostoufi and Chaouki, 2001) and, to a lesser extent, by the fluidised bed diameter and geometry (Grasa and Abanades, 2002). However, increasing the particle mixing by adjusting the inlet air flow rate also supplies a larger evaporative capacity to the bed, resulting in reduced coating efficiencies due to increased spray drying losses (Dewettinck and Huyghebaert, 1998), an effect which will be included in the model in Chapter 4.
**Figure 3.47.** Model-predicted axial thermodynamic profiles during steady state conditions resulting from varying the particle exchange rate ($r_i$): (a) air temperature profile, $\bar{T}_{\text{a,i}}$, (b) air relative humidity profile, $\bar{\phi}_{\text{a,i}}$, (c) average particle temperature profile, $\bar{T}_p$, (d) standard deviation of particle temperature, $s(T_p)$ and (e) average particle moisture content, $\bar{W}_p$ in mg kg$^{-1}$.

**Figure 3.48.** Simulated effect of the particle exchange rate ($r_i$) on the cumulative coating mass distributions ($Y_p$).

### 3.7.3.9. External air properties

From Figures 3.37b and 3.37c, it appears that the external air temperature, $T_e$, has a significant effect on the outlet air humidity. The explanation for the large translation factor $1/K_{r_e}$ is twofold: first, expressing the translation factors in Kelvin results in very small values for $\Delta \phi/\phi$ in Eq. (3.55). Second, as the inlet air humidity is defined as relative air humidity, $\phi_e$, rather than absolute air humidity, $X_e$. In the model, the relative air humidity was kept constant.
when varying the external temperature in the sensitivity analysis and consequently, the absolute air humidity, $X_e$, varied along with $T_e$. Besides affecting the outlet air thermodynamic properties, the external temperature also had a pronounced influence on the bed heat losses. However, the heat loss from the bed to the environment is negligible compared to the heat supplied by the fluidisation air. Figure 3.34 gives a typical value for the model-predicted heat loss in the fluidised bed coating process. On average, the heat losses represent approximately 1 to 2% of the heat supplied by the fluidisation air.

### 3.7.3.10. Particle diameter

The effect of the particle diameter, $d_p$, on the bed thermodynamics was studied. However it should be noted that the particle diameter, contrary to all other modelled input variables, also affects the error on the model-predicted output variables (cfr. Section 3.5). The explanation behind this effect is that $N_{\text{bed}}$, being the number of particles in the (physical) bed, increases with smaller values of the particle diameter when the mass of the bed, $M_{pp,\text{bed}}$, is kept constant. Thus, the scaling factor – $N_{\text{bed}}/N_{\text{sim}}$ in Eqs. (3.45) to (3.48) – which links the cumulative heat and mass transfer of the simulated population ($N_{\text{sim}}$) to the control volume’s air heat and mass balances, is affected when varying the particle diameter. Normally, for each different particle diameter, the number of simulated particles ($N_{\text{sim}}$) should be reassessed to maintain a constant error on the model-predicted output variables. Figure 3.49 illustrates what happens to the error (standard deviation) on the model-predicted outlet air temperature and humidity when the particle diameter is changed when using a constant number of simulated particles, $N_{\text{sim}}$. Other variables that were used in Section 3.5 for error quantification can not be used any longer, as there is an obvious physical relationship between the particle diameter and, for instance, the particle temperature distribution or the coating mass distribution. From Figure 3.49 it can be concluded that, although the model produces a larger error when increasing the particle diameter, it is still within an acceptable range and re-evaluation of the critical $N_{\text{sim}}$ for each value of $d_p$ is not considered necessary.

Another aspect that needs to be taken into consideration when varying the particle diameter is its effect on the fluidisation behaviour. Therefore, the particle exchange rate, $r_i$, and the bed height, $h_{\text{bed}}$, need to be recalculated for each value of the particle diameter. The effect of the particle diameter on the bed height, calculated using the equations in Section 1.4.4., and on the particle exchange rate, calculated using the Peclet number in Eq. (3.52) is illustrated in Figure 3.50. According to Eq. (3.52), an inverse correlation between the axial dispersion coefficient and the particle diameter exists.
Figure 3.49. The simulated effect of the particle size on the standard deviation (or error) of the outlet air temperature, $s(T_{a,\text{out}})$ (■), and the outlet air absolute humidity, $s(X_{a,\text{out}})$ (○) at steady state.

Figure 3.50. The model-predicted particle exchange rate, $r_i$ (○) and bed height, $h_{\text{bed}}$ (■) as a function of the particle diameter, $d_p$. 
Figure 3.51. Model-predicted axial thermodynamic profiles during steady state conditions resulting from varying the particle diameter: (a) air temperature profile, $T_{\text{a},i}$, (b) air relative humidity profile, $\varphi_{\text{a},i}$, (c) average particle temperature profile, $\bar{T}_p$, (d) standard deviation of particle temperature, $s(T_p)$, and (e) average particle moisture content, $\bar{W}_p$ in mg kg$^{-1}$.

From Figure 3.37 and Table 3.6, it can be deduced that the particle diameter has a significant effect on the width of the particle temperature distribution at steady state — and to a lesser extent, on the average particle temperature — and on the particle moisture distribution. When the particle diameter is increased, the standard deviation on the particle temperature increases as can be observed in Figure 3.51, where the model-predicted axial temperature and humidity...
profiles for both the gas and solid phase have been plotted for a particle diameter ranging between 100 and 1000 µm.

Again, as the bed height depends on particle diameter, the axial position is expressed in dimensionless $h/h_{\text{bed}}$ coordinates. The slight decrease in average particle temperature occurring when lowering the particle diameter, could be explained by the fact that the heat loss mechanism between the fluidised bed and the inner reactor wall, as explained in Section 3.3.2, depends on the particle diameter. The effect on the particle temperature distribution width is the result of the increased thermal inertia of the larger particles. The larger the particle, the longer it takes to equilibrate the particle temperature with the control volume’s air temperature. As a result, higher temperature differences between the air and particles occur at larger particle diameters, as illustrated in Figure 3.52.

![Model-predicted axial thermodynamic profiles for $T_{a,i} - T_p$ during steady state conditions resulting from varying the particle diameter.](image)

**Figure 3.52.** Model-predicted axial thermodynamic profiles for $T_{a,i} - T_p$ during steady state conditions resulting from varying the particle diameter.

### 3.7.3.11. Batch size

Just as in the case where the effect of particle diameter was discussed, the same considerations are to be taken into account: first, the scaling factor $N_{\text{bed}}/N_{\text{sim}}$ is changed when the batch size ($M_{\text{bed}}$) is varied and consequently, the error on the model-predicted output variables is also affected. Second, the variables characterising fluidisation, including bed height and particle exchange rate, have to be recalculated for each change in the bed size variable ($M_{\text{bed}}$).

Considering the error on the model-predicted output variables, the effect of batch size – which ranged between 0.5 and 3.0 kg – on the model-predicted outlet air temperature, $T_{a,\text{out}}$, and on the outlet air absolute humidity, $X_{a,\text{out}}$, is illustrated in Figure 3.53. The error on the outlet air absolute humidity was negligible. On the other hand, the error on the outlet air temperature continued to rise to 0.8 °C at $M_{\text{bed}} = 3.0$ kg. It was decided however, that the model generated
error on its output variables is still within an acceptable range to perform the sensitivity analysis without reassessing the critical sample size, \( N_{\text{bed}} \).

**Figure 3.53.** The simulated effect of the batch size on the standard deviation (or error) of the outlet air temperature, \( s(T_{a,\text{out}}) \) (■), and the outlet air absolute humidity, \( s(X_{a,\text{out}}) \) (○), during steady state.

Secondly, the influence of the sample size on the particle exchange rate, \( r_i \), and the bed height, \( h_{\text{bed}} \), needed to be recalculated prior to the sensitivity analysis. The bed height was calculated using the bed voidage, \( \varepsilon_{\text{bed}} \), calculated using the equations in Section 1.4.4.5. Particle exchange rate was calculated using the axial dispersion coefficient in Eq. (3.52). The results are presented in Figure 3.54.

**Figure 3.54.** The model-predicted particle exchange rate, \( r_i \) (○) and bed height, \( h_{\text{bed}} \) (■) as a function of the batch size, \( M_{\text{bed}} \).
Figure 3.55. Model-predicted axial thermodynamic profiles during steady state conditions resulting from varying the batch size ($M_{\text{bed}}$): (a) air temperature profile, $T_{\text{air}}$, (b) air relative humidity profile, $\phi_{\text{air}}$, (c) average particle temperature profile, $T_{\text{p}}$, (d) standard deviation of particle temperature, $s(T_{\text{p}})$, and (e) average particle moisture content, $\bar{W}_{\text{p}}$, in mg kg$^{-1}$.

In the sensitivity study, of which the results are presented in Tables 3.5 and 3.6, and in Figure 3.37, the batch size had only an effect on the particle-related modelled properties including the particle temperature ($T_{\text{p}}$), moisture content ($W_{\text{p}}$) and growth rate distributions ($\delta_{\text{c}}$). More detail, including the model predicted axial temperature and humidity profiles for both the gas
and solid phase is provided in Figure 3.55. The non-linear effects of the batch size on the particle related thermodynamic properties and to a lesser extent, the effects on the bed’s air thermodynamic properties, clearly illustrate the difficulties associated with scaling up the batch fluidised bed coating process. Consequently, the probability of the occurrence of side-effects like agglomeration or spray-drying – which are determined by the bed’s thermodynamic properties – change during scale-up of the coating process. In this respect, the proposed model (especially due to inclusion of a sectioned population balance model) can help in understanding the scale-up mechanisms to determine the process variables of a scaled-up process. Also, a small effect on the heat loss and consequently, on the outlet air temperature exists. However, the relationship between the heat loss and bed (or batch) size is complex: first, a larger bed implies a larger contact surface which increases the overall heat loss rate. Second, the batch size also affects the axial temperature gradient across the bed and the particle temperature distribution. Finally, the heat loss mechanism is also affected by the bed voidage which does not remain constant when the batch size is modified. In fact, the heat loss rate appeared to increase when the batch size was increased, but at $M_{\text{bed}} = 1.75$ kg a peak value for $\Phi_{\text{loss}}$ was predicted, after which the heat loss rate decreased again probably as a result of these described interacting effects.

3.7.3.12. Core material (thermal) properties

The last group of variables that was studied in this sensitivity analysis consisted of core material-related properties. In the model, the following material-related variables were used: the core material specific heat, $C_{p,p}$, the particle density, $\rho_p$, and the core material thermal conductivity, $\lambda_p$. From Tables 3.5 and 3.6, and from Figure 3.37, it can be deduced that the particle density and specific heat have an effect on the heat loss, while the particle density has a rather strong effect on the particle temperature, particle moisture and coating growth rate distribution. The reason for this behaviour is that the particle density is inversely proportional to the number of particles in the bed when the batch size, $M_{\text{bed}}$, is kept constant. The thermal conductivity of the core material, $\lambda_p$, had no effect on any of the model-predicted output variables.

To study the effect of $C_{p,p}$ and $\rho_p$ into more detail, two series of simulations were performed: First, $C_{p,p}$ was varied between 1000 and 5000 J kg$^{-1}$K$^{-1}$ and second, the core material density was varied between 1400 and 3000 kg m$^{-3}$. The model-predicted axial temperature and humidity profiles at steady state coating regime and for both gas and solid phase are presented in Figures 3.56 and 3.57. The effect of $C_{p,p}$ is clearly visible on the axial air and particle temperature profile: as expected, increasing the specific heat of the core particles – or in other words, increasing the particle’s thermal inertia – results in an decreased axial temperature gradient in both gas and solid phase (Figures 3.56a and 3.56c). The change in the relative humidity profile of the fluidisation air (Figure 3.56b) is purely the result of the altered air
temperature profile, the axial absolute humidity profile (not shown in Figure 3.56) was unaffected by $C_{p,p}$.

![Figure 3.56](image)

**Figure 3.56.** Model-predicted axial thermodynamic profiles during steady state conditions resulting from varying the core material's specific heat ($C_{p,p}$): (a) air temperature profile, $T_{a,i}$, (b) air relative humidity profile, $\Phi_{a,i}$, (c) average particle temperature profile, $\bar{T}_p$, (d) standard deviation of particle temperature, $s(T_p)$ and (e) average particle moisture content, $\bar{W}_p$ in mg kg$^{-1}$.

In Figure 3.57, the axial profiles are expressed in dimensionless coordinates because the particle density also affects fluidisation behaviour, including the particle exchange rate, $r_i$, and
the bed height, $h_{\text{bed}}$. The recalculation of these variables is similar as in Sections 3.7.3.4, 3.7.3.10 and 3.7.3.11 and will not be discussed into further detail.

Figure 3.57. Model-predicted axial thermodynamic profiles during steady state conditions resulting from varying the core material’s density ($\rho_p$): (a) air temperature profile, $\overline{T}_{a,i}$, (b) air relative humidity profile, $\overline{\varphi}_{a,i}$, (c) average particle temperature profile, $\overline{T}_p$, (d) standard deviation of particle temperature, $\sigma(T_p)$ and (e) average particle moisture content, $\overline{W}_p$ in mg kg$^{-1}$.

From Figure 3.57, it can be seen that core material density only had an influence on the particle temperature and particle humidity distributions. Decreasing the particle density tends
to increase the axial particle temperature gradient across the bed, while the axial air temperature gradient is much less affected compared to varying the specific heat of the core material as input variable. These effects can be attributed to the increase of the total number of particles present in the bed, $N_{\text{bed}}$, at lower particle densities.

### 3.7.4. Conclusions

From the sensitivity analysis of the model proposed in this chapter, the following conclusions can be drawn:

- When the average fluidised bed thermodynamic properties are considered, the main affecting variables are, in descending order: the inlet air temperature, the inlet air flow rate and the spraying rate. The effect of these variables is clearly visible in the outlet air thermodynamic properties and in both the thermal and vaporisation efficiency. Because of their impact on the bed thermodynamic operation point, these variables are commonly used in coating process control strategies. Although the spraying rate does not have the highest effect (or translation factor) for the thermodynamic operation point of the fluidised bed, it is one of the variables of choice in terms of controllability and responsiveness in a real fluidised bed coating system as opposed to control by means of the inlet air temperature, which has a slower response time, or by means of the inlet air flow rate, which is constrained due to its relationship with the bed’s fluidisation characteristics.

- The variables $r_i$ and $c/n$ play a major role in determining the particle temperature, moisture content and coating growth distributions although they do not alter the outlet air thermodynamic properties. These variables could be referred to as ‘internal’ variables: they are hard to measure in-situ, but have a strong influence in determining coating mass uniformity and agglomeration tendency – through the (non)-uniformity of the particle moisture content. Furthermore, these variables are controllable, but only to a certain extent. For instance, the particle exchange rate could be modified using the inlet air flow rate, but this in turn greatly affects the bed temperature and humidity. Using modifiable reactor geometries like moving baffles or draft tubes, could also be used in controlling the particle exchange rate without affecting the bed’s thermodynamic operation point. However, the introduction of additional reactor complexity neutralises one of the main benefits of top-spray coating, namely that its simple reactor configuration allows the scale-up to larger reactor volumes compared to Wurster or tangential coaters (which are inherently more complex in reactor configuration) and consequently reduces production costs of coating products.

- The use of pneumatic nozzles in the fluidised bed coating process results in the introduction of additional (compressed) air into the bed. Consequently, the impact atomisation air properties on the fluidised bed’s thermodynamics could not be neglected. However, as will be shown in Chapter 4, the atomisation air pressure has
more relevance when considering its effect on the particle temperature and moisture distribution and spray drying losses because the atomisation air pressure is one of the major factors determining the droplet size.

- Some of the core material related properties, including particle density, specific heat, diameter and batch size have a non-negligible effect on the particle temperature and moisture distribution, as well as on the axial temperature and humidity distributions of the bed’s air.
4. Droplet deposition and migration behaviour in liquid-sprayed fluidised beds

4.1. Introduction

In the previous chapter, a thermodynamic model combining population balances for the solid (particle) phase was developed. The combination of population balances and the two-phase heat and mass balances proved its capability to provide further insight in the bed’s thermodynamics and how the coating material is distributed among the fluidised particles. However, it was obvious in the model design and in the sensitivity analysis afterwards, that the proposed model had a few shortcomings, which included:

- The model required knowledge of the relative size of the coating region \( (c/n) \), which is the part of the bed where both spraying droplets and particles coexist and droplet-particle collision and collection take place. However, the size of this region is likely to depend on a number of variables, such as the atomisation air pressure, the coating solution properties (viscosity and surface tension) and the evaporative capacity of the supplied air. Furthermore, the sensitivity analysis revealed that the relative size of the coating region significantly contributes to the particle property variables, including particle temperature, moisture content and coating layer growth distributions.

- In the model, the coating solution is equally distributed over \( c \) coating control volumes and each coating control volume receives \( \dot{M}_{\text{sol}}/c \) coating solution (Figure 4.1a).

![Figure 4.1](image_url)

**Figure 4.1.** The distribution of the coating solution over the different coating control volumes in the model (a), compared with (a more probable) real coating solution distribution in the bed (b).
However, as the spray produced by the nozzle, migrates downward through the porous bed, its droplet concentration decreases. Whether this droplet concentration decrease is linear or non-linear is unknown. Consequently, when \( c \) coating control volumes are present, it is quite likely that the topmost control volume collects the largest volume of the droplets compared to lower positioned control volumes as illustrated in Figure 4.1b.

- Furthermore, it was assumed that within the coating volume, the coating solution was continuously divided over each particle. In reality, the coating solution is discontinuously (as droplets) distributed over the particles. For example, if there are approximately \( 10^7 \) particles in a small fluidised bed of \( M_{\text{bed}} = 1 \) kg and the relative size of the coating volume, \( c/n = 0.2 \), then there are constantly \( 2 \times 10^6 \) particles receiving coating solution. If the spraying rate is set to \( 10 \) g min\(^{-1} \), then – assuming continuous distribution of coating solution – each particle receives about \( 8.3 \times 10^{-11} \) kg s\(^{-1} \). Or, when using a time step of \( \Delta t_{\text{sim}} = 1 \) ms, each particle receives \( 8.3 \times 10^{-14} \) kg cycle\(^{-1} \).

Suppose in a real process, droplets are being produced with an average diameter of 15 \( \mu \)m, then each droplet carries \( 1.77 \times 10^{-12} \) kg, which is much more than the coating solution quantity each particle was assumed to receive in the model proposed in Chapter 3. Figure 4.2 illustrates how the real moisture content profile of a single particle in the coating region of the bed could look like compared to the modelled moisture content profile. In the sensitivity analysis of the model in Chapter 3, it was observed that the drying rate is usually not limiting. Considering that, as a result of the discontinuous supply of coating solution in the form of droplets, the actual moisture content could vary within a significantly larger range (Figure 4.2), the drying rate could become limiting much more rapidly than predicted by the model; an effect which will influence the overall axial bed’s temperature and humidity profiles. Not only is the particle moisture distribution affected by this phenomenon, also the coating mass distribution will be affected by the discontinuous supply of coating material towards the particles.

![Figure 4.2](image)

**Figure 4.2.** The moisture content profile \( (W_p) \) of a single particle while residing in the coating region, comparison between modelled and real moisture profile.
It could be demonstrated that, because of the small diameter of the droplets encountered in a typical fluidised bed coating process, the heat and mass transfers occurring between the droplet and the gas phase before the droplet adheres onto a fluidised particle are significant. If the droplet-gas interactions would be incorporated somehow into the model, it would become possible to predict the extent to which droplets evaporate before impinging on the fluidised particle. Hence, the spray drying loss and coating efficiency (as described by Jones (1985) and Dewettinck and Huyghebaert (1998), respectively) could be calculated by means of the model.

In this chapter, the model proposed in Chapter 3 will be extended through the addition of a third phase, namely the droplet phase. This allows including the complex droplet/gas and droplet/particle interactions resulting in the capability to predict spray drying losses (or coating efficiencies) in the batch fluidised bed coating process. As will be shown later on in this chapter, the inclusion of the droplet phase poses additional problems in solving the model which will be solved by proposing a second model which calculates individual droplet trajectories.

Next, the extended model will be experimentally validated. As already stated in the previous chapter, measuring the outlet air thermodynamic properties of a lab-scale fluidised bed unit (e.g. Glatt GPCG-1) proved insufficient. To that purpose, a larger fluidised bed unit was built, specifically designed to measure local air temperature and humidity profiles inside the bed. Besides thermodynamic validation, the coating efficiency will also be used to validate the model.

Finally, once the extended model is validated, a sensitivity analysis will be carried out. The addition of the droplet phase introduces an additional number of variables in the model, such as the relative position of the nozzle to the bed, the coating solution properties (surface tension, viscosity) and nozzle construction parameters. The sensitivity study in this chapter will mainly focus on these spray-related variables.
4.2. Extending the two-phase model with the droplet phase

4.2.1. Extended model description

The foundation of the extended model remains the same as the base model described in Chapter 3: The fluidised bed was one-dimensionally discretised into \( n \) constant-volume, constant-particle control volumes. However, in each control volume, three different phases are now distinguished: the gas (air) phase, the solid phase (particles) and the droplet phase. Figure 4.3 illustrates a detailed control volume containing the three phases and their interactions. In modelling these three phases within the fluid bed, the following additional assumptions were made besides the assumptions under 3.2:

- The arbitrary distinction between coating and non-coating control volumes is not maintained. Whether coating (or droplet/particle adhesion) takes place in a certain control volume \( S_i \) depends on whether the droplet phase is capable of penetrating the bed towards the \( i^{th} \) control volume, which in turn, is determined by factors such as the evaporative capacity of the bed, bed voidage and kinetic energy of the droplets.
- The particles are mechanically inert; there is neither agglomeration nor attrition. Particles are considered non-porous (e.g. glass beads) and do not absorb coating solution. Due to the low level of liquid loading of the fluidised bed, characteristic of coating processes, the impact of liquid bridge formation on the fluidisation behaviour – and hence, particle exchange rates – is considered not to be significant. According to Schaafsma et al. (1999), particle mixing behaviour and minimum fluidisation gas velocity are not significantly influenced by the relative humidity as long as the relative humidity of the bed’s gas phase does not exceed 0.5.
- The droplet phase travels downward (i.e. countercurrent to the gas phase) through the particle bed. Before contacting the particle surface, droplets exchange heat and mass with the surrounding air. If no successful adhesion occurs before complete droplet evaporation, dry fines are produced. In this model, dry fines are assumed to be completely removed from the bed by the fluidising air. In reality, however, part of the spray dried fines may stick to the surface of a wetted particle, resulting in coating imperfections (Smith and Nienow, 1983).
- Depending on droplet size, droplet kinetic energy and evaporative capacity of the bed, it is possible that droplets will eventually hit the reactor wall or the air distributor, especially in small reactor configurations. These so-called ‘wall losses’ are also taken into account when the droplet phase mass and heat transfer balances are constructed.
The droplet phase was characterised by 5 variables in each control volume: The control volume averaged droplet temperature, \( T_{dp,i} \), the average droplet dry matter content, \( DM_{dp,i} \), the droplet phase mass, \( M_{dp,i} \), the average droplet diameter, \( d_{dp,i} \), and finally, the number of droplets (or droplet number concentration) in each control volume \( S \). Unlike the solid phase (or particles), the number of droplets during the migration of the spray through the bed does not remain constant due to droplet-particle adhesion and premature droplet evaporation. Consequently, to characterise the droplet phase, two continuity equations (instead of one) are required, namely the droplet mass balance and the droplet number balance.

![Image of a control volume illustrating the addition of the droplet phase to the model.](image)

**Figure 4.3.** Scheme of a control volume illustrating the addition of the droplet phase to the model.

### 4.2.2. Heat and mass balances

#### 4.2.2.1. Gas, solids and droplet phase related balances

Using the heat and mass balances from the simple (continuous) model, described in 3.3.1, the addition of the droplet-interaction terms resulted in the following equations for the dynamic heat and mass balances for air, particles, coating material and droplets:

**Particles mass balance in a single control volume (solid phase continuity equation).** The solid phase continuity equation does not require modification, hence Eq. (3.1) is still valid.

**Air mass balance in a single control volume (gas phase continuity equation).** Eqs. (3.2) and (3.3) remain unchanged as the gas phase continuity equations. Although there is no longer arbitrary selection of the number of coating volumes, \( c \), considering the distribution of the coating solution droplets throughout the bed, the atomisation air is still assumed to be distributed over the upper \( c \) control volumes. In Chapter 5 more information will be provided on how the distribution of the atomisation air into the bed is calculated in detail.
**Droplet mass balance in a single control volume (droplet phase continuity equation).** The sink terms in each control volume include the spray drying losses ($\dot{M}_{\text{sd},i}$ in kg dry matter s$^{-1}$), droplet collection onto the fluidised particles (expressed by the droplet collection rate, $r_{c,i}$, in kg solution kg core$^{-1}$s$^{-1}$) and the (possible) droplet collection onto the inner reactor wall ($r_{c,\infty}^i$ in kg solution m$^{-2}$s$^{-1}$). Thus, the droplet phase continuity equation results in:

$$\frac{dM_{dp,i}}{dt} = J_{dp,i+1} - J_{dp,i} - \dot{M}_{\text{sd},i} - r_{c,i}M_{dp,i} - r_{c,\infty}^i A_{w,i}$$

(4.1)

In the droplet mass balance and in Figure 4.3, the droplet phase mass flow rate from $S_{i+1}$ to $S_i$ is given by the variable $J_{dp,i+1}$ to avoid confusion with the variable $M_{dp,i}$ which is the change in droplet phase mass in control volume $S_{i+1}$ over time due to evaporation and/or droplet collection onto particles. For the bottom control volume, $S_1$, the surface of the air distributor needs to be taken into account. Thus, the term $r_{c,1}A_{w,1}$ in Eq. (4.1) needs to be replaced with the term $r_{c,1}(A_{w,1} + 0.25\pi d_b^2)$.

Describing the droplet phase just by its mass balance results in additional difficulties as the droplet phase is not only characterised by its mass, $M_{dp,i}$, but also by the average droplet diameter, $d_{dp,i}$ and the number of droplets (or droplet number concentration) contained within the droplet phase, $N_{dp,i}$. The relationship between these three droplet phase properties is given by:

$$d_{dp,i} = \sqrt[3]{\frac{6M_{dp,i}}{\pi \rho_{dp,i} N_{dp,i}}}$$

(4.2)

In Eq. (4.2), the density of the liquid within the droplet phase in the control volume $S_i$, or $\rho_{dp,i}$ is calculated based on the dry matter content, with $\rho_c$ being the density of the dry coating material:

$$\rho_{dp,i} = DM_{dp,i}\rho_c + (1 - DM_{dp,i})\rho_{\text{water}}$$

(4.3)

Next to the droplet mass balance, the droplet number balance is constructed, which in conjunction with Eqs. (4.1) and (4.2) enables to calculate $M_{dp,i}$, $N_{dp,i}$ and $d_{dp,i}$ at the same time. Before building the droplet number balance, the droplet coalescence and breakage phenomena should be investigated as they are relevant to the droplet number balance as opposed to the droplet mass balance. In Section 1.3.2.2, the different droplet breakage mechanisms have been briefly discussed, along with the importance of the dimensionless Weber number.

Considering that the droplets usually have a diameter lower than 50 µm and using the model to predict droplet trajectories (and velocities) as will be explained in Section 4.3, the dimensionless Weber numbers were calculated as a function of distance from the nozzle using
an atomisation air pressure of 2.5 bar and assuming constant droplet diameter (no evaporation), the results are plotted in Figures 4.4 and 4.5. From these results, it can be observed that the model-predicted Weber number, $W_{edr}$, reaches a value below the threshold values for both drop breakage phenomena, just after the droplets have travelled approximately 6 mm from the nozzle. To conclude, droplet breakage, although important in the vicinity of the nozzle tip (< 6 mm), does not occur significantly in the fluidised bed.

![Figure 4.4](image1.png)

**Figure 4.4.** The calculated droplet Weber number as a function of the vertical distance from the nozzle for different (constant) droplet sizes.

![Figure 4.5](image2.png)

**Figure 4.5.** The calculated values for $W_{edr}/\sqrt{Re_{edr}}$ as a function of the vertical distance from the nozzle for different (constant) droplet sizes.
When using the same conditions as those used to plot Figures 4.4 and 4.5, the coalescence efficiencies could be calculated based on Eqs. (1.6) to (1.8) \( \left( d_{dr} = d_{dr,1} = d_{dr,2} \Rightarrow We_{dr} = We_{dr} \right) \). The results are plotted in Figure 4.6.

\[\begin{align*}
\text{Figure 4.6.} & \quad \text{The calculated droplet coalescence efficiency as a function of the vertical distance from the nozzle for different (constant) droplet sizes.}
\end{align*}\]

In Figure 4.6, it can be seen that the droplet coalescence efficiency quickly reaches the value of 1, at the same distance from the nozzle where droplet breakage becomes non-significant. The remaining problem is that the coalescence efficiency, \( \eta_{\text{coal}} \), only predicts the outcome of a droplet-droplet collision. In the regime where \( \eta_{\text{coal}} = 1 \), Von Smoluchowski’s theory of rapid coagulation could be used to calculate the rate of reduction in number of droplets due to droplet-droplet collision leading to successful coalescence (Vanderdeelen and Van der Meeren, 2000):

\[
\frac{dN_{dr}(t)}{dt} = -4\pi d_{dr} D_{dr} N_{dr}(0)
\]  

(4.4)

When calculating Eq. (4.4), maximum droplet reduction rates (\( dN_{dr}/dt \) in droplets s\(^{-1}\)) in the region where \( \eta_{\text{coal}} = 1 \) were found to range between \( -1.1 N_{dr}(0) \) to \( -1.9 N_{dr}(0) \). Although these values indicate a complete coalescing of the spray, the time it takes for a droplet to travel from the nozzle to the bottom of the fluidised bed (e.g. 0.25 m in a small scale fluidised bed) is generally in the order of 50 to 100 milliseconds as could also be calculated using the model proposed in Section 4.3. Consequently, the droplet coalescence occurring in such a short time span could be neglected (only < 5 % of the total number of droplets).
To conclude, neither droplet coalescence nor droplet breakage significantly contributes to the change in number of droplets within the droplet phase in the fluidised bed. Thus, the droplet number balance, based on Eq. (4.1), can be expressed as:

$$\frac{dN_{dp,i}}{dt} = \dot{N}_{dp,i+1} - \dot{N}_{dp,i} - \frac{6M_{sol,i}}{\pi \rho_{c,i} d_{sol,i}^3} - \frac{6}{\pi \rho_{dp,i} d_{dp,i}^3} \left( r_{c,i}^w A_{w,i} + r_{c,i}^p M_{pp,i} \right)$$

(4.5)

In Eq. (4.5) is $\dot{N}_{dp,i}$ the number of droplets transported from $S_i$ to $S_{i-1}$ per unit time. The number of droplets introduced in the top-most control volume (in top-spray configuration) is given by

$$\dot{N}_{dp,n+1} = \frac{6M_{sol}}{\pi \rho_{sol} d_{dr,noz}^3}$$

(4.6)

With $d_{dr,noz}$ being the average (stable) diameter of the droplets exiting the nozzle.

**Moisture balance of the particles.** Adding the droplet phase/particle-interaction term to Eq. (3.4) results in the following moisture balance equation, with $DM_{dp,i}$ being the local dry matter content of the droplet phase in $S_i$:

$$M_{pp,i} \frac{dW_{p,i}}{dt} = r_{i,i} M_{pp,bed} W_{p,i-1} + r_{r,i} M_{pp,bed} W_{p,i+1} - W_{p,i} M_{pp,bed} \left( r_{c,i} + r_{t,i} \right) - r_{D,i} M_{pp,i}$$

$$+ r_{c,i} M_{pp,i} \left( 1 - DM_{dp,i} \right)$$

(4.7)

**Moisture balance of the gas (air) phase.** As stated in the assumptions under 4.2.1, the amount of compressed air that is released to assist in atomising the coating solution was homogeneously distributed over the upper control volumes. Expanding Eqs. (3.6) and (3.7) with the terms describing the mass exchange between the gas phase and the droplet phase, including (possibly) attached droplets onto the reactor wall, the moisture balance for the gas phase becomes:

$$M_{a,i} \frac{dX_{a,i}}{dt} = G_{a,i} X_{a,i-1} - G_{a,i} X_{a,i} + r_{D,i} M_{pp,i} + r_{D,i} M_{dp,i} + r_{D,i}^w A_{w,i} \quad 1 \leq i \leq n - c$$

(4.8)

$$M_{a,i} \frac{dX_{a,i}}{dt} = G_{a,i} X_{a,i-1} - G_{a,i} X_{a,i} + \frac{1}{c} G_{a,i} X_{a,i} + r_{D,i} M_{pp,i} + r_{D,i} M_{dp,i} + r_{D,i}^w A_{w,i} \quad n - c < i \leq n$$

(4.9)

The moisture balance of the gas phase contains three different drying rate terms: the drying rate describing evaporation at the surface of the wetted particles, $r_{D,i}$, the droplet drying rate, $r_{D,i}^w$, and the the drying rate of droplets attached to the inner reactor wall, $r_{D,i}^w$. 

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"Modelling heat and mass transfer in fluidised bed coating processes"
Moisture balance of the droplet phase. The droplet moisture balance in a control volume $S_i$ is composed of moisture in- and output from the moving droplet phase, evaporation and condensation at the droplet surface and droplet collection onto the particle surface. It is possible that a fraction of the droplets does not impinge on the particle surface, but on the inner walls of the reactor, hence the variable $r_{C,i}^*$ in the collection term of Eq. (4.10):

\[
\frac{d}{dt} \left[ M_{dp,i} \left( 1 - DM_{dp,i} \right) \right] = \left( 1 - DM_{dp,i+1} \right) J_{dp,i+1} - \left( 1 - DM_{dp,i} \right) J_{dp,i} - r_{D,i}^* M_{dp,i} - \left( r_{C,i}^* M_{pp,i} + r_{C,i}^* A_{w,i} \right) \left( 1 - DM_{dp,i} \right) \tag{4.10}
\]

Coating mass balance of the particles. Instead of using arbitrary coating mass distribution among the upper $c$ control volumes as expressed in Eqs. (3.8) and (3.9), the coating mass balance of the droplet-phase extended model is based on the droplet collection rate, $r_{C,i}$, resulting in a single equation valid for all control volumes:

\[
M_{pp,i} \frac{dY_{p,i}}{dt} = M_{pp,bed} r_{r} Y_{p,i-1} + M_{pp,bed} r_{r} Y_{p,i+1} - M_{pp,bed} \left( r_{i} + r_{r} \right) Y_{p,i} + r_{C,i} M_{pp,i} DM_{dp,i} \tag{4.11}
\]

Coating mass balance of the droplets. Depending on the droplet drying rate and the diameter of the droplets in each control volume $S_i$, a fraction of the droplets will be spray dried, resulting in the formation of dry fines. It was assumed that dry fines are entirely removed from the bed by the fluidising air, therefore the coating mass balance of the droplet phase could be written as:

\[
\frac{d}{dt} \left( M_{dp,i} DM_{dp,i} \right) = DM_{dp,i+1} J_{dp,i+1} - DM_{dp,i} J_{dp,i} - \left( r_{C,i}^* M_{pp,i} + r_{C,i}^* A_{w,i} \right) DM_{dp,i} - \dot{M}_{sd,i} \tag{4.12}
\]

Heat balance of the particles. The particle heat balance of the control volume $S_i$ is similar to Eq. (3.11). However, in the latter equation, it was assumed that the droplets had the wet bulb temperature upon contacting the fluidised particles. In reality however, whether droplets attain the wet bulb temperature depends on the droplet size, droplet velocity and the distance between the nozzle and the receiving fluidised particles. Through the introduction of the droplet phase as the third phase in the model, it is possible – by means of the droplet phase heat balance – to model the temperature of the spray within each control volume $S_i$. The resulting particle heat balance is a modified form of Eq. (3.11):

\[
C_{p,p} M_{pp,i} \frac{dT_{p,i}}{dt} = r_{i} C_{p,g} M_{pp,bed} T_{p,i-1} + r_{C} C_{p,C} M_{pp,bed} T_{p,i} - C_{p,g} M_{pp,bed} T_{p,i} \left( r_{i} + r_{r} \right) + r_{C,i} M_{pp,i} C_{p,p,i} \left( T_{dp,i} - T_{p,i} \right) + a_{p,i} A_{pp,i} \left( T_{a,i} - T_{p,i} \right) - r_{D,i} M_{pp,i} Q_{lost,p,i} - \Phi_{loss,p,i} \tag{4.13}
\]
Heat balance of the gas (air) phase. Modifying Eq. (3.13) to accommodate the droplet/gas phase interaction terms, results in the following equation:

\[
M_{a,i}C_{p,a,i}\frac{dT_{a,i}}{dt} = G_{a,i-1}C_{p,a,i-1}T_{a,i-1} - G_{a,i}C_{p,a,i}T_{a,i} + \frac{1}{\alpha_p}G_{a,i}C_{p,at}T_{at} - \alpha_{p,i}A_{pp,i} \left( T_{a,i} - T_{p,i} \right) \\
- \alpha_{dp,i}A_{dp,i} \left( T_{a,i} - T_{dp,i} \right) - r_{D,i}M_{pp,i}C_{p,v,i} \left( T_{a,i} - T_{p,i} \right) - r_{D,i}^sM_{dp,i}C_{p,v,i} \left( T_{a,i} - T_{dp,i} \right) - \Phi_{loss,a,i} \tag{4.14}
\]

The term \( \left( G_{a,i}C_{p,at}T_{at} \right)/c \) in Eq. (4.14) is equal to zero when \( 1 \leq i \leq n - c \). Also, in Eq. (4.14) is \( A_{dp,i} \) the surface of the droplet phase which could be calculated based on Eq. (4.2) and the droplet number balance in Eq. (4.5).

Heat balance of the droplets. Again, the dimensionless Biot number is calculated to verify whether internal heat transfer resistance inside the droplet has a significant influence on the gas/droplet heat transfer. Considering the diameter range of droplets \( 10 \leq d_{dp} \leq 40 \mu m \) it holds that the Biot number:

\[
Bi_{dp} = \frac{\alpha_{dp}d_{dp}}{\lambda_{dp}} << 0.1 \tag{4.15}
\]

Consequently, each droplet or particle is considered to be isothermal and is described by a single temperature (Straatma et al., 1999; Collier et al., 2004). Combining the enthalpy of the droplet phase input and droplet phase output in \( S_s \), convective heat transfer between droplet phase and gas phase (air), evaporation at the droplet surface with the different droplet sink terms – such as droplet collection onto particles or reactor wall and the removal of spray-dried fines – yields the following droplet heat balance:

\[
M_{dp,i}C_{p,dp,i}\frac{dT_{dp,i}}{dt} = J_{dp,i+1}C_{p,dp,i+1}T_{dp,i+1} - J_{dp,i}C_{p,dp,i}T_{dp,i} - \left( r_{C,i}M_{pp,i} + r_{C,i}^sA_{w,i} \right)C_{p,dp,i}T_{dp,i} \\
- \dot{M}_{sd,i}C_{p,z}T_{sd,i} - r_{D,i}^sM_{dp,i}Q_{sat,i} + \alpha_{dp,i}A_{dp,i} \left( T_{a,i} - T_{dp,i} \right) \tag{4.16}
\]

In Eq. (4.16) is \( T_{sd,i} \) the temperature of the spray dried fines and was assumed to equal \( T_{dp,i} \). The specific heat of the aqueous solution, such as \( C_{p,dp,i} \) in Eq. (4.16) is calculated using the following equation (Earle, 1983):

\[
C_{p,dp,i} = 4186 \left( 1 - DM_{dp,i} \right) + C_{p,z}DM_{dp,i} \tag{4.17}
\]

4.2.2. Fluidised bed/reactor wall interactions

The theory of the heat loss mechanisms occurring near the reactor wall and its implementation into the model have been explained in Chapter 3. However, depending on the evaporative capacity of the fluidised bed and both the nozzle and coating solution parameters influencing...
the initial droplet size distribution and droplet exit velocity, a fraction of the produced droplets will impact on the inner reactor wall (Figure 4.7). The heat required to evaporate the wall-collected droplet phase has to be subtracted from the wall element heat balance as given in Eq. (3.19). Consequently, the overall heat balance of a single wall element $S_{w,i}$ could be written as:

$$
C_{p,w} \rho_w A_{w,i} d_w \frac{dT_{w,i}}{dt} = \frac{A_{w,i} C_{hu,w} (T_{a,i} - T_{w,i})}{R_{conv,i}} + \frac{A_{w,i} (1 - C_{hu,w}) (T_{p,i} - T_{w,i})}{R_{p,i}} - \sigma v' \epsilon_w A_{w,i} (T_{w,i}^4 - T_{c}^4)
$$

$$
- \frac{A_{w,i}'}{R_{conv,i}'} (T_{w,i} - T_{c}) + \frac{\pi d_{s,i} d_w}{R_{cond,i-1}} (T_{w,i-1} - T_{w,i}) - \frac{\pi d_{s,i+1} d_w}{R_{cond,i}} (T_{w,i} - T_{w,i+1})
$$

$$
- r_{D,i} A_{w,i} Q_{lat,i} + r_{C,i} A_{w,i} C_{p,dp,i} (T_{dp,i} - T_{w,i})
$$

(4.18)

**Figure 4.7.** Overview of the heat transfers occurring at the reactor wall.
4.3. Modelling individual droplet behaviour in the fluidised bed

4.3.1. Introduction and model scope

In the droplet phase extended model presented in the previous section, the addition of the third phase, i.e. the droplet phase, resulted in the introduction of a set of unknown variables. More specifically, the droplet collection rates \( r_{c,i} \) and \( r_{c,i}^\infty \) and the droplet drying rate \( \dot{M}_{D,i} \) in each control volume are still unknown. Also, the degree to which droplets evaporate \( M_{sd,i} \) resulting in the formation of dry fines as a function of the control volume’s air thermodynamic properties is yet unknown.

In order to quantify these unknown variables, a separate droplet submodel was coupled with the main model (Figure 4.8). The aim of this droplet submodel is to calculate the spatial distribution of the sprayed liquid inside the fluidised bed, the rate at which droplets adhere onto the fluidised particles and the solvent evaporation occurring between droplet production at the nozzle and droplet collection onto the particle surface as a function of the fluidised bed’s air and particle thermodynamic properties.

![Figure 4.8](image)

**Figure 4.8.** Coupling the main (extended) model with the droplet submodel which calculates the droplet-related interaction terms which are fed back as inputs in the main model.

In this submodel, droplet behaviour is determined by tracking the trajectories of individual droplets throughout the computational domain. The following assumptions were made:

- The physical system considered is a single droplet exiting the nozzle and travelling through the gas phase until successful adhesion onto the particle surface or reactor wall, or until the droplet has fully evaporated (Figure 4.9).
- The spray dynamics and gas flow patterns produced by the release of compressed air through the nozzle were assumed to be axisymmetric. Consequently, the droplet model was built as a two-dimensional axisymmetric model where the droplet position and velocity vectors were composed of an axial \( (h', dh'/dt) \) and a radial \( (r, dr/dt) \) component as shown in Figure 4.9. The difference between coordinate...
variable $h$ and $h'$ is that $h'$ uses the nozzle position as the origin, while $h$ uses the centre of the air distributor as origin, or $h' = h_{noz} - h$.

- The forces acting on the droplets are gravity, buoyancy and drag (Lorenzini, 2004).
- Each droplet remains a unique entity until it is either spray dried or collected upon the particle or reactor wall surface. So, droplet coalescence or breakage is not taken into account as already explained in Section 4.2 (Sazhin et al. 2005)
- During its flight, the droplet retains a spherical shape, but the droplet size is variable due to water evaporation before droplet impact on a bed particle.
- The core particles are perfectly spherical and non-porous.
- Considering that the diameter of the droplets produced in fluidised bed coating is generally lower than 40 µm (Guignon et al., 2002), the temperature inside the droplets is considered uniform ($Bi_{dp} << 0.1$, see Eq (4.15)). Likewise, the droplet contents are considered to be homogeneously distributed. Therefore, effects such as skin or crust formation during drying are not taken into account (Farid, 2003).

![Figure 4.9](image)

**Figure 4.9.** The physical system considered in modelling the droplet dynamics along with the jet velocity profiles produced by the pneumatic nozzle as described by Schlichting et al. (2004).

### 4.3.2. Droplet production and droplet motion

The diameter of the droplets that are produced by the pneumatic nozzle depends on a number of factors, relating both to the construction parameters of the nozzle and to the properties of the spraying liquid and the atomisation air, such as liquid viscosity and surface tension (Schaefer and Wörts, 1978; Cole et al., 1995; Juslin et al., 1995). In atomisers assisted by compressed air, liquid break-up occurs directly at the nozzle exit, as was verified in 4.2.2.1, where stable droplet diameter was predicted – using the Weber number – at a distance between 6 and 7 mm from the nozzle.
The initial droplet diameter was calculated using the model for pneumatic nozzles, proposed by Lefebvre (1988) in Eq. (1.1). The droplet exit velocity \( v_{\text{dr, noz}} \) is assumed equal the velocity of the released atomisation air near the nozzle orifice (Link and Schlünder, 1997). This theoretical maximum velocity is thus calculated as:

\[
v_{\text{dr, noz}} \approx v_{\text{at, noz}} = \sqrt{\frac{2 \Delta P_{\text{at}}}{\rho_{\text{at}}}}
\]

(4.19)

Resulting from the second assumption, the balance of forces exerted upon a single droplet can now be written as

\[
\mathbf{F}_{\text{dr}} = M_{\text{dr}} \ddot{a}_{\text{dr}} = M_{\text{dr}} \ddot{g} + \mathbf{F}_{\text{buoy}} + \mathbf{F}_{\text{d}}
\]

(4.20)

In Eq. (4.20) is the third force component, \( \mathbf{F}_{\text{d}} \) the drag force exerted on the droplet. It is calculated as

\[
\mathbf{F}_{\text{d}} = \pi C_D \rho_a \frac{d_{\text{dr}}^2}{8} \left( \bar{\nu}_a + \bar{\nu}_{\text{at}} - \bar{\nu}_{\text{dr}} \right)^2
\]

(4.21)

The effective drag coefficient, \( C_D \), is generally greater than the standard drag coefficient, \( C_D^* \), which is the case if the same droplet moves in an infinite fluid in the absence of solid particles in the fluidised bed. The effective drag coefficient, \( C_D \), can be estimated based on an empirical equation proposed by Mostoufi and Chaouki (1999):

\[
P_{\text{bed}} \cdot C_{D^*} = \frac{m}{\left( \frac{d_{\text{dr}}}{d_p} \right)^0.4}
\]

(4.22)

\[m = 3.02 Ar_p^{0.22} Re_{\text{dr, te}}^{0.33} \left( \frac{d_{\text{dr}}}{d_p} \right)^{0.4}\]

(4.23)

As shown in Eq. (4.23), the droplet effective drag coefficient is a function of the fluidised bed voidage fraction, the droplet Reynolds number at terminal velocity, the solids’ Archimedes number and the ratio of the droplet diameter to the bed particle diameter (Mostoufi and Chaouki, 1999; Wang and Zhu, 2003). The standard drag coefficient \( C_D^* \) in Eq. (4.22) for spherical objects is calculated according to the correlation given by Turton and Levenspiel (1986),

\[
C_D^* = \frac{24}{Re_{\text{dr}}} \left( 1 + 0.173 Re_{\text{dr}}^{0.657} \right) + \frac{0.413}{1 + 16300 Re_{\text{dr}}^{-1.09}}
\]

(4.24)

In order to calculate the drag force on a droplet, the velocity profile, \( \bar{\nu}_a \left( h', r \right) \), produced by the release of compressed air to assist in the atomisation of the coating solution has to be known. As stated by various authors, e.g. Donadono et al. (1980), Becher and Schlünder
(1997) and Zank et al. (2001), the nozzle jet in a fluidised bed shows many similarities to a free axisymmetric jet of which the gas velocity profiles are analogous to Schlichting et al. (2004), (see Figure 4.9):

\[ v_{at,ax}(h',\xi) \approx \frac{1}{(h')^{2/3}} \frac{1}{\left(1+0.25\xi^2\right)^2} \hspace{1cm} (4.25) \]

\[ v_{at,rd}(h',\xi) \approx \frac{\xi - 0.25\xi^3}{\left(1+0.25\xi^2\right)^2} \hspace{1cm} (4.26) \]

In Equations (4.25) and (4.26) is \( \xi \) the dimensionless radial coordinate with

\[ \xi = \frac{2x}{x_{at}(h')} \hspace{1cm} (4.27) \]

The jet radius produced by the pneumatic nozzle, \( x_{at}(h') \), is defined as the radius where the radial gas velocity, \( v_{at,rd}(h', x) \), equals zero.

4.3.3. Droplet thermodynamics

4.3.3.1. Droplet heat balance

During its flight, the droplet exchanges heat and mass with the surrounding gas phase (air), of which the temperature and humidity axial profiles are known from the main model. The energy required for the evaporation and the convective heating of a single droplet equals the heat transferred from the gas phase (Chen et al., 2000; Gupta and Rao, 2001; Macleod et al., 2006). The resulting heat balance can then be written as:

\[ M_{dr} C_{p,dr} \frac{dT_{dr}}{dt} = a_{dr} \pi d_{dr}^2 \left( T_{at,\Lambda(h_{dr})} - T_{dr} \right) - Q_{at} r_{D,dr}^2 M_{dr} \hspace{1cm} (4.28) \]

The operator \( \Lambda(h_{dr}) \) in Eq. (4.28) determines in which control volume, as defined in the main model, the droplet resides at height \( h_{dr} \). The convective heat transfer coefficient \( a_{dr} \), was approximated using the dimensionless Nusselt number which was calculated using the Whitaker equation (cfr. Section 1.5.2.2). Just as in the case of drying of wetted solids, different regimes can be distinguished during the drying of a single droplet as shown in Figure 3.7 (Cole et al., 1995). However, in modelling the droplet vaporisation, only the constant drying rate period was considered – section BC on the drying rate curve in Figure 3.7. The droplet drying rate \( \dot{D}_{r} \) was calculated using Eq. (3.24). For small spherical geometries, such as droplets in the micron range, the vapour pressure at the droplet surface also depends on the diameter. This vapour pressure correction is given by the Kelvin-Laplace equation, Eq. (3.30).
4.3.3.2. Droplet mass balance

The change in droplet mass is given by the drying rate, as long as the moisture content of the droplet is not limiting:

\[
\frac{dM_{dr}}{dt} = -r_d M_{dr} \quad DM_{dr} < 1
\]

\[
\frac{dM_{dr}}{dt} = 0 \quad DM_{dr} = 1
\]

As the mass of coating material in the droplet is invariable, the dry matter content was calculated as:

\[
DM_{dr} = \frac{DM_{dr, noz} M_{dr, noz}}{M_{dr}}
\]

The droplet diameter also varies as a function of the water content of the droplet,

\[
d_{dr} = \left( \frac{6M_{dr}}{\pi} \left( \frac{DM_{dr}}{p_c} + 1 - \frac{DM_{dr}}{1000} \right) \right)^{1/3}
\]

4.3.4. Droplet/particle collision

The droplet trajectory could be terminated according to one of the following scenarios:

- The droplet evaporates entirely \((DM_{dr} = 1)\), producing dry fines. The dry fines are assumed to be entirely collected in the fluidised bed’s filter system.
- The droplet impinges on the reactor wall or on the air distributor. The solvent (water) in the attached droplet evaporates, resulting in the deposition of coating onto the inner reactor wall surfaces.
- The droplet successfully impinges onto a fluidised particle. However, when the droplet trajectory crosses the path of a fluidised particle, adhesion of the droplet onto the particle is not assured. The probability that a liquid droplet is successfully collected by a particle when their paths meet, is expressed by droplet collection efficiency, \(\zeta_{dr}\) (Heinrich et al., 2003).

In Section 1.3.2.3, the equations were given to calculate the impingement efficiency when a droplet approaches a fluidised particle. To calculate the Stokes number in Eq. (1.11), the particle velocity needs to be known. In Section 3.6.2.2, it was shown that, for a given core material and fluidisation gas flow rate, the axial particle dispersion coefficient, \(D_{p,ax}\), could be deduced. Furthermore, according to Bellgardt (1985), the radial dispersion coefficient, \(D_{p,rd}\), is usually lower by a power of 10 compared to the axial dispersion coefficient, or:
Using Einstein’s theory of Brownian motion (1905), the dispersion coefficient – which is analogous to the diffusion coefficient as used in Fick’s law – relates to the average particle displacement ($\Delta h_p$, $\Delta r_p$) in a time span of $\Delta t$, with $h$ and $r$ being the axial and radial particle coordinates, respectively:

\[
\sqrt{\Delta h_p^2} = \sqrt{2D_{p,ax}\Delta t} \quad \text{and} \quad \sqrt{\Delta r_p^2} = \sqrt{2D_{p,rd}\Delta t}
\]

\[
\bar{v}_p = \sqrt{\frac{\Delta h_p^2}{\Delta t^2} + \frac{\Delta r_p^2}{\Delta t^2}}
\]

Once the droplet has made initial contact with the particle surface, consecutive interaction between the droplet and the particle determines whether the droplet remains adhered to or bounces off from the particle surface. The fraction of droplets that remain permanently adhered on the particle surface is expressed by the adhesion probability, $\iota_{dr}$, and depends on the droplet impact kinetic energy and the wettability of the particle substrate (Link and Schlünder, 1997; Heinrich et al., 2003b). The critical impingement velocity for droplets impacting a flat, non-porous and dry surface was described by Link (Link and Schlünder, 1997; Zank et al., 2001). Droplets are reflected from the particle surface above this critical velocity:

\[
v_{crit} = \frac{4\mu_{dr}\left(3\tan\left(\frac{\theta_{dr/p}}{2}\right) + \tan^3\left(\frac{\theta_{dr/p}}{2}\right)\right)^{1/2}}{d_{dr} \rho_{dr} \tan^2\left(\frac{\theta_{dr/p}}{2}\right)}
\]

From Eq. (4.36) it can be seen that the critical impingement velocity not only depends on droplet characteristics such as viscosity, size and density, but also depends on the static liquid/solid contact angle, $\theta_{dr/p}$, which is correlated with the wettability of the receiver substrate (or in this case, the fluidised particle). Contrary to the impingement efficiency, the adhesion probability decreases with increasing droplet size and increasing droplet velocity.

The droplet collection efficiency, $\zeta_{dr}$, resulting from successful droplet impingement followed by droplet adhesion is given by multiplying the impingement efficiency and adhesion probability:

\[
\zeta_{dr} = \chi_{dr}\iota_{dr}
\]
4.3.5. Practical implementation and calculation method

A detailed discussion on how the droplet submodel was implemented within the existing calculation procedure can be found in Appendix B.3. The description in Appendix B.3 is split up in two parts: First, the discussion is focussed on how the main model simulation procedure could be modified to accept the output of the droplet submodel and how the third phase, i.e. the droplet phase, is integrated into the main model. Second, the actual procedures which make up the droplet submodel simulation, as described in Section 4.3, are detailed.
4.4. Statistical issues with stochastic, discrete models: assessing model performance

4.4.1. Introduction

In Chapter 3, prior to model validation and analysis, the critical number of control volumes, \( n \), and the minimum sample size, \( N_{\text{sim}} \), were assessed. It was concluded that if the number of control volumes, \( n \geq 24 \) and the number of simulated particles, \( N_{\text{sim}} \geq 10000 \), the error on the model-predicted output variables became almost independent of neither \( n \) nor \( N_{\text{sim}} \).

With the introduction of the droplet submodel, a third discrete model variable has been added, namely the number of the droplets to simulate during each run of the submodel, \( N_{\text{dr,sim}} \). Again, the impact of droplets to simulate, \( N_{\text{dr,sim}} \), had to be investigated and a critical value for \( N_{\text{dr,sim}} \) has to be established for future simulations.

4.4.2. Methods and calculations

To study the effect of the number of droplets used in the submodel, \( N_{\text{dr,sim}} \), besides the same model-predicted output variables of both gas phase and the particle population (solid phase), as detailed in Section 3.5, the following droplet-related output variables were added in this study:

- The spray drying loss at steady state coating regime, \( \sum_{i=1}^{n} M_{\text{sd,i}} \).
- The height distribution of successful droplet/particle adhesion during steady-state, \( h_{\text{drc}} \).
- The steady-state droplet temperature distribution at the moment of particle-impact, \( T_{\text{drc}} \).
- The steady-state droplet dry matter content distribution at droplet/particle impact, \( DM_{\text{drc}} \).

The effect of the number of modelled droplets was studied with \( N_{\text{dr,sim}} \) ranging between 100 and 6500. This upper limit was the result of memory constraints with the used algorithm and hardware. More specifically, to store the droplet related properties and dimensionless numbers along its computed trajectory, approximately 300 kilobytes of computer memory was required, while a total of 2 gigabyte of memory was available. Each simulation consisted of a coating process of inert glass beads with a (hypothetical) spraying solution containing 10 w\% dry matter and having the properties of water (\( \rho_{\text{sol}}, \mu_{\text{sol}}, \gamma_{\text{sol}} \) and \( \theta_{\text{sol}} \)), more details concerning process conditions are summarised in Table 4.1. The simulated process consisted of two distinct phases: in the heating phase, no coating solution was sprayed (\( \dot{M}_{\text{sol}} = 0 \), droplet submodel inactive). Next, after 600 seconds of simulated time, a step input was given for the spraying rate (\( \dot{M}_{\text{sol}} = 10 \text{ g min}^{-1} \)).
Table 4.1. Model and operational parameters of the simulation.

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<th>Variable</th>
<th>Symbol</th>
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<th>Value</th>
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### Table 4.1. Model and operational parameters of the simulation (continued).

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<td>-</td>
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<td>J kg(^{-1})K(^{-1})</td>
<td>500</td>
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<td>kg m(^{-3})</td>
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<td>Wall mixing constant</td>
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<td>Hz</td>
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<td>Simulated time</td>
<td>( t_{sim} )</td>
<td>s</td>
<td>1500</td>
</tr>
<tr>
<td>Simulation time step</td>
<td>( \Delta t_{sim} )</td>
<td>s</td>
<td>0.001</td>
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<tr>
<td><strong>Simulation (droplet submodel) parameters</strong></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Maximum simulation time (single droplet)</td>
<td>( t_{dr,sim} )</td>
<td>s</td>
<td>0.5</td>
</tr>
<tr>
<td>Maximum time step **</td>
<td>Max(( \Delta t_{dr,sim} ))</td>
<td>s</td>
<td>2.5 \times 10^{-5}</td>
</tr>
<tr>
<td>Minimum time step **</td>
<td>Min(( \Delta t_{dr,sim} ))</td>
<td>s</td>
<td>1.33 \times 10^{-6}</td>
</tr>
<tr>
<td>Maximum droplet temperature difference **</td>
<td>Max(( \Delta T_{dr} ))</td>
<td>°C</td>
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</tr>
<tr>
<td>Minimum droplet temperature difference **</td>
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<td>°C</td>
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</tr>
<tr>
<td>Maximum droplet displacement **</td>
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<td>m</td>
<td>2.0 \times 10^{-4}</td>
</tr>
<tr>
<td>Minimum droplet displacement **</td>
<td>Min(( \Delta x_{dr} ))</td>
<td>m</td>
<td>5.0 \times 10^{-4}</td>
</tr>
</tbody>
</table>

**Remarks:**

* Properties of water at 20 °C, taken from De Man (1999)

** Relating to the adaptive time stepping algorithm in the simulation of the droplet submodel (Appendix B). These parameters are obtained through trial-and-error, where values are selected yielding sufficiently fast calculations, while the rejection due to uncontrollable oscillations is kept below 5%
In Table 5.4, the jet resulting from the release of compressed air into the reactor is characterised by a so-called shape factor. It could be deduced from Eqs. (4.25) and (4.26), which describe axial and radial gas velocity profiles within this jet, that the jet radius, \( x_{\text{noz}}(h') \) has a parabolic profile:

\[
x_{\text{noz}}(h') \sim h'^{1/6}
\]

Consequently, the shape factor is defined as the proportionality in Eq. (4.26), or

\[
x_{\text{noz}}(h') = s_n h'^{1/3}
\]

Full detail, including a simple method in experimentally deriving the shape factor, shall be given in Section 4.6.

4.4.3. Results and discussion

a) Droplet phase related model-predicted variables

Figure 4.10 illustrates the model-predicted spray drying loss during steady state coating regime, expressed in \( w\% \) of the mass flow rate of the coating solution, as a function of the number of simulated droplets, \( N_{dr,sim} \). When considering the effect of increasing number of simulated droplets on the model-predicted spray drying loss, it could be observed that the simulated spray drying loss was likely to reach an asymptotic value beyond \( N_{dr,sim} = 6500 \). It should be stressed that at that point, the differences in simulated spray drying losses were very small (< 0.02 %).

![Figure 4.10](image-url)

**Figure 4.10.** Influence of the number of simulated droplets, \( N_{dr,sim} \) on the model-predicted spray drying loss during steady state coating regime mass, in \( w\% \) of injected coating solution
A similar conclusion could be drawn when considering the droplet collision height, $h_{drc}$ (Figure 4.11), the droplet dry matter content at particle/droplet collision, $DM_{drc}$ (Figure 4.12) and the droplet collision temperature, $T_{drc}$ (Figure 4.13). Each time, the model-predicted output variables appeared to reach steady-state at a number of simulated droplets between 5000 and 6500.

Figure 4.11. Influence of the number of simulated droplets, $N_{dr,sim}$ on the average droplet collection height (○), in m above the air distributor, and its standard deviation (●).

Figure 4.12. Influence of the number of simulated droplets, $N_{dr,sim}$ on the average droplet collection dry matter content (○) and its standard deviation (●).
b) Gas phase (air) related model-predicted variables

Similarly, the effect of the number of simulated droplets on the outlet air properties was studied. The results for the modelled outlet air temperature, $T_{a,\text{out}}$, are given in Figure 4.14, while the effect on the outlet air humidity, $\varphi_{a,\text{out}}$, is presented in Figure 4.15. As opposed to the conclusions drawn with regard to the droplet related properties (section a), the outlet air properties already reach a constant value independent of $N_{dr,\text{sim}}$, when the number of droplets simulated in the submodel was above 3500.

Figure 4.13. Influence of the number of simulated droplets, $N_{dr,\text{sim}}$ on the average droplet collection temperature (-o-) in °C, and its standard deviation (-●-).

Figure 4.14. Influence of the number of simulated droplets, $N_{dr,\text{sim}}$ on the steady state outlet air temperature (-o-) and its standard deviation (-●-).
Figure 4.15. Influence of the number of simulated droplets, $N_{dr,sim}$, on the steady state outlet air relative humidity ($\rightarrow$) and its standard deviation ($\rightarrow$).

When considering the axial profiles of the fluidisation air’s thermodynamic properties ($\overline{T_{a,i}}$ in Figure 4.16, $s(T_{a,i})$ in Figure 4.17, $\overline{\varphi_{a,i}}$ in Figure 4.18 and $s(\varphi_{a,i})$ in Figure 4.19), it could be deduced that the critical (minimum) number of simulated droplets was situated between $N_{dr,sim} = 2500$ and $N_{dr,sim} = 5000$. The distortions in $s(T_{a,i})$ and $s(\varphi_{a,i})$ at low numbers of $N_{dr,sim}$ occur in the location where the majority of the successful droplet/particle adhesion takes pace.

Figure 4.16. Influence of the number of simulated droplets, $N_{dr,sim}$, on the axial average air temperature profile at steady state.
Figure 4.17. Influence of the number of simulated droplets, $N_{dr,sim}$, on the axial profile of the air temperature standard deviation during steady state.

Figure 4.18. Influence of the number of simulated droplets, $N_{dr,sim}$, on the axial profile of the average air relative humidity.
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**Figure 4.19.** Influence of the number of simulated droplets, \( N_{dr,sim} \), on the axial profile of the standard deviation on the air relative humidity.

c) **Particle population related model-predicted variables**

The influence of the number of simulated droplets, \( N_{dr,sim} \), on the model-predicted coating mass distribution, \( Y_p \) after \( t = 900 \) s of spraying is given in Figure 4.20. Figure 4.21 illustrates the effect of the number of simulated droplets on the global particle temperature distribution after reaching steady state conditions. The particle-related properties stabilised at about \( N_{dr,sim} = 5000 \) similar to the droplet-related output variables.

**Figure 4.20.** Influence of the number of simulated droplets, \( N_{dr,sim} \), on the average coating mass, in g coating/kg core after \( t = 900 \) s of spraying (○-○), and its standard deviation (●-●).
Figure 4.21. Influence of the number of simulated droplets, $N_{dr,sim}$, on the average particle temperature at steady state (○) and its standard deviation (●).

4.4.4. Conclusion

The droplet submodel-predicted variables, including spray drying losses and the droplet properties at the moment of droplet/particle impact, appear to be highly sensitive to changes in number of droplets simulated. Increasing the number of simulated droplets beyond $N_{dr,sim} = 5000$ or even $N_{dr,sim} = 6500$ is required to have model-predicted output data which is independent of the sample size, $N_{dr,sim}$. When considering the gas phase or particle population-related output variables, as predicted by the main model, it appears that the critical value for $N_{dr,sim}$ is lower ($N_{dr,sim} \leq 2500$ or 5000). Particle properties ($W_p$, $Y_p$) are more sensitive to $N_{dr,sim}$ than gas phase-related properties ($T_a$, $\varphi_a$), as coating mass and moisture deposition rates fully depend on the results of the droplet submodel (being the most sensitive).

Another factor to take into consideration when choosing the number of droplets in the simulation, is the additional calculation time resulting from the use of the droplet submodel. The reference simulation used in Chapter 3 – with 24 control volumes, including 10000 particles and for a process time of 900 seconds – took about 3 hours to finish. Adding the droplet submodel with $N_{dr,sim} = 5000$, increased the calculation time to approximately 8 hours. In fact, for every additional 1000 droplets, one hour of extra calculation time was needed. To conclude, a number of $N_{dr,sim} = 5000$ was chosen for future simulations, as this number of droplets was considered to result in tolerable model-generated error, while having acceptable calculation time (<8 h).
4.5. Model validation: thermodynamic

4.5.1. Introduction

It is clear from the model description in Sections 4.2 and 4.3 that the model does not modify the overall heat and mass balances of the fluidised bed and thus, the thermodynamic properties of the outlet air do not differ between the model described in Chapter 3 and the model extended with the droplet submodel being the subject of this chapter. Therefore, the use of temperature or humidity measurements of the exhaust air, as applied in Section 3.6, is inadequate to validate the model. A measurement method to gain deeper insights into the temperature and humidity distributions inside the fluidised bed had to be designed.

Several authors have already described measurement set-ups to record the temperature or humidity profiles inside a fluidised bed. These set-ups include the use of a single scanning (rotating) probe (Maronga and Wnukowski, 1997a) or the use of moving arrays of thermocouples through the bed (Heinrich et al., 2003b). In this experimental validation, the use of a single moving probe was chosen as it provides minimal bed disturbance (compared to moving rakes or arrays of sensors), minimum calibration effort (single sensor) and low experimental error (compared to multiple sensors in grid or array lay-out). The disadvantages include the long measurement time – thus steady state conditions must be assured – and the limited section of the bed (i.e. longitudinal or diametral symmetry plane) that could be scanned.

An additional difficulty comprised the small dimensions of the Glatt GPCG-1 and the fact that this pilot plant operates in underpressure (i.e. using a suction fan drawing air through the bed), which could create by-pass airflow if probe access holes were drilled in the reactor shell. Thus, a larger fluidised bed unit, operating in overpressure to facilitate probe access, was designed, specifically to be equipped with the scanning temperature/humidity probe for these thermodynamic validation experiments.

4.5.2. Materials and methods

4.5.2.1. Process equipment

A schematic of the experimental validation set-up is depicted in Figure 4.22, while the actual pilot installation and its major components are displayed in Figure 4.23. The tapered reactor had a bottom diameter of 0.225 m, a top diameter of 0.45 m and a total height of 0.84 m. The reactor shell material consisted of 5 mm thick steel. Fluidisation air was provided by a 2.2 kW high pressure centrifugal fan (Ventomatic CHT160-2T-3, Belgium) of which the pressure characteristics are depicted in Figure 4.24. This type of turbine was chosen as it has a large airflow rate interval with a constant (sufficiently high) pressure difference. Instead of using valves or diaphragms to control the turbine airflow by ‘choking’, an inverter (Omron PV-
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series, 3G3PV-A4040-E, Japan) was applied, which alters the centrifugal fan’s electrical supply frequency, and thus the rotational speed of the electric motor powering the fan. The volumetric air flow rate was measured between the fan and the heating coils by means of 0.1 m diameter rotating vane flow meter (Airflow Developments, VMD20, UK).

The process air was heated by 9 1.2 kW heating coils (Watlow RCN3510S, US), resulting in a total installed heating capacity of 10.8 kW. Because this heating capacity might be too large to adequately control the air temperature when using, for instance, small product batches ($M_{bed}$), the heating coils were arranged in 3 banks of 3 coils each, which could be switched manually on or off. Thus, the maximum heating capacity could be selected between 3.6, 7.2 or 10.8 kW. To control the temperature of the fluidisation air, measured by means of a K-type thermocouple in the inlet air plenum of the fluidised bed, a PID-controller (Cal Controls, 9400, UK) with 3 solid-state relays, one for each heating bank, were used.

Figure 4.22. Scheme of the experimental set-up.
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Figure 4.23. Experimental set-up

Figure 4.24. Pressure versus volumetric air flow rate characteristics of some of the high pressure centrifugal fans of the CHT-series provided by Ventomatic (B), (*) marks the type of fan used in the experimental setup.

The air distributor consisted of a Robusta 172×36 wpi (wires per inch) wire mesh (Spörl KG, Germany). A pneumatic nozzle (Schlick Model 970-S1, Germany) was installed at the tip of a vertical retractable rod, allowing adjustment of the nozzle height. This type of pneumatic
nozzle is identical to the type used in the Glatt GPCG-1. The coating solution was transported to the pneumatic nozzle by means of a peristaltic pump (Watson-Marlow, 505 Du/RL, US).

4.5.2.2. Core material and spraying solution

Micropearl® glass beads (Sovitec, Belgium) with a volume-weighted average diameter, $d_p = 365 \, \mu m$ were used as the core material. For thermodynamic analysis, distilled water was used as the spraying liquid.

4.5.2.3. Scanning measurement system

Steady state spatial air temperature and air relative humidity profiles within the fluidised bed were measured by means of a moving J-shaped probe. Figure 4.25 illustrates the details of the probe positioning element. It basically consisted of two electric motor-driven spindles. Probe displacement was registered by means of relative optical encoders (Hewlett-Packard HEDS-5700, US). A relative optical encoder measures the spindle rotation by means of a slotted disc which periodically interrupts a light beam (Figure 4.26). The result is an alternating voltage signal of which the frequency is directly proportional to the rotation speed of the spindle. The type of encoder used delivered 300 pulses per rotation and by taking the thread characteristics of the spindle (thread M12) into account, the relative displacement of the probe could be derived.

![Figure 4.25](image)

**Figure 4.25.** Schematic overview of the probe positioning element with two degrees-of-freedom.
Figure 4.26. Measurement principle of a single-beam relative photo-encoder

The motor control and sensor signals were provided or registered by means of a multifunction data acquisition and control PC-card (Eagle Technology, PCI-30G, South Africa). A program was written in VB 6.0 (Microsoft, US) to interface with this PC-card to control the movement or scanning pattern of the probe.

The probe consisted of a hollow aluminium tube. Only the tip of the probe was manufactured of PVC ($\lambda = 0.25$ W m$^{-1}$K$^{-1}$, $C_p = 1950$ J kg$^{-1}$K$^{-1}$). The temperature and humidity sensing elements were embedded in the tip of the probe (see Figure 4.27). A defined quantity of reactor air was drawn through the probe using an air sampling pump (SKC Airlight, US). Air temperature was measured by means of a K-type thermocouple, while relative humidity was measured by a capacitive humidity sensor (Honeywell HIH-3610, US) embedded in the tip of the probe. The Honeywell HIH-3610 is a discrete sensor element with integrated amplifier logic, returning a signal between 0 and +/- 3 V for the air relative humidity – which is a signal ideally suited for the data acquisition board (0…10 V input). This humidity sensor was chosen because, according to its manufacturer, it sustains condensing environments with rapid and full recovery (hysteresis). On the other hand, the thermocouple input could not be directly connected to the data acquisition board. A K-type thermocouple signal amplifier and conditioner, transformed the thermocouple voltage into a 0..5 V signal.
The moving probe sampled air along the points of a regular grid with a 0.01 m cell spacing. In each grid point, measurements were repeated 10 times in rapid succession (<1 s). During each experiment, all process parameters (such as inlet air temperature and flow rate, liquid flow rate) were kept constant.

The scanning was initiated when steady state conditions were reached. Steady state conditions were assumed when constant exhaust air temperature and air relative humidity were registered. Exhaust air thermodynamic properties were measured using a thermistor (National Semiconductor LM-35D, US) for air temperature, while air relative humidity was measured using a capacitive humidity sensor (Honeywell HIH-3610, US).

4.5.2.4. Data post-processing

The use of the scanning probe resulted in two-dimensional spatially distributed measurement data. However, the current model only predicts one-dimensional axial thermodynamic profiles for the three different phases involved. Therefore, the experimental axial thermodynamic profiles were derived from the measurements by taking the surface-weighted average at each height. For example, the calculation for $T_a(h_i)$ was carried out as,
\[ T_s(h) = \frac{1}{\chi^3_n} \sum_{i=1}^{n'} \left( \left( x_i + \frac{\Delta x}{2} \right)^2 - \left( x_i - \frac{\Delta x}{2} \right)^2 \right) T_s(h_i, x_i) \] (4.40)

In Eq. (4.40) is \( n' \) the number of radial measurement points. The variables \( x_i \) and \( h_i \) represent the radial and axial coordinates of the \( i \)th measurement points on the scanned grid.

### 4.5.2.5. Modelling and calculations

Due to the change in geometry – the reactor’s volume had been scaled up with a factor 2.25 – variables such as the particle exchange rate, \( r_i \), needed to be recalculated. In Table 4.2 all modified input variables and parameters are summarised, those omitted from Table 4.2 remain to have the same value as mentioned in Table 5.4. Also note that, for the larger reactor domain, it was necessary to halve the time step of the main model, \( \Delta t_{\text{sim}} \), to avoid uncontrolled oscillations in the simulation procedure.

#### Table 4.2. Modified model and operational parameters of the simulation.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Symbol</th>
<th>Unit</th>
<th>Value</th>
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</thead>
<tbody>
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<td><strong>Main model parameters</strong></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Particle exchange rate</td>
<td>( r )</td>
<td>Hz</td>
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</tr>
<tr>
<td><strong>Droplet sub model parameters</strong></td>
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<td></td>
</tr>
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<td>Number of simulated droplets</td>
<td>( N_{dr,\text{sim}} )</td>
<td>-</td>
<td>5000</td>
</tr>
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<td><strong>Fluidisation air properties</strong></td>
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</tr>
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<td>Inlet air volumetric flow rate</td>
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<td>kg s(^{-1})</td>
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<td><strong>Bed dimensions</strong></td>
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</tr>
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<td>( h_{\text{bed}} )</td>
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</tr>
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<td>Reactor height</td>
<td>( h_r )</td>
<td>m</td>
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</tr>
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<td>Reactor bottom diameter</td>
<td>( d_b )</td>
<td>m</td>
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</tr>
<tr>
<td>Reactor top diameter</td>
<td>( d_t )</td>
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</tr>
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<td><strong>Bed material (glass beads)</strong></td>
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<td></td>
<td></td>
</tr>
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<td>Overall mass</td>
<td>( M_{\text{bed}} )</td>
<td>kg</td>
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</tr>
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<td><strong>Nozzle related properties</strong></td>
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<td>Nozzle tip height</td>
<td>( h_{\text{noz}} )</td>
<td>m</td>
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</tr>
<tr>
<td>Liquid orifice diameter</td>
<td>( d_{or} )</td>
<td>m</td>
<td>( 8 \times 10^{-4} )</td>
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Table 4.2. Modified model and operational parameters of the simulation (continued)

<table>
<thead>
<tr>
<th>Variable</th>
<th>Symbol</th>
<th>Unit</th>
<th>Value</th>
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<td>Coating solution properties</td>
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<td>Spraying rate</td>
<td>( \dot{M}_{\text{sol}} )</td>
<td>kg s(^{-1})</td>
<td>(6.05 \times 10^{-5})</td>
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<tr>
<td>Dry matter content</td>
<td>(DM_{\text{sol}})</td>
<td>kg kg(^{-1})</td>
<td>0</td>
</tr>
<tr>
<td>Other parameters</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Reactor wall thickness</td>
<td>(d_{w})</td>
<td>m</td>
<td>0.005</td>
</tr>
<tr>
<td>Reactor wall thermal conductivity</td>
<td>(\lambda_{w})</td>
<td>W m(^{-1})K(^{-1})</td>
<td>50</td>
</tr>
<tr>
<td>Reactor wall specific heat</td>
<td>(C_{p,w})</td>
<td>J kg(^{-1})K(^{-1})</td>
<td>452</td>
</tr>
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<td>Reactor wall density</td>
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<td>kg m(^{-3})</td>
<td>7850</td>
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<tr>
<td>Simulation (main model) parameters</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Simulation time step</td>
<td>(\Delta t_{\text{sim}})</td>
<td>s</td>
<td>0.0005</td>
</tr>
</tbody>
</table>

4.5.3. Results and discussion

In the experimental model validation, the following process variables were changed: the inlet air temperature, \(T_{a,\text{in}}\), the spraying rate, \(\dot{M}_{\text{sol}}\), and the mass of core material in the fluidised bed, \(M_{\text{bed}}\). Table 4.3 summarises the different experiments performed along with their process conditions. The fluidisation air flow rate, \(G_{a,\text{in}}\), remained constant for each value of the batch size, \(M_{\text{bed}}\). The appropriate air flow rate was chosen by visually monitoring the fluidised bed (increasing \(G_{a,\text{in}}\) until the onset of bubbling fluidisation). The onset of fluidisation was also clearly visible on the turbine frequency versus air flow charts, of which an example (for \(M_{\text{bed}} = 2\) kg, \(d_{p} = 365\) µm) is presented in Figure 4.28. As explained in Chapter 1, the pressure gradient across the bed, \(\Delta p_{\text{bed}}\), is directly proportional to the gas flow rate through the fixed bed. This corresponds to the region with the lowest slope on the \(f_{\text{bu}}/v_{a,\text{in}}\)-curve. To illustrate this, the air flow rate at the suction side of the centrifugal fan was also measured and plotted in Figure 4.28. The discrepancy between the two flow rates is due to losses occurring over the centrifugal fan. In the first part of the \(f_{\text{bu}}/v_{a,\text{in}}\)-curve, the fan air loss increases linearly, as the pressure difference across the static bed that the fan has to overcome also increases linearly with the inlet air flow rate \(v_{a,\text{in}}\).

At the onset of fluidisation, the pressure difference across the bed remains more or less constant. The constant \(\Delta p_{\text{bed}}\) results in significantly higher air flow rates, as observed by the steeper second part of the \(f_{\text{bu}}/v_{a,\text{in}}\) curve. During fluidisation, the air losses across the centrifugal fan were constant, as can be seen by both flow rate curves which are more or less parallel to each other.
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Figure 4.28. Linear air flow rate, measured at the suction side of the fan (●), and between the fan and the heating coils (■) as a function of the AC frequency supplied to the electric motor of the centrifugal fan.

The combined results of the axial temperature profiles, predicted by the droplet phase extended model and measured experimentally using a moving thermocouple probe (using Eq. (4.40) to transform two-dimensional spatially distributed data into one-dimensional axial profiles) are presented in Figure 4.29 for experiments 1 to 3, Figure 4.30 for experiments 4 to 6 and Figure 4.31 for experiments 7 to 9 – being all experiments with a batch size of $M_{\text{bed}} = 3$ kg.

From Figure 4.29, it can be seen that there is satisfactory agreement between the modelled and measured axial temperature profiles, although the model predicted slightly higher air temperatures near the air distribution plate. There was a good agreement between the modelled and experimental outlet air temperature ($T_{a,\text{out}} = T_{a,n}$) in all experiments. When using a higher inlet air temperature (Figures 4.31 and 4.32), the model-predicted more uniform temperature gradients compared to the measured axial temperature gradients.
Table 4.3. Process conditions in the thermodynamic validation experiments.

<table>
<thead>
<tr>
<th>No.</th>
<th>$T_{\text{a,in}}$ (°C)</th>
<th>$M_{\text{sol}}$ (g min$^{-1}$)</th>
<th>$P_{\text{at}}$ (bar)</th>
<th>$G_{\text{a,in}}$ (kg s$^{-1}$)</th>
<th>$M_{\text{bed}}$ (kg)</th>
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<tr>
<td>1</td>
<td>50</td>
<td>3.63</td>
<td>1.5</td>
<td>$5.57 \times 10^{-2}$</td>
<td>3.0</td>
</tr>
<tr>
<td>2</td>
<td>50</td>
<td>5.24</td>
<td>1.5</td>
<td>$5.57 \times 10^{-2}$</td>
<td>3.0</td>
</tr>
<tr>
<td>3</td>
<td>50</td>
<td>6.85</td>
<td>1.5</td>
<td>$5.57 \times 10^{-2}$</td>
<td>3.0</td>
</tr>
<tr>
<td>4</td>
<td>60</td>
<td>3.63</td>
<td>1.5</td>
<td>$5.57 \times 10^{-2}$</td>
<td>3.0</td>
</tr>
<tr>
<td>5</td>
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<td>$5.57 \times 10^{-2}$</td>
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</tr>
<tr>
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<tr>
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<tr>
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<tr>
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<td>1.5</td>
<td>$2.14 \times 10^{-2}$</td>
<td>2.0</td>
</tr>
<tr>
<td>19</td>
<td>60</td>
<td>10.07</td>
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<td>$2.14 \times 10^{-2}$</td>
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</tr>
<tr>
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<td>$2.14 \times 10^{-2}$</td>
<td>2.0</td>
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<td>1.5</td>
<td>$2.14 \times 10^{-2}$</td>
<td>2.0</td>
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<tr>
<td>23</td>
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<td>1.5</td>
<td>$2.14 \times 10^{-2}$</td>
<td>2.0</td>
</tr>
<tr>
<td>24</td>
<td>70</td>
<td>10.07</td>
<td>1.5</td>
<td>$2.14 \times 10^{-2}$</td>
<td>2.0</td>
</tr>
</tbody>
</table>

It is difficult to pinpoint the shortcomings of the model on the one hand, or of the measurement method on the other, that cause the deviation between the two different profiles. Considering the model, difficulties arise in the use of the particle exchange rate variable, $r_i$, which is not uniform for fluidised beds having a conical geometry and the simple correlations, set out in Section 3.6.2.2, could be insufficient to adequately characterise the mixing behaviour inside the fluidised bed. The effect of the particle exchange rate variable on the bed’s axial temperature profile has already been illustrated in the sensitivity analysis in Section 3.7.3.8. Finally, as the model is one-dimensional, it totally neglects radial temperature or humidity gradients as well as radial particle transport. As will be demonstrated in Chapter 5, the radial temperature and humidity gradients in the vicinity of the nozzle could even be more pronounced than the axial gradients.
Figure 4.29. Measured versus simulated steady state axial air temperature profile of a water sprayed fluidised bed of 3.0 kg glass beads with $T_{a, in} = 50$ °C and with varying spraying rates.

Figure 4.30. Measured versus simulated steady state axial air temperature profile of a water sprayed fluidised bed of 3.0 kg glass beads with $T_{a, in} = 60$ °C and with varying spraying rates.
Figure 4.31. Measured versus simulated steady state axial air temperature profile of a water sprayed fluidised bed of 3.0 kg glass beads with $T_{a,in} = 70 \, ^\circ C$ and with varying spraying rates.

However, not only the model could be solely responsible for the deviation, also the measurement method has some problems associated with it. First, it is difficult to actually measure the air temperature, $T_a$. As particles continuously impinge on the probe, they transport part of their heat towards the probe, thus the measured temperature will lie between $T_a$ and $T_p$. To reduce this effect to a certain extent, plastic materials (such as PVC or nylon, which have much lower thermal conductivity than aluminium) were selected for probe tip construction. Insulating the probe tip could even more reduce the effect of $T_p$ in air temperature measurement, but would increase the volume of the probe, thus causing larger disturbances in the bed’s air flow. Another problem is that the probe surface is wetted when it nears the nozzle and as a result, dry bulb temperature measurements are no longer assured. Also, particles could stick to the wetted probe surface. Therefore, a minimum safe distance of 0.05 m from the nozzle was respected in the probe’s scanning pattern.

Similar conclusions could be drawn when the experiments using a batch size of $M_{\text{bed}} = 2.0 \, \text{kg}$ are studied, being experiments 10-24 in Table 4.3. At low inlet air temperatures and low spraying rates, there is a good agreement between the measured and modelled axial air temperature profiles, the exception being the temperatures just above the air distributor. When considering higher inlet air temperatures or higher spraying rates, predicting or comparing the air temperature profile, especially its non-linear nature, becomes increasingly difficult. Nonetheless, there was a good agreement between the modelled and experimental outlet air temperature ($T_{a,out} = T_{a,n}$).
Figure 4.32. Measured versus simulated steady state axial air temperature profiles of a water sprayed fluidised bed of 2.0 kg glass beads with $T_{a,in} = 50 \, ^\circ C$ and with varying spraying rates.

Figure 4.33. Measured versus simulated steady state axial air temperature profiles of a water sprayed fluidised bed of 2.0 kg glass beads with $T_{a,in} = 60 \, ^\circ C$ and with varying spraying rates.
Figure 4.34. Measured versus simulated steady state axial air temperature profiles of a water sprayed fluidised bed of 2.0 kg glass beads with $T_{a,in} = 70 \, ^\circ C$ and with varying spraying rates.

4.5.4. Conclusions

Simulation of the thermodynamic properties of the fluidised bed during coating operations by means of the droplet phase extended model, as detailed in this chapter, was quite reliable. The model had no difficulties in predicting the exhaust air properties, but had some problems with predicting the precise axial thermodynamic profiles. The likely causes are: one-dimensionality of the model, the characterisation of the particle exchange rate, $r_i$, and experimental difficulties including particle and droplet interference in the air temperature measurements.

Besides thermodynamic validation, the model was also verified in its ability to predict the spray drying losses occurring during fluidised bed coating. This second part of the validation study will be thoroughly discussed in Section 4.6.
4.6. Model validation: coating efficiency

4.6.1. Introduction

The first part of this chapter dealt with the integration of the different interactions between the droplet phase on the one hand and the gas/solid phases on the other hand into the model, described in Chapter 3. The extended model was validated using spatial thermodynamic measurements within the liquid-sprayed fluidised bed. However, one of the strengths of the model proposed in this chapter is its ability to predict the extent to which droplets (completely) evaporate before impinging on the surface of the fluidised particles.

In this section, the droplet phase extended model will be validated in its ability to predict spray drying losses by comparing the results with experimental coating data of a Glatt GPCG-1, generated by Dewettinck (1997).

4.6.2. Materials and methods

4.6.2.1. Determination of the coating efficiency

For the determination of the coating mass and coating efficiency, $\eta_c$, the experimental data by Dewettinck (1997) were used. In this research work, the core material was 1 kg of NaCl crystals which were coated with a 5 w% sodium caseinate solution. The adhered sodium caseinate, which is a protein, was then quantitatively determined using the Lowry method. The coating efficiency was subsequently calculated as the amount of protein retrieved on the particles, compared to the total amount of coating material injected into the bed throughout the batch coating process.

All coating experiments were performed in the Glatt GPCG-1 fluidised bed unit using the top-spray insert with the nozzle in the upper position ($h_{noz} = 0.225$ m). In each coating experiment, the spraying rate and the inlet air flow rate were kept constant ($\dot{M}_{sol} = 7$ g min$^{-1}$, $v_{a,in} = 3$ m s$^{-1}$ corresponding to $G_{a,in} = 1.34 \times 10^{-2}$ kg s$^{-1}$), while the particle diameter, $d_p$, atomisation air pressure, $P_{at}$, and the inlet air temperature, $T_{a,in}$, were varied between different experiments as will be shown further on in Tables 4.4 and 4.5. Each coating experiment was terminated when the total amount of coating solution introduced into the bed was equal to 0.5 kg. After finishing the coating process, coating efficiency was determined by means of the Lowry-method.

Also, during each experiment, the steady-state bed temperature, $T_{bed}$, was recorded. The bed temperature was measured by means of a shielded T-type thermocouple suspended in the fluidised bed approximately $h_{probe} = 0.06$ m above the air distributor. As this probe was freely suspended into the bed, its measured temperature was a combination of both the temperature...
of the fluidisation air passing the probe tip, and of the temperature of the fluidised particles impinging on the probe. To compare the measured with the model-predicted bed temperature, the model-predicted bed temperature – based on the model-predicted particle and gas phase temperatures in the vicinity of the nozzle – was calculated using the following equation:

\[
T_{\text{bed}} = e_{\text{bed}} T_{a,A(h_{\text{probe})}} + (1 - e_{\text{bed}}) T_{p,A(h_{\text{probe})}}
\]

In Eq. (4.41) is \( T_{p,A(h_{\text{probe})}} \) the average temperature of the particles and \( T_{a,A(h_{\text{probe})}} \) the air temperature, both within the control volume containing the pneumatic nozzle.

### 4.6.2.2. Determination of the spraying jet shape factor

As already highlighted in Section 4.3.5, one of the remaining unknown process variables is the atomisation gas jet shape factor, \( s_{\text{at}} \). This shape factor determines the aperture of the parabolic jet formed by the release of compressed air into the reactor chamber, as defined by Eqs. (4.38) and (4.39). As stated in Eqs. (4.26) and (4.27), the jet boundary (or jet radius) is defined by the location where the radial gas velocity is equal to zero. The radial gas velocity for any position within this jet boundary always carries a positive sign (i.e. radial gas flow is directed outwards from the centre), while outside the jet boundary, the radial gas velocity has a negative sign (i.e. radial gas flow is directed towards the jet centre), or

\[
\begin{align*}
\text{at,rd} (h', x) &> 0 & x < s_{\text{at}} h'^{1/3} \\
\text{at,rd} (h', x) &\leq 0 & x \geq s_{\text{at}} h'^{1/3}
\end{align*}
\]

Because of the drag force reversal occurring at the jet boundary, droplets travelling through the gas jet produced by the atomisation air generally remain within the boundaries of this gas jet. Consequently, the shape of the gas jet and the spray jet could be considered to be the same and the behaviour of the gas (compressed air) jet could be easily analysed by means of the spraying pattern.

As a rough approximation of the spatial distribution of the spray and to determine the value of the shape factor, \( s_{\text{at}} \), a simple method, similar to Wauters et al. (2002), was used. This method consists of spraying over an array of \( 20 \times 20 \) cuvettes – which are normally used for chemical analysis by light adsorption. Each cuvette had a width of \( 12.5 \times 12.5 \) mm and an opening of \( 10.0 \times 10.0 \) mm. The pneumatic nozzle (Schlick 970 S1) was suspended 0.2 m above the centre of the cuvette array and the spraying solution used was distilled water. The amount of collected liquid was measured afterwards by weighing the cuvettes by means of an electronic balance (Mettler, PE-6000, Belgium). Spray deposition was expressed by means of the spray flux, in the amount of liquid (kg) collected per time unit (s), and per surface area (m\(^2\)).
This method to characterise the spray flux has some disadvantages including low spatial resolution (i.e. the size of the cuvettes) and reduced recovery yields of the spray onto the collector array. Possible yield-reducing causes are evaporation and deflection of the droplet trajectories near the collector array (obstacle).

4.6.2.3. Calculations and simulation method

An overview of the model and process variables, constant in each coating process simulation, is given in Table 4.4, while the varied parameters and variables are given in Table 4.5. Considering the input variables and parameters, the following remarks should be taken into account:

- As the influence of the particle diameter was studied, some fluidisation characteristics were needed to be reassessed. More specifically, the particle exchange rate, \( r_i \), using Eq. (3.52) and the bed height (\( h_{\text{bed}} \)), the bed voidage at minimum fluidisation (\( \varepsilon_{\text{mf}} \)) the bubble frequency (\( f_{\text{bu}} \)) were recalculated using the equations in Section 1.4.4, the results are shown in Table 4.5.

- Salt crystals are shaped cubically, consequently their sphericity is lower than 1. The sphericity, \( \psi_p \), which is the degree to which the shape of a particle approaches that of a perfect sphere, is calculated as (Perry, 1984; Bayram, 2004):

\[
\psi_p = \frac{6V_p}{A_p d'_p} \quad (4.44)
\]

Where \( V_p \) is the volume of the particle, \( A_p \) its surface and \( d'_p \) the equivalent diameter, being the diameter of a sphere with equal volume. For a cube, Eq. (4.44) yields:

\[
\psi_p = \left( \frac{\pi}{6} \right)^{\frac{1}{3}} = 0.806 \quad (4.45)
\]

- Viscosity and surface tension of a 5 w% solution of sodium caseinate were based on measurements made by Dewettinck (1997). These values were taken at 20°C because the droplet drying process is usually adiabatic. Previous droplet submodel simulation results have shown the droplet temperature to range between 18 to 25 °C when using reference process conditions (cfr. Figure 4.13).

- To reduce calculation time, the heating phase was no longer simulated. Instead, the following initial conditions were assumed:

\[
\forall i \in [1 \ldots n]: T_{\text{a,i}}(0) = T_p(0) = T_{\text{a,in}} \quad (4.46)
\]
A process time of $t_{\text{sim}} = 900$ s was found to be sufficiently long to reach steady state conditions using the initial conditions in Eq. (4.46).

**Table 4.4.** Constant model and operational variables of the simulation.

<table>
<thead>
<tr>
<th><strong>Variable</strong></th>
<th><strong>Symbol</strong></th>
<th><strong>Unit</strong></th>
<th><strong>Value</strong></th>
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<tr>
<td><strong>Main model parameters</strong></td>
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<tr>
<td>Coating control volumes</td>
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<td>5000</td>
</tr>
<tr>
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<td></td>
<td></td>
<td>Levefbre (1988)</td>
</tr>
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<td><strong>Fluidisation air properties</strong></td>
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<tr>
<td>Inlet air volumetric flow rate</td>
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<td>Reactor top diameter</td>
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<td>Specific heat</td>
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*Modelling heat and mass transfer in fluidised bed coating processes*
Table 4.4. Constant model and operational variables of the simulation (continued).

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<th>Variable</th>
<th>Symbol</th>
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<td><strong>External air (atmospheric) conditions</strong></td>
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<tr>
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</tr>
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<td><strong>Simulation (droplet submodel) parameters</strong></td>
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<td>Maximum simulation time (single trajectory)</td>
<td>$t_{dr,sim}$</td>
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<td>Minimum tolerable droplet temperature difference</td>
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### Table 4.5. Studied process variables.

<table>
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<th>Values studied</th>
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<tr>
<td>Particle diameter</td>
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</tr>
<tr>
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<td>$h_{bed}$</td>
<td>m</td>
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</tr>
<tr>
<td>Particle exchange rate</td>
<td>$r_i$</td>
<td>Hz</td>
<td>2.48 – 2.34 – 2.14</td>
</tr>
<tr>
<td>Bed voidage at minimum fluidisation velocity</td>
<td>$\epsilon_{mf}$</td>
<td>–</td>
<td>0.48 – 0.46 – 0.45</td>
</tr>
<tr>
<td>Bubble frequency</td>
<td>$f_{bu}$</td>
<td>Hz</td>
<td>4185 – 4825 – 5780</td>
</tr>
<tr>
<td>Inlet air temperature</td>
<td>$T_{a, in}$</td>
<td>°C</td>
<td>70 – 78 – 86</td>
</tr>
<tr>
<td>Atomisation air pressure</td>
<td>$P_{at}$</td>
<td>bar</td>
<td>1.5 – 2.5 – 3.5</td>
</tr>
<tr>
<td>Atomisation air mass flow rate</td>
<td>$G_{at}$</td>
<td>kg s$^{-1}$</td>
<td>1.45×10$^{-3}$ – 2.04×10$^{-3}$ – 2.38×10$^{-3}$</td>
</tr>
<tr>
<td>Shape factor of atomisation air jet$^*$</td>
<td>$s_{at}$</td>
<td>m$^{2/3}$</td>
<td>0.0467 – 0.0446 – 0.0426</td>
</tr>
</tbody>
</table>

*Remarks:*

$^*$The experimental determination of $s_{at}$ is given in Section 4.6.3.1

### 4.6.3. Results and discussion

#### 4.6.3.1. Spray characterisation and shape factor determination

In Figure 4.35, an example of the spray flux in a plane perpendicular to nozzle’s central symmetry axis and at a distance $h' = 0.20$ m from the nozzle, is shown. This graph shows how the spray flux varies with the radial position underneath the nozzle. As expected, the spray flux was axisymmetric and had the highest fluxes in the centre, i.e. directly under the nozzle tip.

To determine the shape factor, $s_{at}$, a projected (cross-sectional) circular region with its centre on the axial nozzle axis was defined in which 90 % of all sprayed liquid was collected, which is illustrated in Figure 4.36. Once the diameter, $d_{at}(h')$ has been retrieved, the shape factor is then calculated as:

$$s_{at} = \frac{d_{at}(h')}{h'^{2/3}} \quad (4.47)$$

As different atomisation air pressures were applied in the experimental determination of the coating efficiency, the spray produced by the pneumatic nozzle was characterised when its air pressure was varied. A series of experiments was carried out with $P_{at}$ varying between 1.0 and 3.0 bar, using a constant spraying rate of 8.46 g min$^{-1}$. The result of these experiments is shown in Figure 4.36 for the shape factor as determined using Eq. (4.47).
Figure 4.35. Measured spray fluxes, in g m$^{-2}$ s$^{-1}$, registered on a vertical plane 0.20 m below the spraying nozzle. Distilled water was used as spraying with a spraying rate of $M_{\text{sol}} = 8.46$ g min$^{-1}$ and at a pressure of $P_{\text{at}} = 1.5$ bar.

Figure 4.36. The determination of cross-sectional spray surface at position $h'$ for the calculation of the spray jet shape factor, for a spraying rate of $M_{\text{sol}} = 8.46$ g min$^{-1}$ and a pressure of $P_{\text{at}} = 1.5$ bar.
In Figure 4.37, there is a strong linear relationship ($R^2 = 0.918$) between the shape factor and the atomisation air pressure within the range of conditions examined. Higher atomisation air pressures will result in slightly more narrow spray jets and vice versa. As the coating experiments, performed by Dewettinck (1997), were based on varying atomisation pressures, the linear correlation – as shown in Figure 4.37 – was used to calculate the spray jet shape factor, which is a prerequisite in the droplet submodel.

To compare, in a study by Juslin et al. (1995a, b), the cone angle was measured using image analysis of the water spray pattern produced by a Schlick pneumatic nozzle, type 940.S7 but using a liquid orifice having a diameter, $d_{or} = 1.2 \times 10^{-3}$ m. Depending on the liquid feed rate and the atomisation air pressure, the cross-sectional diameter of the spray at a distance of 0.15 m of the nozzle ranged between 0.041 m and 0.064 m. Using Eq. (4.47), these results correspond to a shape factor, $s_{at}$, ranging between 0.077 m$^{2/3}$ and 0.120 m$^{2/3}$.

4.6.3.2. Bed thermodynamics validation

The measured bed temperature during steady state coating regime, $T_{bed,\text{exp}}$, was compared with the model-predicted bed temperature, $T_{bed,\text{sim}}$. The results are shown in Figure 4.38, while the regression analysis results are given in Table 4.6. The model-predicted bed temperature appeared to be significantly influenced by the particle diameter, especially at higher inlet air temperatures, as shown by the vertical spreading of $T_{bed,\text{sim}}$-values; an effect which could be explained by the calculation of the bed temperature from the simulation output data as given in Eq. (4.41).
From the results of the regression analysis, it could be deduced that a good correlation was attained between the experiment and the model, although the model tended to overestimate the bed temperature as can be seen by the asymmetric 95%-confidence interval of the intercept, $b$. Previous thermodynamic validation in Section 4.5 revealed that the model tended to underestimate the air temperature – and ultimately, bed temperature – in the lower sections of the fluidised bed, as the temperature profiles had a more non-linear character compared to the nearly linear model-predicted axial air temperature gradients. This observation is in apparent contradiction with the results presented in Figure 4.38. However, the method of measuring the bed temperature, $T_{\text{bed}}$, by means of a shielded probe suspended in the bed is likely to result in lower temperatures due to evaporative cooling. Indeed, visual inspection of the temperature probe after conducting coating experiments using coloured coating solutions often revealed a clearly distinguishable layer of coating material sticking on the probe. This indicates that part of the coating material somehow must have contacted the probe in a dissolved state, either by direct droplet contact or by collision of wetted particles resulting in evaporative cooling of the probe tip.

![Figure 4.38](image_url)

**Figure 4.38.** Measured bed temperature, $T_{\text{bed,exp}}$, during steady state coating versus the model-predicted bed temperature, $T_{\text{bed,sim}}$.

**Table 4.6.**
Regression analysis between model-predicted and experimental bed temperature, including the sum of squared residuals and the 95%-confidence intervals.

<table>
<thead>
<tr>
<th>$T_{\text{bed,exp}} = a T_{\text{bed,sim}} + b$</th>
<th>$R^2$</th>
<th>$SSR$</th>
<th>Slope, $a$</th>
<th>95% confidence interval of slope</th>
<th>Intercept, $b$</th>
<th>95% confidence interval of intercept</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.972</td>
<td>164.5</td>
<td>1.160</td>
<td>[1.065; 1.257]</td>
<td>-4.564</td>
<td>[-8.814; -0.313]</td>
</tr>
</tbody>
</table>
4.6.3.3. Validation of the coating efficiency

The experimental spray drying losses were compared with the model-predicted spray drying losses and the results are graphically represented in Figure 4.39, while the regression analysis is summarised in Table 4.7. From these results it can be concluded that modest correlation was achieved, given the complex thermodynamic nature (multivariateness) of the spray drying effect. As can be seen from the regression analysis, the model tended to underestimate the spray drying loss \(a\), slope = 0.769), but this could be due to assumption,

\[
\left[ \frac{\dot{M}_{sd}}{DM_{sol}M_{sol, exp}} \right] = (1 - \eta_c)
\]  

(4.48)

In other words, any loss in coating material throughout the process, \(1 - \eta_c\), was assumed to be solely the result of the spray drying of the coating solution. In reality, however, effects such as attrition of deposited coating material and subsequent entrainment of coating material in the filter will contribute in decreasing the overall coating efficiency and thus, the experimental spray drying losses may be well below \(1 - \eta_c\), possibly explaining the underestimation in the model.

Figure 4.39. The measured spray drying loss, \(1 - \eta_{c,exp}\), during steady state coating versus the model-predicted spray drying loss.
Table 4.7.
Regression analysis between model-predicted and experimental spray drying loss, including the sum of squared residuals and the 95%-confidence intervals.

\[
\frac{\dot{M}_{sd}}{\dot{M}_{sol}M_{sol,exp}} = a \left( \frac{\dot{M}_{sd}}{\dot{M}_{sol}M_{sol,sim}} \right) + b \quad \text{assuming} \quad \frac{\dot{M}_{sd}}{\dot{M}_{sol}M_{sol,exp}} = (1 - \eta_c)
\]

<table>
<thead>
<tr>
<th>$R^2$</th>
<th>$SSR$</th>
<th>Slope, $a$</th>
<th>95% confidence interval of slope</th>
<th>Intercept, $b$</th>
<th>95% confidence interval of intercept</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.701</td>
<td>0.058</td>
<td>0.769</td>
<td>[0.528; 1.010]</td>
<td>0.096</td>
<td>[0.017; 0.175]</td>
</tr>
</tbody>
</table>

4.6.4. Conclusions

Although the model was shown to be quite reliable in predicting the overall bed thermodynamics, it has proven to be capable of roughly estimating the spray drying losses. An additional difficulty in this kind of validation experiments is that not the spray drying in se is measured, but the overall process efficiency. The latter is also affected by attrition and entrainment; effects which are still not implemented into the model. The obvious result is an underestimation of the model-predicted spray drying losses.
4.7. Modelling results and discussion

4.7.1. Introduction

In this section, the model-predicted results are discussed in more detail. More specifically, the effect of extending the basic model (Chapter 3) with the droplet phase and its discrete, stochastic solution method (this chapter), on the model-predicted results will be studied in detail. Finally, a sensitivity analysis of the process variable affecting the droplet phase characteristics will be carried out.

4.7.2. Methods and calculation

4.7.2.1. Reference case simulation and model comparison

To compare the results of both models, both reference cases from the sensitivity analysis were used. The model variables and parameters to simulate the reference case were given in Table 3.1, for the basic model and in Table 4.1 for the droplet phase extended model. The unknown variables such as bed height, relative size of the coating region \((c/n)\), only for the basic model) and average particle exchange rates, \(r_i\), are calculated using the equations highlighted in Section 3.6.2.2.

Each reference case simulation consisted of a coating process of inert glass spheres which was simulated from start-up to stationary conditions using the reactor and bed dimensions of the Glatt GPCG-1 fluidised bed unit. Two distinct phases were distinguished in the dynamic simulation. Initially, no coating was spraying until steady state conditions were reached (heating phase). This stage’s duration was approximately 500 seconds. Next, a step input was given for the spraying rate (here \(M_{\text{sol}} = 10 \text{ g min}^{-1}\)). The simulation was continued until steady state conditions were reached again.

4.7.2.2. Sensitivity analysis

As in Chapter 3, the translation factor \((1/K_n)\), as expressed in Eq. (3.55) was used to evaluate the sensitivity of the different model-predicted output variables to changes in the input variables (i.e. process variables, material properties). However, in this chapter, the sensitivity analysis was limited to the output variables that purely relate to the droplet phase and its generation by means of a pneumatic nozzle. Table 4.8 summarises all the input variables to which the sensitivity analysis was applied in this chapter. As shown in Table 4.8, variables such fluidisation air flow rate, \(G_{a,\text{in}}\) (or \(v_{a,\text{in}}\)) were also included, because the fluidisation air flow rate also affects the bed voidage, \(\varepsilon_{\text{bed}}\), and thus potentially influences the droplet penetration into the fluidised bed.
Droplet deposition and migration behaviour in liquid-sprayed fluidised beds

Table 4.8. The different input variables assessed in the sensitivity analysis.

<table>
<thead>
<tr>
<th>Variable or parameter</th>
<th>Symbol</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Fluidisation air properties</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Linear fluidisation air velocity (inlet duct)*</td>
<td>$v_{a,\text{in}}$</td>
<td>m s$^{-1}$</td>
</tr>
<tr>
<td>Inlet air temperature</td>
<td>$T_{a,\text{in}}$</td>
<td>K</td>
</tr>
<tr>
<td><strong>Atomisation air properties</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Atomisation air pressure**</td>
<td>$p_{at}$</td>
<td>bar</td>
</tr>
<tr>
<td>Atomisation air relative humidity</td>
<td>$\phi_{at}$</td>
<td>-</td>
</tr>
<tr>
<td>Atomisation air temperature</td>
<td>$T_{at}$</td>
<td>K</td>
</tr>
<tr>
<td><strong>Spraying solution properties</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Spraying rate</td>
<td>$M_{\text{sol}}$</td>
<td>g min$^{-1}$</td>
</tr>
<tr>
<td>Spraying solution temperature</td>
<td>$T_{\text{sol}}$</td>
<td>K</td>
</tr>
<tr>
<td>Spraying solution viscosity</td>
<td>$\mu_{\text{sol}}$</td>
<td>Pa s</td>
</tr>
<tr>
<td>Spraying solution surface tension</td>
<td>$\gamma_{\text{sol}}$</td>
<td>N m$^{-3}$</td>
</tr>
<tr>
<td><strong>Nozzle properties</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nozzle height</td>
<td>$h_{\text{noz}}$</td>
<td>m</td>
</tr>
<tr>
<td>Nozzle liquid orifice diameter</td>
<td>$d_{or}$</td>
<td>m</td>
</tr>
<tr>
<td><strong>Bed material properties</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Particle diameter</td>
<td>$d_p$</td>
<td>$\mu$m</td>
</tr>
<tr>
<td>Particle exchange rate</td>
<td>$r_i$</td>
<td>Hz</td>
</tr>
</tbody>
</table>

Remarks
* The linear air velocity at the inlet duct corresponds to the air velocity as measured by the pitot probe in the Glatt GPCG-1 (see Figure 3.30). The recalculate into volumetric air flow rate, the linear air flow rate has to be multiplied with the duct cross-sectional area, being $3.744 \times 10^{-3}$ m$^2$.
** The atomisation pressure is recalculated to the atomisation air mass flow rate using Eqs. (3.49) and (3.50).

In addition to the output variables as used in Chapter 3’s validation study, the following outputs were used:

- The overall spray drying loss, $\dot{M}_{\text{sd}}$, calculated in the model as $\sum_{i=1}^{n} \dot{M}_{\text{sd}_i}$.

As the model was validated in its capability to predict spray drying losses in Section 4.6, sensitivity analysis could help in evaluating the effects of varying process conditions on the degree of spray drying in order to assess an optimal strategy.
The droplet property distributions at the moment of particle/droplet impact. More specifically, $T_{\text{dec}}, D_{\text{M,dec}}$ and $h_{\text{dec}}$ were studied as these variables could have a significant impact on the droplet spreading characteristics on the core particle’s surface and ultimately, alter the quality of the coating layer. For instance, Dewettinck (1997) conjectured that coating solution’s glass transition temperature – which depends on the dry matter concentration – plays a key role in determining the coating quality through affecting the coating solution’s rheology and stickiness at the moment the solution is deposited on the particle’s surface until it has been entirely evaporated. The lower the glass transition temperature, the less likely the coating solution will spread evenly, resulting in ‘orange peel’-like coatings. However, linking the droplet properties to the quality of the coating layer and ultimately, its performance in real food matrices is beyond the scope of this research work.

4.7.3. Results and discussion

4.7.3.1. Reference case simulation and model results comparison

The axial air temperature and relative humidity profiles as predicted by both models are presented in Figure 4.40. Although the exhaust air temperature, $T_{a,\text{out}} = T_{a,n}$, and humidity, $\varphi_{a,\text{out}} = \varphi_{a,n}$, are almost equal for both models, their profiles are considerably different: the droplet phase extended model tends to predict lower air temperatures and higher air humidities. Because there is no pre-set volume in which the coating solutions deposition takes place, as in the basic model ($c/n = 3/24$), there is no abrupt change in air humidity in the upper part of the fluidised bed. In fact, the volume of the bed in which droplets penetrated (the coating volume) was predicted – by means of the submodel – to be significantly higher than 12.5 % of the total bed as assumed in the basic model. More specifically, a value of 37.5 % of the bed volume was occupied by the droplet phase. This larger coating volume is also the explanation for the lower air temperatures and higher air humidities. Increasing the $c/n$-ratio to 37.5 % in the basic model will result in axial thermodynamic profiles being more comparable to those of the extended model, but in general, the $c/n$-ratio is unknown prior to the simulation. The prediction of the degree of droplet penetration into the fluidised bed implemented in the extended model is one of the major benefits of this model over the basic model.

Similarly, the axial particle temperature and moisture content profiles during steady state have been plotted in Figure 4.41 for $\bar{T}_p$ and $\bar{W}_p$, and in Figure 4.42 for $s(T_p)$ and $s(W_p)$. As the particle temperature is closely related to the fluidum temperature in which it resides, the same conclusions could be drawn as for the air temperatures: including the droplet phase into the model resulted in the prediction of significantly lower particle temperatures (approx. 4 °C).
Figure 4.40. Axial air temperature and relative humidity profiles during steady state coating conditions predicted by the droplet phase extended model ($\overline{T}_{a,l}$: ●, $\overline{\phi}_{a,l}$: ○), compared to the basic model ($\overline{T}_{a,l}$: ○, $\overline{\phi}_{a,l}$: ●).

Figure 4.41. Average axial particle temperature and moisture content profiles during steady state coating conditions predicted by the droplet phase extended model ($\overline{T}_{p}$: ●, $\overline{W}_{p}$: ○), compared to the basic model ($\overline{T}_{p}$: ○, $\overline{W}_{p}$: ●).
Figure 4.42. Standard deviation axial profiles of the particle temperature and moisture content during steady state coating conditions predicted by the droplet phase extended model ($s(T_p)$: ■, $s(W_p)$: ◇), compared to the basic model ($s(T_p)$: ○, $s(W_p)$: ▽).

Figure 4.43 illustrates the simulated particle temperature distributions. The typical particle temperature distribution consists of a low-temperature peak and a high-temperature peak corresponding to wetted (hence, drying) and fully dried particles, respectively. From these temperature distributions it can be observed that the extended model predicted a significant larger fraction of particles at low temperature, compared to the basic model. The explanation behind this phenomenon is the assumption of non-uniform distribution of coating solution among the particles (i.e. fewer particles receive larger quantities of coating solution) in the extended model, compared to the continuous distribution used in the basic model.

When considering the simulated coating mass distributions after 900 seconds of coating regime (Figure 4.44), the following differences could be noticed: First of all, the average coating mass, was smaller for the droplet phase extended model than the simple model. The difference in average coating mass was equal to the sum of the spray drying losses and wall adhesion, factors which are only included in the droplet phase extended model. For the reference case, the overall spray drying losses were estimated to be around 3.8 w% - which was quite low. The low fraction of spray dried coating solution could be explained by the low position of the spraying nozzle. The wall contact losses were negligible in the reference case.
Figure 4.43. Model-predicted particle temperature distribution, $T_p$ in °C, using the basic model (−) and using the droplet-phase extended model (−).

Figure 4.44. Model-predicted coating mass distribution, $Y_p$ in kg coating kg core$^{-1}$, using the basic model (−) and using the droplet-phase extended model (−).

The coating mass distribution was also narrower for the droplet phase extended model which is in apparent contradiction with the model assumption of discontinuous deposition of coating solution (through droplets) as opposed to the continuous deposition used in the basic model. Basically, in the extended model, two mechanisms potentially affect the coating mass distribution: The frequency of particle migration (mixing) from and towards the coating volume and, the discontinuous deposition of coating solution on the particle surface. When
the model-predicted particle residence time in the coating volume is plotted against the hypothetical number of droplets available to each particle residing in the coating volume (Figure 4.45), the majority of the particles (> 60 %) receive one or more droplets during their passage through the bed’s coating volume. Only a smaller fraction of the particles has less than 100 % chance to receive a droplet during this passage. Thus, the contribution of the discontinuous coating solution deposition to the variance of $Y_p$ is relatively small compared to particle exchange occurring between the coating and the non-coating volumes of the bed.

**Figure 4.45.** Simulated distribution of the particle residence time in the coating volume (●), compared to the hypothetical number of coating solution droplets available to each particle during its passage through the bed’s coating volume (○). To assess the size of the coating volume as predicted by the droplet phase extended model, the relative droplet number concentration, $N_{dp,i}/N_{dp,n}$ versus droplet height in the fluidised bed is plotted in Figure 4.46.

*Modelling heat and mass transfer in fluidised bed coating processes*
In Eqs. (3.38) and (3.39) is $\zeta_{\text{dr}}$ the average droplet collection efficiency for the entire fluidised bed. The results in Figure 4.46 clearly indicate that the decrease in droplet number concentration is not linear as stated in Eqs. (4.49) and (4.50). The reason these linear equations are less suited is because of a large gradient in droplet collection efficiency across the fluidised bed. As the high-velocity droplets rapidly decelerate by the sharp increase in drag force upon penetrating the fluidised bed – as stated by the correlation by Mostoufi and Chaouki (1999) in Eqs. (4.22) and (4.23) – the impingement efficiency, $\chi_{\text{dr}}$, decreases, while the droplet adhesion probability, $\iota_{\text{dr}}$, increases. The net result is a negative droplet collection efficiency gradient across the fluidised bed, whereas the linear equations by Löffler (1988) are based on a single bed-averaged droplet collection efficiency.

The effect of this negative collection efficiency gradient is a reduced droplet deposition rate in the upper layers (just below the nozzle) of the fluidised bed, whereas there will be a distinct region below these upper layers with maximised droplet collection rate. To illustrate, the
simulated dry matter deposition rate $r_{C,i} \times M_{pp,i}$ is plotted against the axial bed position in Figure 4.47. Due to the exhaustion of the downward moving droplet phase and the increasing collection efficiency closer to the nozzle, a region with maximised droplet/particle transfer could be distinguished. The position of this region in the bed depends on a number of process variables, including the atomisation pressure, bed voidage and the viscosity of the coating solution. The effects of these process variables will be discussed into more detail in Section 4.7.3.2.

In analogy with the basic model, as detailed in Chapter 3, the coating volume could be derived. This represents the volume in which both particles and droplets coexist and droplet/particle capture takes place. Based in Figure 4.46 and by defining the coating volume as the volume where 90 % of the total droplet collection takes place, the model-predicted coating volume comprised approximately 0.036 m of the bed below the nozzle, which corresponds to 37.5 % of the total bed volume.

![Figure 4.47. Model-predicted dry matter collection rate at steady state coating regime, $r_{C,i}$, as a function of bed position, $h$ (- -) and the modelled steady state droplet phase/gas phase heat exchange rate in W(- -).](image)

### 4.7.3.2. Sensitivity analysis: effect of process variables

The results of translation factors calculation are given in Table 4.9 for the exhaust air properties and the thermodynamic efficiency, in Table 4.10 for the particle-related model-predicted output variables and in Table 4.11 for the model-predicted droplet phase-related properties. The calculated translation factors from these tables have been represented...
graphically in Figure 4.48. In these graphs, both the translation factors for a negative input variable variation \((\Delta \omega < 0)\) as well as for a positive variation \((\Delta \omega > 0)\) are given.

**Table 4.9.** Translation factors (central difference, calculated as the average from both single-sided differences) for the exhaust air-related properties and the thermodynamic efficiency of the process.

<table>
<thead>
<tr>
<th>Input variable ((\omega))</th>
<th>Output variable ((\Omega))</th>
<th>(\bar{T}_{a,\text{out}})</th>
<th>(\bar{\Phi}_{a,\text{out}})</th>
<th>(\bar{X}_{a,\text{out}})</th>
<th>(\bar{n}_{\text{vap}})</th>
<th>(\eta_{\text{th}})</th>
<th>(\Phi_{\text{loss}})</th>
</tr>
</thead>
<tbody>
<tr>
<td>(v_{\text{a,in}}) (m s(^{-1}))</td>
<td>6.30 \times 10^{-2}</td>
<td>-1.67</td>
<td>-4.87 \times 10^{-1}</td>
<td>-2.56</td>
<td>-0.99</td>
<td>1.57</td>
<td></td>
</tr>
<tr>
<td>(P_{a}) (bar)</td>
<td>-4.10 \times 10^{-3}</td>
<td>2.09 \times 10^{-2}</td>
<td>-4.76 \times 10^{-2}</td>
<td>-2.88 \times 10^{-3}</td>
<td>-2.46 \times 10^{-2}</td>
<td>-2.11 \times 10^{-2}</td>
<td></td>
</tr>
<tr>
<td>(T_{a}) (K)</td>
<td>1.02 \times 10^{-3}</td>
<td>-1.03</td>
<td>6.71 \times 10^{-1}</td>
<td>-8.38 \times 10^{-1}</td>
<td>-6.93 \times 10^{-1}</td>
<td>5.24 \times 10^{1}</td>
<td></td>
</tr>
<tr>
<td>(\varphi_{\text{at}})</td>
<td>8.22 \times 10^{-4}</td>
<td>2.53 \times 10^{-2}</td>
<td>3.97 \times 10^{-2}</td>
<td>5.88 \times 10^{-2}</td>
<td>-1.55 \times 10^{-2}</td>
<td>5.89 \times 10^{2}</td>
<td></td>
</tr>
<tr>
<td>(\dot{M}_{\text{sol}}) (g min(^{-1}))</td>
<td>-5.80 \times 10^{-2}</td>
<td>1.43</td>
<td>5.49 \times 10^{-1}</td>
<td>2.20</td>
<td>1.01</td>
<td>1.49 \times 10^{1}</td>
<td></td>
</tr>
<tr>
<td>(T_{\text{sol}}) (K)</td>
<td>7.11 \times 10^{-3}</td>
<td>-1.16 \times 10^{-1}</td>
<td>4.64 \times 10^{-4}</td>
<td>-1.45 \times 10^{-1}</td>
<td>-1.33 \times 10^{-1}</td>
<td>3.76 \times 10^{1}</td>
<td></td>
</tr>
<tr>
<td>(\mu_{\text{sol}})</td>
<td>4.03 \times 10^{-4}</td>
<td>5.16 \times 10^{-3}</td>
<td>6.86 \times 10^{-4}</td>
<td>6.10 \times 10^{-3}</td>
<td>-1.27 \times 10^{-4}</td>
<td>-2.27 \times 10^{1}</td>
<td></td>
</tr>
<tr>
<td>(\gamma_{\text{sol}})</td>
<td>4.13 \times 10^{-3}</td>
<td>-6.79 \times 10^{-2}</td>
<td>4.15 \times 10^{-3}</td>
<td>-8.38 \times 10^{-2}</td>
<td>-6.93 \times 10^{-3}</td>
<td>-2.20 \times 10^{2}</td>
<td></td>
</tr>
<tr>
<td>(d_{\text{or}})</td>
<td>3.15 \times 10^{-3}</td>
<td>-5.13 \times 10^{-2}</td>
<td>1.31 \times 10^{-3}</td>
<td>6.37 \times 10^{-2}</td>
<td>-1.00 \times 10^{-3}</td>
<td>1.08 \times 10^{1}</td>
<td></td>
</tr>
<tr>
<td>(h_{\text{noz}}) (m)</td>
<td>-1.01 \times 10^{-3}</td>
<td>1.77 \times 10^{-2}</td>
<td>7.02 \times 10^{-4}</td>
<td>2.33 \times 10^{-2}</td>
<td>3.12 \times 10^{-4}</td>
<td>1.20</td>
<td></td>
</tr>
<tr>
<td>(r_{i}) (Hz)</td>
<td>6.38 \times 10^{-4}</td>
<td>-1.08 \times 10^{-2}</td>
<td>-1.57 \times 10^{-4}</td>
<td>1.36 \times 10^{-2}</td>
<td>-2.03 \times 10^{-4}</td>
<td>-2.06 \times 10^{-4}</td>
<td></td>
</tr>
<tr>
<td>(d_{p}) (µm)</td>
<td>-1.47 \times 10^{-4}</td>
<td>2.42 \times 10^{-3}</td>
<td>8.93 \times 10^{-4}</td>
<td>3.43 \times 10^{-3}</td>
<td>4.68 \times 10^{-5}</td>
<td>-5.24 \times 10^{-2}</td>
<td></td>
</tr>
</tbody>
</table>
Table 4.10. Translation factors (central difference, calculated as the average from both single-sided differences) for the particle-related output variables.

<table>
<thead>
<tr>
<th>Output variable ((\Omega))</th>
<th>(T_p)</th>
<th>(s(T_p))</th>
<th>(W_p)</th>
<th>(s(W_p))</th>
<th>(\delta_c)</th>
<th>(s(\delta_c))</th>
</tr>
</thead>
<tbody>
<tr>
<td>(v_{a,in}) (m s(^{-1}))</td>
<td>6.67 \times 10^{-2}</td>
<td>4.26 \times 10^{1}</td>
<td>-6.49 \times 10^{1}</td>
<td>-2.55 \times 10^{1}</td>
<td>-7.70 \times 10^{2}</td>
<td>7.56 \times 10^{4}</td>
</tr>
<tr>
<td>(P_a) (bar)</td>
<td>-1.61 \times 10^{-1}</td>
<td>1.13 \times 10^{1}</td>
<td>-3.06 \times 10^{1}</td>
<td>-8.49 \times 10^{1}</td>
<td>7.78 \times 10^{3}</td>
<td>2.06 \times 10^{2}</td>
</tr>
<tr>
<td>(T_a) (K)</td>
<td>6.46 \times 10^{-2}</td>
<td>-1.33</td>
<td>-2.96 \times 10^{2}</td>
<td>1.07 \times 10^{1}</td>
<td>-2.85 \times 10^{2}</td>
<td>-3.13 \times 10^{4}</td>
</tr>
<tr>
<td>(\varphi_{a})</td>
<td>7.28 \times 10^{-4}</td>
<td>-9.09 \times 10^{3}</td>
<td>-1.00 \times 10^{2}</td>
<td>2.50 \times 10^{3}</td>
<td>2.46 \times 10^{3}</td>
<td>2.15 \times 10^{2}</td>
</tr>
<tr>
<td>(\dot{M}_{sol}) (g min(^{-1}))</td>
<td>5.43 \times 10^{-2}</td>
<td>5.26 \times 10^{1}</td>
<td>1.88</td>
<td>1.37</td>
<td>1.16</td>
<td>9.92 \times 10^{1}</td>
</tr>
<tr>
<td>(T_{sol}) (K)</td>
<td>-2.93 \times 10^{-2}</td>
<td>5.26 \times 10^{1}</td>
<td>5.74 \times 10^{1}</td>
<td>6.76</td>
<td>5.86 \times 10^{1}</td>
<td>1.52</td>
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<tr>
<td>(\mu_{sol})</td>
<td>5.06 \times 10^{-4}</td>
<td>-7.41 \times 10^{3}</td>
<td>1.00 \times 10^{1}</td>
<td>1.68 \times 10^{1}</td>
<td>1.35 \times 10^{2}</td>
<td>1.98 \times 10^{2}</td>
</tr>
<tr>
<td>(\gamma_{sol})</td>
<td>-1.39 \times 10^{-3}</td>
<td>-3.21 \times 10^{1}</td>
<td>4.44 \times 10^{2}</td>
<td>2.45 \times 10^{1}</td>
<td>-7.22 \times 10^{3}</td>
<td>1.41 \times 10^{1}</td>
</tr>
<tr>
<td>(d_{a}) (m)</td>
<td>-5.90 \times 10^{-3}</td>
<td>-2.31 \times 10^{1}</td>
<td>3.30 \times 10^{1}</td>
<td>1.02</td>
<td>1.29 \times 10^{2}</td>
<td>-8.34 \times 10^{1}</td>
</tr>
<tr>
<td>(h_{sol}) (m)</td>
<td>4.79 \times 10^{-2}</td>
<td>8.10 \times 10^{1}</td>
<td>-6.35 \times 10^{1}</td>
<td>-4.78 \times 10^{1}</td>
<td>-6.11 \times 10^{2}</td>
<td>2.13</td>
</tr>
<tr>
<td>(r_{i}) (Hz)</td>
<td>-1.36 \times 10^{-2}</td>
<td>-1.36 \times 10^{2}</td>
<td>1.48 \times 10^{2}</td>
<td>1.17 \times 10^{2}</td>
<td>-2.75 \times 10^{4}</td>
<td>-5.77 \times 10^{1}</td>
</tr>
<tr>
<td>(d_{p}) ((\mu)m)</td>
<td>9.43 \times 10^{-3}</td>
<td>6.58 \times 10^{1}</td>
<td>2.75</td>
<td>1.39</td>
<td>2.41 \times 10^{3}</td>
<td>-4.15 \times 10^{1}</td>
</tr>
</tbody>
</table>

Table 4.11. Translation factors (central difference, calculated as the average from both single-sided differences) for the droplet phase-related properties.

<table>
<thead>
<tr>
<th>Output variable ((\Omega))</th>
<th>(\dot{M}_{sd})</th>
<th>(\bar{h}_{dc})</th>
<th>(s(h_{dc}))</th>
<th>(\bar{T}_{dc})</th>
<th>(\bar{D}_{M,dc})</th>
</tr>
</thead>
<tbody>
<tr>
<td>(v_{a,in}) (m s(^{-1}))</td>
<td>1.87</td>
<td>1.42</td>
<td>1.41 \times 10^{1}</td>
<td>2.20 \times 10^{2}</td>
<td>1.10 \times 10^{4}</td>
</tr>
<tr>
<td>(P_a) (bar)</td>
<td>-2.35 \times 10^{-1}</td>
<td>6.70 \times 10^{-2}</td>
<td>-2.87 \times 10^{2}</td>
<td>1.40 \times 10^{3}</td>
<td>4.19 \times 10^{2}</td>
</tr>
<tr>
<td>(T_a) (K)</td>
<td>1.51</td>
<td>-1.30 \times 10^{1}</td>
<td>3.83 \times 10^{1}</td>
<td>1.96 \times 10^{2}</td>
<td>2.65 \times 10^{1}</td>
</tr>
<tr>
<td>(\varphi_{a})</td>
<td>-1.51 \times 10^{1}</td>
<td>8.04 \times 10^{3}</td>
<td>-4.80 \times 10^{3}</td>
<td>4.99 \times 10^{3}</td>
<td>7.99 \times 10^{2}</td>
</tr>
<tr>
<td>(\dot{M}_{sol}) (g min(^{-1}))</td>
<td>-5.89 \times 10^{-1}</td>
<td>-3.44 \times 10^{2}</td>
<td>9.87 \times 10^{2}</td>
<td>-8.51 \times 10^{3}</td>
<td>-5.84 \times 10^{1}</td>
</tr>
<tr>
<td>(T_{sol}) (K)</td>
<td>15.0</td>
<td>-3.74</td>
<td>4.18 \times 10^{1}</td>
<td>1.59 \times 10^{1}</td>
<td>6.25 \times 10^{4}</td>
</tr>
<tr>
<td>(\mu_{sol})</td>
<td>-7.41 \times 10^{-1}</td>
<td>-2.02 \times 10^{2}</td>
<td>3.41 \times 10^{2}</td>
<td>-7.18 \times 10^{3}</td>
<td>-1.73 \times 10^{2}</td>
</tr>
<tr>
<td>(\gamma_{sol})</td>
<td>-2.40 \times 10^{-1}</td>
<td>-3.32 \times 10^{1}</td>
<td>2.76 \times 10^{1}</td>
<td>-4.48 \times 10^{1}</td>
<td>-1.87 \times 10^{1}</td>
</tr>
<tr>
<td>(d_{a}) (m)</td>
<td>-7.88 \times 10^{-1}</td>
<td>-3.33 \times 10^{1}</td>
<td>5.67 \times 10^{1}</td>
<td>3.32 \times 10^{3}</td>
<td>-9.36 \times 10^{2}</td>
</tr>
<tr>
<td>(h_{sol}) (m)</td>
<td>2.97 \times 10^{-1}</td>
<td>1.29</td>
<td>-3.38 \times 10^{-3}</td>
<td>4.21 \times 10^{-3}</td>
<td>-1.90</td>
</tr>
<tr>
<td>(r_{i}) (Hz)</td>
<td>1.32 \times 10^{2}</td>
<td>5.27 \times 10^{-3}</td>
<td>1.37 \times 10^{2}</td>
<td>1.57 \times 10^{3}</td>
<td>-8.27 \times 10^{2}</td>
</tr>
<tr>
<td>(d_{p}) ((\mu)m)</td>
<td>1.84 \times 10^{-1}</td>
<td>9.09 \times 10^{-3}</td>
<td>-2.36 \times 10^{-1}</td>
<td>-5.22 \times 10^{4}</td>
<td>1.55 \times 10^{2}</td>
</tr>
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</table>
Figure 4.48. Translation factors describing the predicted effects of variation on the outlet air temperature, $\bar{T}_{a,\text{out}}$ (a), the outlet air relative humidity, $\bar{\varphi}_{a,\text{out}}$ (b), and the outlet air humidity, $\bar{X}_{a,\text{out}}$ (c). Single-sided difference, □: $\Delta \omega < 0$, ■: $\Delta \omega > 0$. 

Modelling heat and mass transfer in fluidised bed coating processes
Figure 4.48. Translation factors describing the predicted effects of variation on the vaporisation efficiency, $\bar{\eta}_{\text{vap}}$(d), the thermal efficiency, $\eta_{\text{th}}$(e), and the overall heat loss rates, $\Phi_{\text{loss}}$(f). Single-sided difference, $\Delta \omega < 0$, $\Delta \omega > 0$. 
Figure 4.48. Translation factors describing the predicted effects of variation on the average particle temperature, $\bar{T}_p$ (g), the particle temperature standard deviation, $s(T_p)$ (h), and the average particle moisture content, $\bar{W}_p$ (i). Single-sided difference, □: $\Delta \omega < 0$, ■: $\Delta \omega > 0$. 
Figure 4.48. Translation factors describing the predicted effects of variation on the particle moisture content standard deviation, \( s(W_p) \) (j), the average coating growth rate \( \bar{\delta}_c \) (k), and its standard deviation, \( s(\delta_c) \) (l). Single-sided difference, ■: \( \Delta \omega < 0 \), ▲: \( \Delta \omega > 0 \).
Figure 4.48. Translation factors describing the predicted effects of variation on the spray drying loss, $M_{sd}$ (m), the average droplet collision height, $h_{drc}$ (n) and the standard deviation on the droplet collision height, $s(h_{drc})$ (o). Single-sided difference, $\Delta \omega < 0$, $\Delta \omega > 0$. 
In the following sections, the effect of each input variable is discussed:

4.7.3.3. Inlet air flow rate

Adjusting the fluidisation air flow rate alters the bed height, $h_{\text{bed}}$ and the particle exchange rate, $r_i$, as detailed in Figure 3.39. Therefore, to exclude the effect of shortening the droplet path length by altering the bed height, the position of the nozzle was adjusted in such a way that the nozzle tip coincided with the bed height in each simulation.

Increasing the air flow rate significantly increased the evaporative capacity of the bed and thus increases the overall spray drying loss, as shown in Figures 4.48m and 4.49a. In Figure 4.49b, the model-predicted steady state heat exchange between the droplet phase and the gas
phase is plotted against the inlet air flow rate. At low fluidisation air flow rates, the droplet/gas heat transfer was negligible, while at high values fluidisation air flow rates, a significant increase in heat transfer between the droplet phase and the fluidising air in the bed’s coating volume was observed, indicating a higher potential for premature droplet evaporation.

![Figure 4.49](image)

**Figure 4.49.** Model-predicted spray drying losses (relative to the reference case) as a function of the volumetric inlet air flow rate (a), and the simulated axial droplet/gas phase heat exchange profiles as a function of the volumetric inlet air flow rate (b).

The bed’s coating volume – which is computed in the model as the region where 90 % of the droplet collection on the fluidised particles takes place and is shown in Figure 4.50a – was reduced when using a higher inlet air flow rate. A number of effects could explain this increase of the bed’s coating (or spraying) volume, including the correlation between the evaporation rate and inlet air flow rate, larger bed expansion for higher inlet air flow rates and the relationship between drag force – exerted on the droplets – and the inlet air flow rate. The predominant factor is the increase in bed voidage ($\varepsilon_{\text{bed}}$) and hence, bed volume ($h_{\text{bed}}$) when increasing the fluidisation air flow rate, as shown in Figure 4.50a. Nevertheless, even when considering the volume of the bed’s coating region in absolute value – in m³ in stead of % of the total bed volume – a reduction can still be observed for higher inlet air flow rates.

Besides a reduction in the bed’s volume where effective droplet collection takes place, the droplets appeared to penetrate deeper into the bed at lower fluidisation air flow rates, as shown in Figure 4.50b. The droplet evaporation, being insignificant at lower air flow rates, leads to a droplet size reduction along its trajectory which results in a more rapid deceleration on the one hand and in an increase in adhesion efficiency on the other hand, explaining the lower droplet collection height, $h_{\text{drc}}$ at lower inlet air flow rates, $G_{a,\text{in}}$ (or $V_{a,\text{in}}$).
Figure 4.50. Model-predicted relative size of the bed’s coating volume (■) and bed voidage (▲) as a function of the volumetric inlet air flow rate (a); the model-predicted axial dry matter deposition rate ($r_{Cj}$ in mg s$^{-1}$) profile, as a function of volumetric inlet air flow rate (b).

Figure 4.51. Model-predicted coating layer growth rate ($\delta_c$ in $\mu$m s$^{-1}$) during steady state coating regime as a function of volumetric fluidisation air flow rate: average (■), maximum (▲) and minimum growth rate (●).

Finally, the effect on the coating layer growth rate ($\delta_c$ in $\mu$m s$^{-1}$) is illustrated in Figure 4.51. Although the fluidisation air flow rate has a predictable decreasing effect on the average growth rate due to the increase in premature droplet evaporation, its effect on the growth rate distribution is less trivial: an optimum (i.e. narrow) growth rate distribution was observed around $V_{a,\text{in}} = 0.013$ m s$^{-1}$. The relationship between the inlet air flow rate and the coating
layer growth rate distribution illustrates the rather complex interactions between fluidisation air flow rate, the bed’s coating volume size, the local droplet collection (impingement + adhesion) probabilities and the local droplet drying kinetics.

4.7.3.4. Atomisation air properties

The effects of the atomisation air pressure on droplet collection mechanisms and bed thermodynamics are complex. Firstly, increasing the atomisation air pressure will increase the volumetric air flow rate of cooler and drier (compared to the fluidisation air) atomisation air. Consequently, the air temperature in the upper part of the bed decreases which in turn, slows down the droplet evaporation rate. Secondly, as shown by Lefebvre (1998) and Juslin et al. (1995), increasing the air pressure also decreases the Sauter droplet diameter and increases the initial droplet velocity. The relationship between the droplet size and droplet evaporation rate is nearly linear (Leclère et al., 2004). Furthermore, smaller droplets tend to decelerate faster and will therefore be more susceptible to complete evaporation. Finally, the droplet collection probability, $\zeta_{dr}$, depends on the droplet size. Whereas smaller droplets have – as shown in Section 1.3.2.3 – decreased impingement efficiency ($\chi_{dr}$), their adhesion probability ($\zeta_{ad}$) is increased. Furthermore, smaller droplets – produced at higher atomisation air pressures – also have higher velocities which in turn, result in improved impingement efficiencies and thus counteract any decrease in impingement efficiency caused by the reduced droplet diameter.
As a result of these complex and counteracting phenomena, the average droplet path length is only slightly reduced with increasing atomisation air pressures (Figure 4.48n, and Table 4.11). In Figure 4.52a, the modelled droplet penetration distribution is given for different atomisation air pressures, while Figure 4.53b gives the axial coating deposition rate profile \( r_{Ci} \) as a function of atomisation air pressure, \( P_{at} \). The calculated size of the bed’s coating volume is presented in Figure 4.53a.

Similarly, the effect of varying the atomisation air pressure on the spray drying losses, and hence the coating efficiency, was somewhat less pronounced (Figure 4.48m and Table 4.11). According to the coating experiments of Dewettinck and Huyghebaert (1998) and of Saleh et al. (1999), an optimal atomisation air pressure could be found, which resulted in minimal spray drying losses or highest coating efficiency. This effect could be reproduced using the presented model. Figure 4.52c shows the model-predicted spray drying losses, compared to the reference conditions of \( P_{at} = 2.5 \) bar, as a function of atomisation air pressure. Figure 4.52b illustrates the model-predicted axial droplet/air heat exchange profile as a function of atomisation air pressure. According to the model-predicted results, a pressure of \( P_{at} = 3 \) bar yielded minimal spray drying losses and minimal droplet/air heat transfer.
Figure 4.53. Model-predicted relative size of the bed’s coating volume as a function of the atomisation air pressure ($P_{at}$ in bar) (a); the model-predicted axial dry matter deposition rate ($r_{C,i}$ in mg s$^{-1}$) profile (b).

The contribution of the atomisation air temperature to the air temperature and air humidity gradients in the fluidised bed has been detailed in Chapter 3. Due to the use of Kelvin for both input and output variables in the calculation of the translation factor, these translation factors were considerably larger than, for instance, atomisation air pressure. Nonetheless, increasing the temperature of the atomisation air had an augmenting effect on the spray drying losses as shown in Figures 4.48m and 4.54a. The increase in spray drying loss was accompanied by increased droplet collision temperatures, $T_{drc}$, and dry matter contents, $DM_{drc}$, as demonstrated in Figures 4.48p and 4.48q. Because of the increased reduction in droplet diameter at higher values of the atomisation air temperature and its effect on local droplet collection efficiency and droplet movement, the droplets were captured over a larger portion of the fluidised bed as shown in Figures 4.48n and 4.48o.

Finally, the atomisation air relative humidity, $\varphi_{at}$, had – albeit less pronounced – an opposite effect as the atomisation air temperature, $T_{at}$. In Figure 4.54b the model-predicted spray drying loss is given as a function of atomisation air relative humidity. Because the mixing of the gas phase in the upper part of the bed with atomisation air with increasing humidity, the bed’s local evaporative capacity was reduced resulting in a reduction of the premature droplet evaporation (or spray drying) as shown in Figure 4.54b.
4.7.3.5. Spraying rate and coating solution properties

In the sensitivity analysis, the solution’s dry matter content \( (DM_{sol}) \) was omitted as an input variable because, firstly – as shown in 3.7.3.6 – the bed thermodynamics purely relate to the solvent feed rate. Consequently, the thermodynamic effect of varying dry matter content of the coating solution can be reduced to varying \( \dot{M}_{sol} \). Secondly, the effect of dry matter content on the coating mass distribution is perfectly linear. Instead of studying the effect of the coating solution’s dry matter content, the solution’s viscosity and surface tension were added in this sensitivity analysis as they play an important role in the droplet formation process and droplet/particle collection efficiency (although these properties depend on the type of dissolved coating material and its concentration).

The spraying rate is the principal variable influencing the bed’s temperature and humidity. Increasing the spraying rate yields larger temperature and humidity gradients in the fluidised bed (Hemati et al., 2003). As a result of the lower air temperatures and higher air humidity in the upper part – i.e. close to the spraying nozzle – of the bed when increasing the spraying rate, the droplet evaporation rate was reduced, resulting in lower spray drying losses (Figures 4.48m and 4.55c). From Figure 4.55c it can be observed that the spray drying loss curve had a sigmoidal shape, while in Figure 4.55b, where the droplet/gas-phase heat exchanges are illustrated, the modelled heat transfer reached a maximum value at \( \dot{M}_{sol} = 10 \text{ g min}^{-1} \). The reason for this occurrence is that, at lower spraying rates, the amount of freely available coating solution in the bed (i.e. droplets) becomes limiting while at higher spraying rates, the higher air humidity in the bed is the limiting factor for the droplet evaporation rate.

As a result of the reduction in droplet evaporation rate, a slightly deeper droplet penetration depth (Figures 4.55a and 4.56b) was predicted. But the overall effect on the calculated bed’s
coating volume size was less pronounced - or more specifically, within the studied range of 2 to 20 g min⁻¹, the coating volume variation was approximately 4% of the total bed volume – as shown in Figure 4.56a.

![Diagram](image)

**Figure 4.55.** Model-predicted droplet penetration height ($h_{dpc}$ in m) as a function of spraying rate ($\dot{M}_{sol}$ in g min⁻¹): average (■); 10th (▲) and 90th (●) percentile of droplet penetration depth (a), the simulated axial droplet/gas-phase heat exchange profiles (b) and spray drying losses, relative to the reference case $\dot{M}_{sol} = 10$ g min⁻¹ (c).

Finally, the effect of the spraying rate on the coating layer growth rate, $\delta_c$, was studied. Whereas in Chapter 3, a pure linear relationship was found between spraying rate and coating layer growth rate, the predicted growth rate using the extended model was somewhat non-linear, as shown in Figure 4.57. The inclusion of the spray drying loss phenomenon in the
extended model, with its S-shaped curve in Figure 4.55c, forms the basis for the non-linear growth behaviour of the coating layer.

![Graph](image-url)

**Figure 4.56.** Model-predicted relative size of the bed’s coating volume as a function of the spraying rate (in g min\(^{-1}\)) (a); the model-predicted axial dry matter deposition rate \(r_{C,i}\) in mg s\(^{-1}\) profile, as a function of the spraying rate (b).

![Graph](image-url)

**Figure 4.57.** Model-predicted coating layer growth rate \(\delta_c\) in µm s\(^{-1}\)) during steady state coating regime as a function of spraying rate: average (■), maximum (▲) and minimum growth rate (●).

With respect to the impact of the temperature of the coating solution, \(T_{sol}\), it can be observed from the translation factors in Figure 4.48, that the temperature at which the coating solution is injected into the bed had a large impact on the spray drying losses. Consequently, the
particle moisture distribution, $W_p$, the collision height distribution, $h_{dec}$, droplet temperature, $T_{dec}$, and dry matter content, $DM_{dec}$, at droplet/particle-impact were largely affected by the coating solution’s temperature.

A first explanation lies in the expression of the input variable – $\omega$ in Eq. (3.55), being the solution temperature ($T_{sol}$) – in Kelvin rather then degrees Celsius and thus, the term $\partial \omega / \omega$ in Eq. (3.55) is significantly smaller. However, aside from the biased translation factors, the coating solution temperature still has a pronounced, physical-based effect on the droplet phase-related variables. For instance, the model-predicted spray drying loss – as shown in Figure 4.58a – varied between 35 and 200 % of spray drying loss encountered in the reference case (being $M_{sd} = 3.8 \text{ w}\%$ of the total amount of coating material injected into the bed) when the coating solution’s temperature, $T_{sol}$, varied between 15 and 50 °C. The physical explanation behind this strong correlation is that, when assuming the droplet drying process to be adiabatic, an additional drying loss of 0.17 w% was obtained per Kelvin temperature difference between the initial droplet temperature and the adiabatic drying temperature. This additional drying loss due to the initial droplet temperature resulted in the large effect on the overall spray drying losses in Figure 4.58a.

![Figure 4.58. Model-predicted spray drying losses (relative to the reference case) (a) and the simulated axial droplet/gas-phase heat exchange profile as a function of the temperature of the injected coating solution temperature, $T_{sol}$ (b).](image)

The increase in premature droplet evaporation due to elevated coating solution temperatures also resulted in a decrease in the bed’s coating volume size, as shown in Figure 4.59. Another consequence of the increased spray drying loss, is that the average growth rate, as shown in Figure 4.60, decreased with increasing coating solution temperature, but the growth rate standard deviation – which expresses the ‘uniformity’ by which the coating material is...
distributed among the particle population – increased. In fact, for all input variables studied in this sensitivity analysis, there appeared to be a negative correlation between the overall spray drying losses, $M_{sd}$, and the coating layer growth rate standard deviation.

![Figure 4.59](image_url)  
**Figure 4.59.** Model-predicted relative size of the bed’s coating volume (a) and the steady state axial dry matter deposition rate ($r_{C,i}$ in mg s$^{-1}$) profile, as a function of the coating solution temperature, $T_{sol}$ (b).

![Figure 4.60](image_url)  
**Figure 4.60.** Model-predicted coating layer growth rate ($\delta_c$ in µm s$^{-1}$) during steady state coating regime as a function of coating solution temperature: average (■), maximum (▲) and minimum growth rate (●).

A third studied input variable was the coating solution viscosity, $\mu_{sol}$. Based on the translation factors in Figure 4.48, there was only a minor effect on the particle moisture content...
distribution ($W_p$), steady-state heat loss and the droplet collision depth distribution. The only exception was the overall steady-state spray drying loss ($M_{sd}$ in Figure 4.48m). As detailed in Sections 4.3.2 and 4.3.4, the viscosity of the coating solution played an important role in the droplet formation process and the successful collection of droplets onto the surface of the fluidised particles. According to Eq. (1.1) – giving the droplet size model used in this study – increasing the viscosity increases the diameter of the droplets exiting the nozzle. Because of the larger droplets at higher values for the coating solution viscosity, the droplet/gas-phase mass transfer was considerably reduced resulting in lowered spray drying losses as shown in Figure 4.61a.

Increasing the droplet viscosity results in larger adhesion probabilities, $\text{ad}$ (Panda et al., 2001a,b). However, as shown in Eq. (4.36), this effect is counteracted by the increasing droplet diameter, as larger droplets will have smaller critical impingement velocities. An increase in droplet diameter also improves the droplet impingement efficiency, $\chi_{dr}$. Furthermore, the good wettabili ty (or small contact angle, $\theta_{dp}$) generally results in the impingement efficiency being the limiting factor in determining the overall collection efficiency, as shown in Eq. (4.37). Based on these assumptions, the higher the droplet viscosity, the closer it gets collected near the nozzle as can be observed, albeit less pronounced, in Figure 4.61b. The peak values observed in Figure 4.61b indicate the transition from droplet impingement efficiency towards droplet adhesion probability being the limiting factor in determining the overall droplet collection efficiency, $\zeta_{dr}$. Increasing the viscosity of the coating solution also appeared to increase the size of the coating volume as demonstrated in Figure 4.61c.

The negative correlation between droplet collision depth (or $h_{dc}$) and droplet diameter is in apparent contradiction with the model-predicted results obtained from varying the atomisation air pressure in Section 4.7.3.4, where a higher atomisation air pressure resulted in smaller droplets, which collided at a smaller distance from the nozzle than larger ones (in case of using lower atomisation air pressures). The explanation behind the opposite effect of coating solution viscosity and atomisation air pressure is that: first, the critical impingement velocity is not modified (increased) through the atomisation air pressure as opposed to coating solution viscosity. Secondly, increasing the atomisation air pressure also increases the initial droplet velocity, which in turn, has a beneficial effect on the impingement efficiency.
Figure 4.61. Model-predicted steady state spray drying losses, relative to the reference case $M_{sol} = 10 \text{ g min}^{-1}$, as a function of the coating solution viscosity (a), average ($\square$); 10th ($\triangle$) and 90th ($\bullet$) percentile of droplet penetration depth (b) and relative size of the bed’s coating volume as a function of the viscosity of the coating solution, $\mu_{sol}$ (c).

Similar to the viscosity, $\mu_{sol}$, increasing the surface tension of the coating solution, $\gamma_{sol}$, results in the production of larger droplets. The difference is that in Eq. (4.36) the effect of increasing the droplet diameter (at higher values of $\mu_{sol}$) was more or less neutralised by the $\mu_{dr}$-term (= $\mu_{sol}$) in the nominator of Eq. (4.36). This neutralising effect, however, is absent in case of varying the droplet size through adjusting the coating solution surface tension. The increased droplet adhesion probabilities at higher values of the surface tension of the coating solution, will give rise to an increased volume of the spraying region (Figures 4.62b and 4.62c), wider particle temperature and moisture content distributions (Figures 4.48h and 4.48j) and less spray drying losses (Figure 4.62a).
The results dealing with the influence of coating solution surface tension, are in agreement with the experimental observations of both Panda et al. (2001a,b) and Hemati et al. (2003), who reported that coating solution surface tension is supposed to have a higher influence on droplet deposition and particle growth compared to the viscosity of the coating solution.

![Graphs](a) (b) (c)

**Figure 4.62.** Model-predicted steady state spray drying losses, relative to the reference case $M_{\text{sol}} = 10 \text{ g min}^{-1}$, as a function of the coating solution surface tension (a), average (■); $10^{\text{th}}$ (▲) and $90^{\text{th}}$ (▲) percentile of droplet penetration depth (b) and relative size of the bed’s coating volume as a function of the viscosity of the coating solution, $\mu_{\text{sol}}$ (c).
4.7.3.6. Nozzle height

Figure 4.63a shows the results for the effect of nozzle position, $h_{\text{noz}}$, on the droplet penetration depth distribution in a fluidised bed with bed height, $h_{\text{bed}} = 0.093$ m. From a certain nozzle height ($h_{\text{noz}} > 0.13$ m), little influence on the droplet penetration depth was observed. By increasing the distance between the nozzle and the fluidised bed, the volume of the spraying region – the region where collision between particles and droplets occurs – is reduced and consequently, as shown in Figures 4.64a and 4.64b, the particle temperature and moisture content have wider distributions, as shown in Figures 4.48h and 4.48j. The pronounced translation factors for droplet depth distribution in Figures 4.48n and 4.48o result from the fact that the reference condition used a nozzle height, $h_{\text{noz}} = 0.10$ m, a region where strong correlation exists between nozzle height and droplet penetration depth (cfr. Figure 4.63a). By comparing the results from Figure 4.63a with the spray drying losses shown in Figure 4.63c, it can be seen that spray drying losses continue to increase significantly, even beyond a nozzle height of $h_{\text{noz}} = 0.13$ m.
Figure 4.63. Model-predicted droplet penetration height ($h_{drc}$ in m) as a function of nozzle height ($h_{noz}$ in m): average (■); 10th (▲) and 90th (◆) percentile of droplet penetration height (a), the simulated axial droplet/gas-phase heat exchange profiles (b) and spray drying losses (relative to the reference case, $\dot{M}_{sol} = 10$ g min$^{-1}$) as a function of the nozzle height.

Figure 4.64. Model-predicted relative size of the bed’s coating volume as a function of the nozzle height ($h_{noz}$ in m) (a); the model-predicted axial dry matter deposition rate ($r_{C,i}$ in mg s$^{-1}$) profile, as a function of nozzle height (b).
The impact of the nozzle position on the coating layer growth rate $\delta_c$, is presented in Figure 4.65. As expected, the average growth rate $\delta_c$ decreases with increasing nozzle height due to the elevated spray drying losses. At a height of $h_{noz} = 0.110$ to $0.115$ m, the growth rate distribution was at its widest, and a peak value for the maximum growth rate was attained. From Figure 4.63a, it can be seen that this nozzle position corresponds to the height where the bed’s coating volume starts to coincide with the top of the fluidised bed. The physical meaning is that, at a height of $h_{noz} = 0.110$ to $0.115$ m, part of the droplets have decelerated sufficiently in order to have high droplet collection efficiencies at the onset of droplet penetration into the bed, so this fraction of the droplets is immediately captured upon entry into the bed. These droplets will have the least amount of solvent (water) removed during their trajectories and thus contributing the largest amount of solvent (water) to the particle surface, $W_p$. Above this critical nozzle height, the decelerated droplets are exposed much longer to the fluidising air, creating significantly larger spray drying losses, as shown in Figure 4.63c.

A second nozzle-related variable that was added in this sensitivity analysis was the diameter of the nozzle’s liquid orifice, $d_{or}$. Next to the spraying rate, coating solution viscosity, coating solution surface tension and atomisation air pressure, the orifice diameter is the fifth and final variable affecting the droplet diameter according to Eq. (1.1). More specifically, the droplet surface-weighted mean diameter is proportional to $\sqrt{d_{or}}$. But, compared to the other droplet diameter-altering variables, the orifice diameter does not alter the bed’s thermodynamics, nor droplet initial velocities and it does not induce additional interactions (e.g. $\mu_{sol}$ or $\gamma_{sol}$), aside from the altered droplet diameter, on droplet impingement efficiency or adhesion probability.

**Figure 4.65.** Model-predicted coating layer growth rate ($\delta_c$ in $\mu$m s$^{-1}$) during steady state coating regime as a function of nozzle height: average (-■-), maximum (-▲-) and minimum growth rate (-◆-).
In Figure 4.66a, the model-predicted droplet penetration depth is given as a function of the diameter of the liquid orifice of the pneumatic nozzle. Because increasing the orifice diameter results in the production of larger droplets and because the droplet collection efficiency, \( \zeta_{\text{dr}} \), is not additionally affected by changes in coating solution viscosity or surface tension, the heavier droplets will penetrate deeper into the bed due to their inertia. As a result of the larger droplets, there is a profound effect of \( d_{\text{or}} \) on the particle temperature, humidity and coating layer growth rate distributions as shown in Figures 4.48g to 4.48l.

**Figure 4.66.** Model-predicted droplet penetration height \( (h_{\text{dc}} \text{ in m}) \): average (■■■); 10\(^{\text{th}}\) (▲▲▲) and 90\(^{\text{th}}\) (●●●) percentile droplet penetration height (a), the simulated axial droplet/gas-phase heat exchange profiles (b) and spray drying losses (relative to the reference case, \( M_{\text{sol}} = 10 \text{ g min}^{-1} \)) as a function of the nozzle liquid orifice diameter \( (d_{\text{or}} \text{ in m}) \).
For smaller nozzle orifice diameters, the droplets not only penetrate less deep into the fluidised bed, the size of the region into which the droplets adhere onto the particles (i.e. the bed’s coating volume) is considerably smaller. Figure 4.67a illustrates the model-predicted size of the bed’s coating volume, while Figure 4.67b shows the axial droplet collection rate profile as a function of orifice diameter, \(d_{\text{or}}\).

Finally, the coating layer growth rate, \(\delta_c\), is illustrated in Figure 4.68. The decreasing width of the growth rate distribution at higher values of the orifice diameter appears to contradict the fact that higher values for \(d_{\text{or}}\) result in larger droplets. However, as already demonstrated in Section 4.7.3.1, there are two major factors influencing the coating mass distribution (or growth rate): the particle residence time in the bed’s coating volume – which is the result of the particle exchange rate and the relative size of the bed’s coating volume – and droplet deposition. As was demonstrated in Figure 4.45, the droplet deposition is usually not the limiting factor in determining the width of the coating mass (or growth rate) distribution. Instead, the relative size of the bed’s coating volume, as shown in Figure 4.67a, nearly doubles within the studied diameter range which explains the decreasing coating layer growth rate distribution width, \(s(\delta_c)\), as a function of \(d_{\text{or}}\).

\[\text{Figure 4.67.} \quad \text{Model-predicted relative size of the bed’s coating volume as a function of the nozzle liquid orifice diameter} \ (d_{\text{or}} \text{ in m}) \ (a); \text{the model-predicted axial dry matter deposition rate} \ (r_{C,i} \text{ in mg s}^{-1}) \text{ profile, as a function of nozzle liquid orifice diameter} \ (b).\]
4.7.3.7. Particle exchange rate

The reason for including the particle exchange rate, \( r_i \), in this sensitivity analysis is that \( r_i \) is linked with the average particle velocity (Eq. (4.35)) and thus affects the droplet impingement efficiency. However, as shown in Figure 4.48n, the effect of the particle exchange rate on the droplet/particle collection height distribution was negligible. In fact, modifying the particle exchange rate had a negligible effect on any variable relating to droplet/particle deposition, as shown in Table 4.11. The only model-predicted output variables sensitive to the particle exchange rate are the particle temperature \( (T_p) \), moisture \( (W_p) \) and coating mass \( (Y_p) \) or growth rate, \( \delta_c \) as shown in Figure 4.69) distributions, and the heat loss. The effect of particle exchange rate on thermodynamic variables such as \( T_p, W_p \) and \( Q_{\text{loss}} \) could be attributed to the increased degree of bed homogeneity at higher values of the particle exchange rate. Coating mass or growth rate distribution is primarily influenced by the altered residence time of the particle in the bed’s coating volume.

Figure 4.68. Model-predicted average coating layer growth rate \( (\delta_c \text{ in } \mu m \text{ s}^{-1}) \) during steady state coating regime as a function of liquid orifice diameter \( (d_{or} \text{ in m}) \): average (■), maximum (▲) and minimum growth rate (●).
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Figure 4.69. Model-predicted average coating layer growth rate ($\delta_c$ in $\mu$m s$^{-1}$) during steady state coating regime as a function of particle exchange rate ($r_i$ in Hz): average (- - ), maximum (▲) and minimum growth rate (●).

4.7.3.8. Particle diameter

Similar to the inlet air flow rate, the particle diameter ($d_p$) alters the bed’s voidage ($\varepsilon_{\text{bed}}$) and particle exchange rate ($r_i$). However, the particle diameter, as opposed to the inlet air flow rate, $G_{a,\text{in}}$, does not modify the overall bed’s heat and mass balances.

When considering the translation factors as presented in Figure 4.48, it could be observed that the particle diameter had little influence on the droplet-related variables, except for the droplet collection height. Although the translation factor for $\Omega = \dot{M}_{\text{sd}}$ is relatively small, the effect of the particle size on the spray drying loss is not negligible, as demonstrated in Figure 4.70a. Two phenomena are responsible for the increased spray drying loss when larger sized particles are used as the core material. First, as demonstrated in Section 3.7.3.10, the particle diameter has a pronounced effect on the air temperature and humidity gradients across the fluidised bed because the bed’s mixing behaviour, expressed through $r_i$, depends on the particle diameter. Second, the size of the bed’s volume where droplets are collected by the fluidised particles decreases with larger sized particles. The impingement efficiency decreases with larger particles. However, the larger particle diameter, the lower the bed voidage (more dense), thus the probability for droplet/particle contact increases, regardless of the droplet collection efficiency. The net result of the changes in both bed voidage and droplet collection efficiency is given in Figure 4.71, where a decreasing size of the bed’s coating volume was observed at higher values for the particle diameter.

Consequently, a decreasing size of the bed’s coating volume is normally associated with decreasing spray drying loss. When the particle diameter was changed, this relationship only remained valid for $d_p < 350$ $\mu$m (Figure 4.70a), but the opposite was true in case of increasing
the particle diameter beyond $d_p > 350 \, \mu m$. The remaining factor, the changes of the temperature and humidity gradients across the bed resulting from altering $d_p$ (and thus $r_i$) – more specifically higher air temperatures and lower air humidities when particles have a larger diameter – are likely to be the explanation behind the positive correlation between the particle diameter and spray drying losses when $d_p > 350 \, \mu m$.

![Figure 4.70](image1.png)

**Figure 4.70.** Model-predicted spray drying losses (relative to the reference case) (a) and the simulated axial droplet/gas-phase heat exchange profile as a function of the particle diameter ($d_p$ in $\mu m$) (b).

![Figure 4.71](image2.png)

**Figure 4.71.** Model-predicted relative size of the bed’s coating (a); the model-predicted axial dry matter deposition rate ($r_{C,i}$ in mg s$^{-1}$) profile, as a function of the particle diameter ($d_p$ in $\mu m$) (b).
4.7.4. Conclusions

Once the droplet phase extended model was experimentally validated in both the bed’s thermodynamics (Section 4.5) and spray drying losses (Section 4.6), a sensitivity analysis was performed to assess the impact of the different input variables on the dynamics of the batch fluidised bed coating process. From the sensitivity analysis of the model proposed in this chapter, the following conclusions could be drawn:

The effects of variables such as particle diameter, exchange rate and inlet air flow rate on the particle temperature and coating mass (excluding moisture content) distribution and the gas phase axial temperature and humidity profiles, described in Chapter 3, could be reproduced using the droplet-phase extended model. Particle moisture content distributions were different due to the assumption of continuous coating solution distribution in the basic model as opposed to the distribution by means of the droplet phase in the extended model.

The degree to which spray drying losses occur is the result of complex interactions between the three different phases. Within the group of variables studied, the temperature of the coating solution, the inlet air flow rate, atomisation air temperature and spraying rate were found to be most significant. Also material and configuration related properties, such as the coating solution’s viscosity and, to a lesser extent, the surface tension as well as the position of the nozzle above the fluidised bed and the diameter of the nozzle’s liquid orifice had a pronounced effect on premature droplet evaporation. Due to the used method of quantifying the model sensitivity, the effect of atomisation air temperature and coating solution temperature on the occurrence of spray drying was biased (overestimated). It is important to note that not every process variable included in this sensitivity analysis has the same relevance to practical coating applications, while others are restricted in their range. For example, the current model will predict larger droplets as beneficial in controlling spray drying losses, but does not take the elevated risk of particle agglomeration into consideration.

Next to the spray drying losses, the effect of the process variables on the size of the coating volume – defined as the part of the bed where successful droplet/particle adhesion takes place – was investigated. The importance of the size of this coating volume has been thoroughly discussed in Chapter 3, where a strong correlation between the particle temperature, moisture content and coating mass (or, coating layer growth rate) distributions and the relative size of the bed’s coating volume was found. Within the group of variables, the following variables were found to be the most influential, in decreasing order of importance: the diameter of the nozzle’s liquid orifice, coating solution surface tension, atomisation air temperature, coating solution temperature, location of the spraying nozzle above the fluidised bed, particle diameter and finally, the coating solution spraying rate. The same remark regarding the exaggeration of the effects of the atomisation air and the coating solution temperature due to the definition of sensitivity has to be made. Also remarkable was the small effect of the
atomisation air pressure on the bed’s coating volume size due to counteracting effects (droplet size vs. droplet velocity).

Finally, the effect on the coating layer growth rate distribution was studied. The growth rate distribution expresses the uniformity by which the coating material is deposited on the particles and narrow growth rate distributions are beneficial to the performance of encapsulated products in practical applications (i.e. in real food matrices). The average growth rate is purely related to the degree of spray drying loss, while the growth rate standard deviation depends on numerous process variables. In descending order of importance, the following process variables were found to be important in determining the width of the coating layer growth rate distribution: The location of the nozzle above the bed, the coating solution temperature, the coating solution spraying rate, the fluidisation air flow rate, the nozzle’s liquid orifice diameter, the particle exchange rate and finally, particle diameter.

Not all process variables were studied in this model sensitivity analysis. In Figure 4.72 an overview is given of all interactions that were found between the studied process variables and the different phenomena involved in the fluidised bed coating process, more specifically, the phenomena related to droplet migration and droplet/particle deposition. The process variables omitted in this chapter’s sensitivity analysis have been added as well, along with their likely interactions. Finally, some interactions which have not been modelled, but are presumably important in real coating applications are also indicated. More specifically, the effect of the release of atomisation air on the bed’s fluidisation behaviour was not yet modelled, but will be (partially) addressed to in Chapter 5.
Figure 4.72. An overview of the phenomena and their interactions (→), as stated in the droplet phase extended model description, involved in determining the different gas-, droplet- and solid-phase related properties. The interaction between the process variables (→, interaction present in the model; ←, probable interaction, but not present in the model) and the physical phenomena (at droplet level) is given (⊙, process variable present; ◐, process variable not present in the droplet phase extended model) as well.
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5. Modelling the dynamic two-dimensional axisymmetric
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5.1. Introduction

Chapters 3 and 4 focused on the development of a 2- and a 3-phase model, respectively. In these models, the fluidised bed was one-dimensionally discretised into flat, disc-like (or more specifically, truncated conically shaped) segments, as shown in Figure 5.1a. Although this approach gave satisfactory results, it also demonstrated some important shortcomings, most noticeably:

- As stated by Bellgardt (1985), the radial dispersion of particles in a fluidised bed is usually an order of magnitude smaller than the axial dispersion. Consequently, the radial temperature and moisture gradients of the fluidised bed can be larger than axial gradients. According to Peng and Fan (1997), particle movement in tapered fluidised beds operating at a gas velocity ranging between minimum fluidisation and the onset of turbulent fluidisation (cfr. Chapter 1) is more or less structured, with upward moving particles in the central region (core) of the bed and downward movement of particles in the bed’s annulus, which also has a lower voidage ($\varepsilon_{\text{bed}}$) than the bed’s core region.

- The bed’s coating volume, or the region where droplet/particle adhesion is established, was modelled as a predetermined or droplet submodel-simulated truncated cone-shaped volume stretching the entire diameter of the reactor. In reality, however, the coating volume is likely to be shaped like a narrow cone below the nozzle.

- When the pneumatic nozzle is positioned closely to the top of the fluidised bed or submerged into the bed, the large gas velocities near the nozzle due to the release of compressed air will effectively create a coherent void (or jet cavity) where particles are suppressed (Ariyapadi et al., 2003; 2005).

To remove these shortcomings in the existing fluidised bed coating model, it is obvious that two- or three-dimensional bed discretisation schemes are required instead of just one-dimensional axisymmetric portioning of the bed. To illustrate this, a 2D axisymmetric model has been drawn next to the existing 1D model in Figure 5.1. From Figure 5.1 it is clear that the axisymmetric model allows for the incorporation of phenomena such as jet cavitation, radial particle transport and a more accurately shaped coating volume which was not possible using the existing 1D models.

The first part of this Chapter consists of a detailed description of the development of the existing model into a 2D axisymmetric model. As will be demonstrated throughout the description of the modelling framework, in order to calibrate the 2D model, extensive data
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concerning particle mixing behaviour and gas flow during fluidisation is required. Whereas in
the 1D model, the transport of both phases was characterised by plug flow (gas phase) and
simple axial particle exchange, in the 2D model, both the radial and axial transport of both the
gas and solid phases have to be fully characterised. In the calibration of the 2D model, both
the particle mixing behaviour and gas flow could either be quantified using models – such as
hybrid Eulerian-Langragian CFD-models (see Chapter 2) – or through experiments (Werther,
1999). It should be stressed, however, that the focus of this chapter’s study is on presenting an
overall method (or framework) of simulating the fluidised bed coating process rather than
extensively modelling, including validation, of these component processes.

The second part of this Chapter presents some preliminary modelling results compared with
the experimental temperature and humidity distributions, which were retrieved using the same
experimental set-up as described in Chapter 4.

Figure 5.1. Comparison of the level of detail that can be incorporated in fluidised bed
cysting coating modelling using the 2D axisymmetric model (a) and its 1D counterpart
as detailed in Chapters 3 and 4 (b).
5.2. Model description

5.2.1. Model overview and assumptions

In order to develop the model, the fluidised bed was axisymmetrically discretised into \( n \times m \) control volumes, resulting in ring shaped control volumes. A detail of a single control volume is given in Figure 5.2. Throughout the simulation of a single coating process, each control volume had a constant size, given by

\[
V_{S}(i, j) = \frac{\pi \Delta h_{S}(i, j)}{3} \left( d_{S}^{2}(i, j) + d_{S}^{2}(i + 1, j) + d_{S}(i, j)d_{S}(i + 1, j) \right)
- \frac{\pi \Delta h_{S}(i, j)}{3} \left( d_{S}^{2}(i, j - 1) + d_{S}^{2}(i + 1, j - 1) + d_{S}(i, j - 1)d_{S}(i + 1, j - 1) \right)
\]

with \( \Delta h_{S}(i, j) = h_{S}(i + 1, j) - h_{S}(i, j) \) \hfill (5.1)

Within each control volume, three different phases were distinguished: the gas (air) phase, the solid phase (core particles) and the droplet phase. The solid phase mass per control volume, \( M_{pp}(i, j) \), remained constant throughout the simulated coating process. In case of a monodisperse particle population (no segregation nor agglomeration present) a constant mass of solid phase implies a constant number of particles per control volume, \( N_{p}(i, j) \). In its simplest form, the fluidised bed is represented by a single value for the bed voidage, \( \varepsilon_{bed} \). In reality, however, the (time-averaged) bed voidage fraction in a tapered fluidised bed depends on the location in the bed (Peng and Fan, 1997).

Assuming a constant bed voidage, the number of particles contained in a control volume \( S(i, j) \) is

\[
N_{p}(i, j) = \frac{6(1 - \varepsilon_{bed})V_{S}(i, j)}{\pi d_{p}^{3}} \hfill (5.2)
\]

All assumptions made under Section 3.2, regarding the basic gas/solid-model, and in Section 4.2.1, dealing with the modelling of the droplet phase extension, are still valid in this 2D axisymmetric model. Additionally, the following assumptions were added to the 2D model:

- One noticeable difference between the existing 1D model and the model proposed in this Chapter, is the possibility to define control volumes which only contain gas and/or droplet phase but no solids, or \( M_{pp}(i, j) = 0 \). This type of control volume is particularly useful in modelling the jet cavity in those cases where the pneumatic nozzle is positioned close to the bed’s surface or submerged into the fluidised bed.
- The droplet phase is assumed to migrate in one direction for both the axial and the radial component, i.e., the droplet phase in \( S(i, j) \) only migrates outwards to \( S(i, j + 1) \) or downwards to \( S(i - 1, j) \).
5.2.2. Heat and mass balances

Again, in each control volume $S(i, j)$, the dynamic heat and mass balances for air, particles and droplets were established. A detailed overview of the control volume’s constituting phases, the interactions between them and the variables used to characterise these phases is given in Figure 5.3.

To simplify the terms in the balance equations describing mass transfer between two adjacent control volumes, the flux terms ($G_a$ for the gas phase, $r$ for the solid phase and $J_{dp}$ for the droplet phase) were distributed over each boundary surface of the control volume. These terms were written with the subscript $k$ ($k = 1...4$) and the following convention was applied in assigning the boundary surfaces in the axisymmetric grid:

- $k = 1$ indicates the boundary surface between $S(i, j)$ and $S(i+1, j)$,
- $k = 2$ represents the boundary surface between $S(i, j)$ and $S(i, j+1)$,
- $k = 3$ indicates the boundary surface between $S(i-1, j)$ and $S(i, j)$ and finally,
- $k = 4$ represents the boundary surface between $S(i, j-1)$ and $S(i, j)$.

For example, $r_1(i, j)$ represents the particle exchange from $S(i, j)$ towards $S(i+1, j)$, while $r_3(i+1, j)$ represents the particle exchange from $S(i+1, j)$ towards $S(i, j)$. Certain calculations require the integration of fluxes over all the boundary surfaces of the control volume $S(i, j)$, therefore the operator $n_k(i)$ was defined. This operator returns the control volume indices at the $k^{th}$ boundary surface of $S(i, j)$, for example: $n_1(i, j) = (i + 1, j)$.
Figure 5.3. Detailed overview of a control volume used in the 2D-axisymmetric grid.

Remarks

* The solid phase (particles) can be absent (jet cavity control volumes)

** The heat loss terms and wall contact losses only apply for control volumes adjacent to the reactor wall or, $S(i, j); j - m$

*** $G_d(i, j)$ is either negative (inbound gas transport) or positive (outbound gas transport). In case of inbound transport, the gas properties ($T_d$ and $\varrho_d$) of the appropriate neighbouring control volume (source) have to be used
In each control volume, the heat and mass balance equations were obtained. Note that, in analogy with Chapters 3 and 4, the droplet and solid (particles) phases were at first represented as continuous phases and were treated similar to the gas phase. Later, the population balances for both particles and droplets were introduced. For the continuous model, the following heat and mass balance equations were obtained:

**Particle balance in a single control volume (solid phase continuity equation).** Under the assumption that the particle mass inside the control volume, \( M_{pp}(i, j) \), is constant, the solid phase (or particle) balance could be written as, with \( M_{pp,bed} \) being the total mass of particles in the fluidised bed:

\[
\frac{dM_{pp}(i, j)}{dt} = M_{pp,bed} \left( \sum_{k=1}^{4} r_k (n_k(i, j)) - \sum_{k=1}^{4} r_k(i, j) \right) = 0 \tag{5.3}
\]

Note the fundamental difference in modelling of the particle exchange between the 1D and 2D-axisymmetric discretised fluidised bed: In the 1D-model, particle flow was always symmetric, i.e., the amount of solid material travelling from \( S_i \) towards \( S_{i+1} \) was always equal to the amount travelling from \( S_{i+1} \) to \( S_i \). However, in the 2D-model, this symmetry is no longer necessary to have solid phase continuity \( (dM_{pp}/dt = 0) \) and consequently, preferential particle migration through the control volume (in structured particle flows) can be simulated as well.

**Air mass balance in a single control volume (gas phase continuity equation).** This equation describes the mass conservation of dry air in a single control volume:

\[
G_{at}(i, j) + \sum_{k=1}^{4} G_{a,k}(i, j) = 0 \tag{5.4}
\]

In Eq. (5.4) is \( G_{at}(i, j) \) the term describing the input of atomisation air into the modelled bed domain. Figure 5.4 illustrates in which cells the term \( G_{at}(i, j) \) is used to calculate the contribution of the atomisation air to the bed’s heat and mass balances. When the nozzle is suspended above the bed \( (h_{noz} > h_{bed}) \), the upper cells \( (S(i, j): j = m) \) – positioned within the nozzle jet of which the radius is calculated according to Eq. (4.26) – are considered to be the source cells for atomisation air. The exact mass flow rates \( G_{a,k}(i, j) \) are calculated by integrating Eq. (4.25) along the radial coordinate. In case of a nozzle submerged into the bed \( (h_{noz} < h_{bed}) \), then only the cell in which the nozzle is positioned is considered a source cell and \( G_{at}(i, j) = G_{at} \).
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Figure 5.4. Determination of source cells of atomisation air in case of (a) nozzle suspended above the bed, or (b) nozzle submerged into the fluidised bed.

Droplet mass balance in a single control volume (droplet phase continuity equation). Resulting from the second additional model assumption under 5.2.1, the droplet mass balance is given by the equation,

\[
\frac{dM_{dp}(i,j)}{dt} = J_{dp,1}(i,j) - \sum_{k=2}^{3} J_{dp,k}(i,j) - M_{ad}(i,j) - r_C(i,j)M_{pp}(i,j) - r_C^e(i,j)A_w(i,j) \tag{5.5}
\]

In Eq. (5.5) is \( J_{dp,k}(i,j) \) the droplet mass flow rate, in kg s\(^{-1}\), from or towards \( S(i,j) \) at the \( k \)th boundary surface. The term \( r_C(i,j) \) represents the droplet collection rate in the control volume \( S(i,j) \) expressed as kg droplet phase kg core\(^{-1}\) s\(^{-1}\), while the term \( M_{pp}(i,j) \) is the total mass of core material present in \( S(i,j) \). The last term in Eq. (5.5) represents the loss in droplets due to the adhesion on the reactor wall and has to be included only in the control volumes adjacent to the reactor wall, or \( S(i,j) \): \( j = m \).

Moisture balance of the particles. The change in particle moisture content, \( W_p(i,j) \), is given by

\[
\frac{dW_p(i,j)}{dt} = \frac{M_{pp,bed}(i,j)}{M_{pp}(i,j)} \left( \sum_{k=1}^{4} r_k(n_k(i,j))W_p(n_k(i,j)) - \sum_{k=1}^{4} r_k(i,j)W_p(i,j) - r_D(i,j) \right) + r_C(i,j)(1 - DM_{dp}(i,j)) \tag{5.6}
\]

The first two terms in Equation (5.6) represent the moisture addition and removal resulting from particle transport towards or from \( S(i,j) \), respectively. The drying term, \( r_D(i,j) \) and the droplet collection term, \( r_C(i,j) \) complement the particles’ moisture balance.

Moisture balance in the gas (air) phase. Before constructing the gas phase-related balance equations, it is important to stress that the gas transfer term, \( G_{ad,k}(i,j) \), could either be positive (gas output) or negative (gas input into the control volume). In building the balances, in-
outbound gas transport terms need to be clearly separated from each other. Therefore, an operator \( m_k \) was defined:

\[
S(i, j), k \in [1...4]: \begin{cases} 
G_{s,k}(i, j) \leq 0 \Rightarrow m_k(i, j) = 0 \\
G_{s,k}(i, j) > 0 \Rightarrow m_k(i, j) = 1
\end{cases} 
\] (5.7)

The resulting moisture balance for the air inside a single control volume is similar to Eq. (4.9), except for the gas exchange with neighbourin g cells and the contribution of the atomisation air, \( G_{at}(i, j) \), which is only non-zero in the source cells as defined in Figure 5.4:

\[
M_a(i, j) \frac{dX_a(i, j)}{dt} = r_d(i, j)M_{pp}(i, j) + r_d^*(i, j)M_{dp}(i, j) + r_d^w(i, j)A_w(i, j)
\]

\[
+ \sum_{k=1}^{4} (1-m_k(i, j))G_{s,k}(i, j)X_a(n_k(i, j))
\]

\[
- \sum_{k=1}^{4} m_k(i, j)G_{s,k}(i, j)X_a(i, j) + G_{at}(i, j)X_{at}
\] (5.8)

**Moisture balance of the droplet phase.** Assuming that the droplet phase migration is only limited to movement from \( S(i, j) \) towards \( S(i-1, j) \) and \( S(i, j+1) \) as shown in Figure 5.3, the moisture balance of the droplet phase is given by,

\[
\frac{d}{dt} \left[ M_{dp}(i, j)(1-DM_{dp}(i, j)) \right] = (1-DM_{dp}(i+1, j))J_{dp}(i, j)
\]

\[
+ (1-DM_{dp}(i, j-1))J_{dp-4}(i, j) - r_d(i, j)M_{dp}(i, j)
\]

\[
- (1-DM_{dp}(i, j)) \left( \sum_{k=2}^{3} J_{dp,k}(i, j) + r_c(i, j)M_{pp}(i, j) + r_c^w(i, j)A_w(i, j) \right)
\] (5.9)

The different terms in Eq. (5.9) describe the input of droplet phase from \( S(i+1, j) \) and \( S(i, j-1) \), removal of moisture due to evaporation at the droplet’s surface, removal of the droplet phase towards \( S(i, j+1) \) and \( S(i-1, j) \) and droplet collection either onto particles, \( r_c \), either on the inner wall of the reactor, \( r_c^w \).

**Coating mass balance of the particles.** The coating mass distribution is given by the variable \( Y_p \), expressed as mass unit of coating material per mass unit of core particles. Again, the only difference with Eq. (4.11) is the larger number of different particle exchanges with neighbouring volumes that have to be included:

\[
\frac{dY_p(i, j)}{dt} = \frac{M_{pp,bed}}{M_{pp}(i, j)} \left( \sum_{k=1}^{4} r_k(n_k(i, j))Y_p(n_k(i, j)) - \sum_{k=1}^{4} r_k(i, j)Y_p(i, j) \right)
\]

\[
+ r_c(i, j)DM_{dp}(i, j)
\] (5.10)
Coating mass balance of the droplets. Analogous to Eq. (5.9), the coating mass \((DM_{dp} \times M_{dp})\) balance for the droplets becomes

\[
\frac{d}{dt} \left(M_{dp}(i,j)DM_{dp}(i,j)\right) = DM_{dp}(i+1,j)J_{dp,1}(i,j) + DM_{dp}(i,j-1)J_{dp,4}(i,j) - DM_{dp}(i,j)\left(\sum_{k=2}^{4} J_{dp,k}(i,j) + r_c(i,j)M_{pp}(i,j) + r_{cw}(i,j)A_p(i,j)\right) - M_{sd}
\] (5.11)

Heat balance of the particles. This balance consists of the following terms: the heat carried by the exchanged particles \((\Phi_p)\), the convective heat transfer between particles and gas phase (radiative heat transfer was considered to be negligible, as stated in Chapter 3), \(\Phi_{a,p}\), the heat contributed by the collected droplets, \(\Phi_C\), heat removed by evaporation of deposited coating solution on the particle surface, \(\Phi_D\), and finally, direct heat losses through particle/wall-contact \(\Phi_{loss,p}\) or:

\[
M_{pp}(i,j)C_{pp}\frac{dT_{pl}}{dt} = \Phi_p(i,j) + \Phi_{a,p}(i,j) + \Phi_C(i,j) - \Phi_D(i,j) - \Phi_{loss,p}(i,j)
\] (5.12)

The different terms of Eq. (5.12) are given by

\[
\Phi_p(i,j) = M_{pp}(i,j)C_{pp}\left(\sum_{k=1}^{4} r_k(n_k(i,j))T_p(n_k(i,j)) - \sum_{k=1}^{4} r_k(i,j)T_p(i,j)\right)
\] (5.13)

\[
\Phi_{a,p}(i,j) = \alpha_p(i,j)A_{pp}(i,j)(T_p(i,j) - T_p(i,j))
\] (5.14)

\[
\Phi_D(i,j) = r_D(i,j)M_{pp}(i,j)Q_{wall}(i,j)
\] (5.15)

\[
\Phi_C(i,j) = r_c(i,j)M_{pp}(i,j)C_{pp}(i,j)(T_{dp}(i,j) - T_p(i,j))
\] (5.16)

The convective heat transfer coefficient, \(\alpha_p\), in Eq. (5.14), and the drying rate of the deposited coating solution on the particle surface, \(r_D\), in Eq. (5.15) were approximated using the dimensionless Nusselt and Sherwood numbers, discussed in Section 3.3.3. In Eq. (5.16) is \(T_{dp}(i,j)\) the temperature of the droplets in \(S(i,j)\) which, through simulation, was found to quickly approach the wet bulb temperature. The droplet collection rate, \(r_C\) in Eq. (5.16), was calculated by means of the droplet submodel (Chapter 4), which has been modified to cooperate with the 2D axisymmetric model. Finally, the calculation of the heat loss rate, \(\Phi_{loss,p}(i,j)\), was identical to the existing one-dimensional model as discussed in Section 3.3.2.

Heat balance of the gas phase (air). In both the droplet and particle heat balances, the internal heat transfer resistance was considered negligible, similar as in Section 3.3.1. The heat balance of the gas phase is given in Eq. (5.17) and consists of the following terms: the heat transported by the gas phase, which is a mixture of atomisation and fluidisation air, \(\Phi_a\), the
convective particle/gas heat transfer, \( \Phi_{a,p} \) given in Eq. (5.14), the convective droplet/gas heat transfer, \( \Phi_{a,dp} \), the heat required to bring the water vapour at the surface of particles and/or droplets to air temperature, \( \Phi_v \), and finally, the heat losses, \( \Phi_{loss,a} \), which were only taken into account in the outer control volumes \( (S(i,j) : j = m) \):

\[
M_{a}(i,j)C_{p,a}(i,j)\frac{dT_a(i,j)}{dt} = \Phi_a(i,j) - \Phi_{a,p}(i,j) - \Phi_{a,dp}(i,j) - \Phi_v(i,j) - \Phi_{loss,a}(i,j) \tag{5.17}
\]

With the different terms in Eq. (5.17) being,

\[
\Phi_a(i,j) = \sum_{k=1}^{4} \left( 1 - m_k(i,j) \right) G_{a,k}(i,j) C_{p,a}(n_k(i,j)) T_a(n_k(i,j)) - \sum_{k=1}^{4} m_k(i,j) G_{a,k}(i,j) C_{p,a}(i,j) T_a(i,j) \tag{5.18}
\]

\[
\Phi_{a,dp}(i,j) = \alpha_{dp}(i,j) A_{dp}(i,j) \left( T_a(i,j) - T_{dp}(i,j) \right) \tag{5.19}
\]

\[
\Phi_v(i,j) = C_{p,v}(i,j) r_D(i,j) M_{pp}(i,j) \left( T_a(i,j) - T_p(i,j) \right) + C_{p,v}(i,j) r_D^\infty(i,j) A_w(i,j) \left( T_a(i,j) - T_w(i,j) \right) + C_{p,v}(i,j) r_D(i,j) A_{dp}(i,j) \left( T_a(i,j) - T_{dp}(i,j) \right) \tag{5.20}
\]

Usually, the last two terms (\( \Phi_v \) and \( \Phi_{loss,a} \)) in Eq. (5.17) are relatively small compared to the remaining terms. The heat loss calculations were given in Section 3.3.2.

**Heat balance of the droplets.** The following heat transfers have to be included in the droplet phase heat balance: heat transport by the moving droplet phase (\( \Phi_{dp} \)), convective heat transfer between the droplets and the gas phase (\( \Phi_{a,dp} \)), heat transport by droplet collection (\( \Phi_C \)), evaporative heat transfer (\( \Phi_D^* \)) and heat removed by the formation and removal of spray dried fines (\( \Phi_{sd} \)):

\[
M_{dp}(i,j) C_{p,dp}(i,j) \frac{dT_{dp}(i,j)}{dt} = \Phi_{dp}(i,j) + \Phi_{a,dp}(i,j) - \Phi_C(i,j) - \Phi_{sd}(i,j) \tag{5.21}
\]

The heat transfer between gas and droplet phase (\( \Phi_{a,dp} \)) is given by Eq. (5.19), the heat transport by droplet removal from the droplet phase (\( \Phi_C \)) is given by Eq. (5.15). The remaining heat transfer terms of Eq. (5.21) are:


\[ \Phi_{dp}(i, j) = J_{dp, 1}(i + 1, j)C_{p,dp}(i + 1, j) + J_{dp, 4}(i, j - 1)C_{p,dp}(i, j - 1) \]
\[ - \sum_{k=2}^{3} J_{dp,k}(i, j)C_{p,dp}(i, j)T_{dp}(i, j) \]  \hfill (5.22)

\[ \Phi_{D}^{*}(i, j) = r_{D}(i, j)M_{dp}(i, j)[Q_{lat}(i, j) + C_{p,v}(i, j)(T_{s}(i, j) - T_{dp}(i, j))] \]  \hfill (5.23)

\[ \Phi_{sd}(i, j) = \dot{M}_{sd}(i, j)C_{p,c}(i, j)T_{sd}(i, j) \]  \hfill (5.24)

In Eq. (5.24) is \( T_{sd}(i, j) \) the temperature at which the spray dried fines are formed in the control volume \( S(i, j) \). It was assumed to equal the droplet phase temperature in that particular control volume, or

\[ T_{sd}(i, j) = T_{dp}(i, j) \]  \hfill (5.25)

*Heat balance of a wall element.* In Section 4.2.2.2, the heat balance was presented for a reactor wall element, which was considered a ring-shaped massive control volume characterised by a single temperature \( T_{w} \) for each ring-like control volume. The same heat balance equation as presented in Eq. (4.18) was also used, unmodified, in the 2D axisymmetric model presented in this chapter.
5.3. Model calibration

5.3.1. Introduction

As already stated in the introduction of this chapter, the model requires extensive calibration data concerning both the solid and gas phase movement during fluidisation. The study in this chapter primarily deals with the description of an overall framework for the simulation of the batch fluidised bed coating process, rather than providing detailed analysis – through modelling or experimentation – of the different processes (mixing, gas flow) during fluidisation. In order to be able to study and to validate the results predicted by means of the proposed modelling framework, two simplified models were proposed for the crude estimation of the required model calibration data.

5.3.2. Particle mixing behaviour

In the one-dimensional model proposed in Chapter 3, the particle mixing behaviour was characterised by the (axial) particle exchange rate, $r_i$. The two-dimensional axisymmetric model requires both axial, $r_1(i, j)$ and $r_3(i, j)$, and, $r_2(i, j)$ and $r_4(i, j)$, radial exchange rates. In its simplest form, both the axial and radial particle exchange could be assumed constant throughout the entire bed, or:

\[
\begin{align*}
\forall i \in [1...n], j \in [1...m]:
&
\begin{cases}
    r_1(i, j) = r_3(i, j) = c \\
    r_2(i, j) = r_4(i, j) = c'
\end{cases}
\end{align*}
\]

$\quad c, c' = \text{constant}$ (5.26)

Details of the calculation of the axial particle exchange rate (or dispersion coefficient) were given in Eqs. (3.51) and (3.52). According to Bellgardt (1985), the relationship between axial and radial exchange rates in fluidised beds could be generalised to:

\[
r_1 = r_3 = 10r_2 = 10r_4
\]

The same assumption considering particle mixing behaviour was made by Heinrich et al. (2003b) in modelling the fluidised bed granulation process. However, the main disadvantage of the assumptions under Eq. (5.27) is the inability to include structured particle movement into the model, which was suggested by Peng and Fan (1997). To solve this problem, particle exchange was predicted by means of a simplified particle trajectory (or Langrangian) model. A detailed description of this model is described in Appendix C.

5.3.3. Gas flow

The shift from the one-dimensional towards a two-dimensional axisymmetric model also requires deeper insight into the air flow patterns occurring inside the fluidised bed. The simple assumption of plug flow is no longer valid because of the addition of atomisation air to the
fluidisation air inside the bed. It its simplest form, the mixing of atomisation air is assumed to take place in a confined number of control volumes, an approach which was used in the one-dimensional models (i.e. homogeneously dividing $G_{at}$ over the upper $c$ control volumes). However, the increased spatial complexity of the model in combination with the lack of a-priori knowledge of the mixing behaviour of fluidisation and atomisation air, rule out this approach.

In order to calculate the gas flow rates in the bed with respect to the reactor geometry and the flow rates of the supplied gas (both fluidisation and atomisation air), different approaches are possible. For ‘simple’ fluid dynamic problems (such as free jets), a mathematical solution to the mass, energy and momentum conservation equations (i.e. Navier-Stokes equations) governing the fluid flow often exists. In case of more complex fluid dynamic problems (for example, more complex geometry), the use of numerical or computational fluid dynamics (CFD) is necessary. Because the latter method was beyond the scope of this research, the use of existing theoretical solutions was preferred as a method for rapid estimation of the flow field inside the reactor geometry. A detailed description of the theoretical solution to the gas flow, used in this study, is given in Appendix D.
5.4. Materials and methods

5.4.1. Model calculation procedures

5.4.1.1. Introduction

The model presented in Section 5.2 is a deterministic model in which both the droplet and solid phase (particles) were characterised by a single set of properties per control volume, i.e. a single value for droplet and solid phase temperatures, $T_{dp}$ and $T_p$, per control volume.

Similar to Section 3.4, the model was transformed into a population balance model, in particular with respect to the solid phase. As a result, the heat and mass balance equations of the solid phase in Section 5.2 (being Eqs. (5.3), (5.6), (5.10) and (5.12)) were replaced with the population balance equation (Eq. (3.31)) and the individual particle heat and mass balance equations. These individual particle heat and mass balances remain unaltered, regardless of the discretisation scheme (1D or 2D) used. Further details of the individual balances are given in Sections 3.4.2 (in the absence of the droplet phase) and 4.3 (including droplet phase interaction).

The integration of the droplet phase is not different from the method explained in Chapter 4: the droplet/gas phase and droplet/solid phase interactions are handled by the droplet submodel, which is described in detail in Section 4.3.

5.4.1.2. Main model

The largest difference between the 1D and 2D axisymmetric discretisation schemes is situated in the gas phase calculations, but not in the solid phase – individual particle balance equations are independent of the discretisation scheme used – nor the droplet phase, which is handled by the same droplet submodel as in Chapter 4. As a consequence, few changes were required to transform the general calculation procedure, as described in Appendix B, from its 1D into its 2D axisymmetric form. An overview of the calculation procedure is presented in Figure 5.5. Among the most noticeable differences between the 1D and 2D calculation procedures are the following:

- The heat and mass balance equations for each control volume within the 2D grid are solved applying the following sequence:
  $S(1,1) \rightarrow S(1,m), S(2,1) \rightarrow S(2,m), \ldots, S(n,1) \rightarrow S(n,m)$.
- Instead of just calculating the interaction terms regarding the droplet phase using the submodel (described in Section 4.3), prior to solving the heat and mass balance equations, the air and particle flow pattern need to be assessed. Because of the long calculation time required to simulate the Lagrangian submodel, the submodel is only calculated during the first simulation cycle or when dynamic simulations with changes in process conditions (such as a step input for $G_a$) are used. The assumption of a steady
state particle flow (or bed fluidisation behaviour) throughout the coating process is only valid if (a) the increase in particle size through deposition of coating mass is negligibly small and (b), the humidity of the fluidisation air is below 0.5 to avoid significant occurrence of liquid bridges during particle-particle contact (Schaafsma et al., 1999).

- Heat losses are only calculated in cells adjacent to the reactor wall \((S(i, j): j = m)\) instead of subtracting heat losses in any cell in the 1D model.
- Finally, exhaust air temperature and air humidity are calculated as,

\[
T_{a,\text{out}} = \frac{\sum_{j=1}^{m} G_{a,j}(n, j) C_{p,a}(n, j) T_e(n, j)}{\sum_{j=1}^{m} G_{a,j}(n, j) C_{p,a}(n, j)} \tag{5.28}
\]

\[
X_{a,\text{out}} = \frac{\sum_{j=1}^{m} X_a(n, j) G_{a,j}(n, j)}{\sum_{j=1}^{m} G_{a,j}(n, j)} \tag{5.29}
\]
5.4.2. Experimental validation

The model-predicted air temperature and air humidity distributions inside the fluidised bed during steady coating regime were compared with experimentally measured distributions of temperature and relative humidity. The experimental set-up has already been discussed in Section 4.5. The only difference with the experimental validation as described in Chapter 4 was the method of post-processing the temperature and humidity data measured with the scanning probe. As the raw data retrieved from the scanning probe were already spatially distributed on a 2D grid, integration by means Eq. (4.40) was no longer necessary.
5.5. Preliminary results and discussion

5.5.1. Introduction

The main objective of the model presented in this chapter is the prediction of dynamic, spatial temperature and humidity distributions of both the gas and solid phases as well as the calculation of the dynamic coating mass distributions during batch fluidised bed coating. The results will be compared against the experimental temperature and humidity measurements.

It is important to stress that the presented results are preliminary: First, the model has been calibrated with gas and solid phase flow data calculated by means of the simplified models, as described in Appendices C and D. Second, a thorough analysis on the effect of the number of particles ($N_{\text{sim}}$) or the number of control volumes used in the discretisation ($n \times m$) on the produced statistical error of the model-predicted output variables has not yet been performed, since we only want to illustrate the model in its current state.

5.5.2. Exchange rate submodel results

The different model parameters used in this submodel are summarised in Table 5.1. A total simulation time ($t'_{\text{sim}}$) of 10 seconds proved to be sufficient to reach steady state conditions as is shown in Figure 5.6. In Figure 5.6, the evolution of the modelled axial particle exchange rate at some selected heights ($h = 0.03, 0.06, 0.09$ and $0.12$ m) of the modelled reactor geometry is plotted. These axial particle exchange rates correspond to the number of particle crossings counted through the entire horizontal bed cross-sections at the aforementioned heights. Based on the observations in Figure 5.6, steady state particle movement was reached after approximately 5 seconds, while, a damped oscillatory transition regime was observed in the simulation between $t' = 0$ and $t' = 5$ seconds.
Table 5.1. Model and operational parameters of the Langragian model (Appendix C)

<table>
<thead>
<tr>
<th>Variable</th>
<th>Symbol</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Model parameters</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Number of simulated particles*</td>
<td>$N_{sim}$</td>
<td>-</td>
<td>58000</td>
</tr>
<tr>
<td>Simulated time</td>
<td>$t_{sim}$</td>
<td>s</td>
<td>10</td>
</tr>
<tr>
<td>Simulation time step</td>
<td>$\Delta t_{sim}$</td>
<td>s</td>
<td>0.0005</td>
</tr>
<tr>
<td>Fluidisation air properties</td>
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<td></td>
</tr>
<tr>
<td>Inlet air temperature</td>
<td>$T_{a,in}$</td>
<td>°C</td>
<td>70</td>
</tr>
<tr>
<td>Inlet air flow rate</td>
<td>$G_{a,in}$</td>
<td>kg s$^{-1}$</td>
<td>0.0214</td>
</tr>
<tr>
<td>Inlet air absolute humidity</td>
<td>$X_{a,in}$</td>
<td>kg kg$^{-1}$</td>
<td>0.00726</td>
</tr>
<tr>
<td>Bed dimensions</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bed height</td>
<td>$h_{bed}$</td>
<td>m</td>
<td>0.1</td>
</tr>
<tr>
<td>Reactor height</td>
<td>$h_{r}$</td>
<td>m</td>
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</tr>
<tr>
<td>Reactor bottom diameter</td>
<td>$d_{b}$</td>
<td>m</td>
<td>0.225</td>
</tr>
<tr>
<td>Reactor top diameter</td>
<td>$d_{t}$</td>
<td>m</td>
<td>0.45</td>
</tr>
<tr>
<td>Bed material (glass beads)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Overall mass</td>
<td>$M_{bed}$</td>
<td>kg</td>
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</tr>
<tr>
<td>Particle diameter</td>
<td>$d_{p}$</td>
<td>µm</td>
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</tr>
<tr>
<td>Particle sphericity</td>
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<td>1</td>
</tr>
<tr>
<td>Specific density</td>
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</tr>
<tr>
<td>Poisson’s ratio</td>
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<td>0.22</td>
</tr>
<tr>
<td>Elasticity modulus</td>
<td>$E_{p}$</td>
<td>Pa</td>
<td>$72 \times 10^{9}$</td>
</tr>
<tr>
<td>Wall material (stainless steel)</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Poisson’s ratio</td>
<td>$\nu_{p}$</td>
<td>-</td>
<td>0.31</td>
</tr>
<tr>
<td>Elasticity modulus</td>
<td>$E_{p}$</td>
<td>Pa</td>
<td>$193 \times 10^{9}$</td>
</tr>
</tbody>
</table>

Remarks:
*The number of particles to include in the simulation has been calculated using Eq. (C.12)
Modelling the dynamic two-dimensional axisymmetric temperature and humidity distributions

Figure 5.6. Simulated axial particle exchange rates for glass beads with a diameter of 365 μm at 0.03 m (■), 0.06 m (○), 0.09 m (▲) and 0.12 m (△) above the air distributor.

Figure 5.7a shows a plot of the simulated average particle velocities during steady state using the conditions mentioned in Table 5.1. A clear circular or spouted bed-like particle flow pattern was found, with upward moving particles in the core of the bed, while particles descend along the reactor walls. This spout-like particle motion pattern in tapered fluidised beds was postulated by Peng and Fan (1997) and has been experimentally verified by Schaafsma (2000) and Depypere (2005) using PEPT measurements.

The theoretical bed height, calculated using the equations set out in Section 3.6.2.2, was approximately 0.1 m. However, Figure 5.7a still shows that particles attain relatively large velocities above \( h = 0.1 \) m, which appears to contradict with the theoretical bed height of 0.1 m. Comparing the particle flow pattern with the axial particle exchange rates recorded during simulation (Figure 5.7b) shows that over 90% of all axial particle exchanges occur below 0.1 m (the theoretic bed height). Consequently, the recorded particle velocities above \( h = 0.1 \) m stem from a minor fraction of particles which surpass the fluidised bed in a spout-like manner.
Modelling the dynamic two-dimensional axisymmetric temperature and humidity distributions

Modelling heat and mass transfer in fluidised bed coating processes

Figure 5.7. Simulated spatial distribution of steady-state particle velocity in m s\(^{-1}\) (a) and steady-state axial particle exchange rates (in Hz) as a function of height above the air distributor (b).

Finally, the simulation was repeated with varying particle densities (\(\rho_a = 1400\) to 2600 kg m\(^{-3}\) with increments of 300 kg m\(^{-3}\)) and varying surplus gas velocities (\(v_a - v_{mf} = 0.0\) to 0.5 m s\(^{-1}\) with increments of 0.05 m s\(^{-1}\)) and verified against the findings of Mostoufi and Chaouki (2001). To enable comparison, the whole-bed axial particle exchange rate (or alternatively, the axial dispersion coefficient \((D_{p,ax})\) using Eq. (3.51)) was calculated as the average value of the model-predicted spatially distributed particle exchange rates. The results are plotted in Figure 5.8.

Figure 5.8. The model-predicted axial (or vertical) dispersion coefficients for particles with a diameter of \(d_p = 365\) µm, and as a function of surplus gas velocity (= \(v_a - v_{mf}\) in m s\(^{-1}\)) and particle density (\(\rho_a\) in kg m\(^{-3}\)).
As expected, a linear relationship was found between the whole-bed axial dispersion coefficient \( (D_{\text{p,ax}}) \) and the surplus gas velocity (Pallarès and Johnsson, 2006). Furthermore, the axial dispersion coefficient at a given \( v_a - v_{\text{mf}} \) was found to increases with the increase in particle density, due to the higher gas velocities \( (v_{\text{mf}}) \) and the lower bed heights \( (h_{\text{bed}}) \) in beds with increasing particle density (Limtrakul et al., 2005). Next, the axial dispersion coefficients were used to calculate the model-predicted dimensionless Peclet number of which the results are shown in Figure 5.9. Considering the experimental findings by Mostoufi and Chaouki (2001) regarding a Peclet number of 0.02 for sand particles with \( d_p = 385 \, \mu m \) and the model-predicted Peclet numbers between 0 and 0.014 within the range of surplus gas velocities examined, it was concluded that the model-predicted particle exchange rates (or dispersion coefficients) were within the range of the experimental vertical dispersion coefficients and were suited to be used in the overall model, whose results are given in Section 5.5.4.

**Figure 5.9.** The model-predicted dimensionless Peclet numbers for particles with a diameter of \( d_p = 365 \, \mu m \), and as a function of surplus gas velocity \( (= v_a - v_{\text{mf}} \) in \( \text{m s}^{-1} \)) and particle density \( (\rho_a \text{ in kg m}^{-3}) \).

### 5.5.3. Gas flow field calculation

Next to analysing the model-predicted particle flow inside the fluidised bed, the gas flow field was assessed using the model equations of Appendix D. Three different series of calculations were performed, where the atomisation air pressure, \( P_{\text{at}} \), nozzle position, \( h_{\text{noz}} \), and gas jet shape factor, \( s_{\text{at}} \), variables were varied. Table 5.2 lists the fixed variables while Table 5.3 summarises the different variable combinations used in each of these 3 series.
### Table 5.3. Fixed variables in the gas (air) flow field calculations

<table>
<thead>
<tr>
<th>Variable</th>
<th>Symbol</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Fluidisation air properties</strong></td>
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<td></td>
</tr>
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<td>Inlet air mass flow rate</td>
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<td>kg s$^{-1}$</td>
<td>0.0214</td>
</tr>
<tr>
<td>Inlet air temperature</td>
<td>$T_{a,in}$</td>
<td>°C</td>
<td>50</td>
</tr>
<tr>
<td>Inlet air absolute humidity</td>
<td>$X_{a,in}$</td>
<td>kg kg$^{-1}$</td>
<td>0.00726</td>
</tr>
<tr>
<td><strong>Bed dimensions</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bed height</td>
<td>$h_{bed}$</td>
<td>m</td>
<td>0.1</td>
</tr>
<tr>
<td>Reactor height</td>
<td>$h_r$</td>
<td>m</td>
<td>0.84</td>
</tr>
<tr>
<td>Reactor bottom diameter</td>
<td>$d_b$</td>
<td>m</td>
<td>0.225</td>
</tr>
<tr>
<td>Reactor top diameter</td>
<td>$d_t$</td>
<td>m</td>
<td>0.45</td>
</tr>
<tr>
<td><strong>Atomisation air properties</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Atomisation air temperature</td>
<td>$T_{at}$</td>
<td>°C</td>
<td>20</td>
</tr>
<tr>
<td>Atomisation air relative humidity</td>
<td>$\phi_{at}$</td>
<td></td>
<td>0.3</td>
</tr>
</tbody>
</table>

### Table 5.3. Modified variables in the gas (air) flow field calculations

<table>
<thead>
<tr>
<th>Series</th>
<th>$P_{at}$ (bar)</th>
<th>$G_{at}$ (kg s$^{-1}$)</th>
<th>$h_{noz}$ (m)</th>
<th>$s_{at}$ (m$^{2/3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Series I</td>
<td>0.5 – 1.5 – 2.5 – 3.5</td>
<td>$6.02 \times 10^4 – 1.45 \times 10^3 – 2.04 \times 10^{-3} – 2.38 \times 10^3$</td>
<td>0.100</td>
<td>0.05</td>
</tr>
<tr>
<td>Series II</td>
<td>2.5</td>
<td>$2.04 \times 10^3$</td>
<td>0.100 – 0.125 – 0.150 – 0.175</td>
<td>0.05</td>
</tr>
<tr>
<td>Series III</td>
<td>2.5</td>
<td>$2.04 \times 10^3$</td>
<td>0.100</td>
<td>0.04 – 0.05 – 0.06 – 0.07</td>
</tr>
</tbody>
</table>

*Remark: the atomisation air pressure ($P_{at}$) and atomisation air mass flow rate ($G_{at}$) are related*

In Figure 5.10, the model-predicted gas flow patterns in the central region of the bed ($0 \leq h \leq h_{bed} = 0.1 \text{m}$, $0 \leq r \leq 0.06 \text{m}$) are plotted against the atomisation air pressure (Series I). It is important to note that the equations describing the axial and radial gas velocity profiles, produced by a free jet (Appendix D), do not take into account the effects of the wall nor the porous gas distributor.

As expected, increasing the atomisation air pressure (related to the atomisation air flow rate) did not modify the gas flow pattern as such, but only resulted in increased (downward) gas velocities below the nozzle. Also, increasing the atomisation air pressure also resulted in larger amounts of upward moving fluidisation air being entrained into the jet produced by the nozzle. The entrainment of fluidisation air is clearly visible in the bed region adjacent to the nozzle ($h = 0.10 \text{ m}$), especially at larger atomisation air pressures. The significance of the downward moving gas flow is twofold. First, a significantly colder region in the core of the
bed is created, potentially resulting in agglomeration side effects. Second, the larger the gas velocities produced by the gas jet (nozzle) – in combination with the counter-current particle flow, – the lower the droplet adhesion efficiency resulting in an increased risk of spray drying and low-quality of the produced coating films.

![Figure 5.11.](image)

**Figure 5.11.** The (theoretical) gas velocity flow in the fluidised bed for varying atomisation pressures: (a) $P_{at} = 0.5$ bar, (b) $P_{at} = 1.5$ bar, (c) $P_{at} = 2.5$ bar and (d), $P_{at} = 3.5$ bar.

Figure 5.11. illustrates the effect of varying the nozzle height (Series II) on the gas velocity profiles with respect to the reactor region being used in the main model (being the lower 0.1 m of the reactor). Because of the relatively narrow shape of the jet ($s_{at} = 0.05$), the effect of the nozzle height on the gas flow pattern was limited. Increasing the nozzle height (with respect to the bed height) resulted in two noticeable effects: First, there was an overall reduction in downward gas velocities below the nozzle and second, entrainment of fluidisation air into the jet – as far as the bed region is concerned ($0 \leq h_{bed} \leq 0.1 m$) – was no
longer noticeable when the distance between the nozzle and the bed was increased above 0.025 m.

(a)  
(b)  
(c)  
(d)  

![Graphs depicting gas flow profiles](image)

**Figure 5.11.** The (theoretical) gas flow profiles in the fluidised bed when varying the nozzle height: (a) $h_{noz} = 0.100$ m, (b) $h_{noz} = 0.125$ m, (c) $h_{noz} = 0.150$ m and (d), $h_{noz} = 0.175$ m.

Finally, Figure 5.12 demonstrates the effect of jet width, expressed by means of the shape factor $s_{at}$ on the gas flow pattern (Series III). Widening the jet produced by the release of atomisation gas results in a more rapid deceleration of the atomisation air, which is likely to result in smaller temperature gradients inside the bed (due to the more ‘homogeneous’ distribution of the compressed air) and in a reduction of spray drying losses.
Modelling the dynamic two-dimensional axisymmetric temperature and humidity distributions

Figure 5.12. The (theoretical) gas flow profiles in the fluidised bed when varying the gas jet shape factor: (a) $s_{at} = 0.04 \text{ m}^{2/3}$, (b) $s_{at} = 0.05 \text{ m}^{2/3}$, (c) $s_{at} = 0.06 \text{ m}^{2/3}$ and (d), $s_{at} = 0.07 \text{ m}^{2/3}$.

5.5.4. Overall model results and validation

Table 5.4 summarises the different process conditions and model parameters used in the 2D model. The bed was discretised using a grid of $8 \times 8$ control volumes. For the initial conditions, both the temperature of all modelled particles and the air in each control volume were set to the inlet air temperature ($T_{a,in} = T_p = T_a(i, j)$). As such, the heating phase was not simulated to reduce calculation time. As shown in Figure 5.13, a simulated time of $t = 500 \text{ s}$ was sufficiently long to reach steady state conditions.
Table 5.4. Model and operational parameters of the simulation.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Symbol</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Main model parameters</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Control volumes</td>
<td>$m \times n$</td>
<td>-</td>
<td>8 \times 8</td>
</tr>
<tr>
<td>Number of simulated particles</td>
<td>$N_{sim}$</td>
<td>-</td>
<td>20016</td>
</tr>
<tr>
<td>Particle exchange rate</td>
<td>$r_k$</td>
<td>Hz</td>
<td>Appendix C</td>
</tr>
<tr>
<td><strong>Droplet sub model parameters</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Number of simulated droplets</td>
<td>$N_{dr,sim}$</td>
<td>-</td>
<td>5000</td>
</tr>
<tr>
<td>Droplet size model (atomisation)</td>
<td></td>
<td></td>
<td>Levebfre  (1988)</td>
</tr>
<tr>
<td><strong>Fluidisation air properties</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Inlet air temperature</td>
<td>$T_{a,in}$</td>
<td>°C</td>
<td>51.6**</td>
</tr>
<tr>
<td>Inlet air flow rate</td>
<td>$G_{a,in}$</td>
<td>kg s$^{-1}$</td>
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</tr>
<tr>
<td>Inlet air absolute humidity</td>
<td>$X_{a,in}$</td>
<td>kg kg$^{-1}$</td>
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</tr>
<tr>
<td><strong>Bed dimensions</strong></td>
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<td></td>
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<tr>
<td>Bed height</td>
<td>$h_{bed}$</td>
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<td>0.1</td>
</tr>
<tr>
<td>Reactor height</td>
<td>$h_r$</td>
<td>m</td>
<td>0.84</td>
</tr>
<tr>
<td>Reactor bottom diameter</td>
<td>$d_b$</td>
<td>m</td>
<td>0.225</td>
</tr>
<tr>
<td>Reactor top diameter</td>
<td>$d_t$</td>
<td>m</td>
<td>0.45</td>
</tr>
<tr>
<td>Nozzle tip height</td>
<td>$h_{noz}$</td>
<td>m</td>
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</tr>
<tr>
<td><strong>Bed material (glass beads)</strong></td>
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<td></td>
</tr>
<tr>
<td>Overall mass</td>
<td>$M_{bed}$</td>
<td>kg</td>
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<tr>
<td>Particle diameter</td>
<td>$d_p$</td>
<td>µm</td>
<td>365</td>
</tr>
<tr>
<td>Particle sphericity</td>
<td>$\psi_p$</td>
<td>-</td>
<td>1</td>
</tr>
<tr>
<td>Specific density</td>
<td>$\rho_p$</td>
<td>kg m$^{-3}$</td>
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<tr>
<td>Specific heat</td>
<td>$C_{p,p}$</td>
<td>J kg$^{-1}$K$^{-1}$</td>
<td>837</td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>$\lambda_p$</td>
<td>Wm$^{-1}$K$^{-1}$</td>
<td>0.8</td>
</tr>
<tr>
<td><strong>Coating solution properties</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Spraying rate</td>
<td>$\dot{M}_{sol}$</td>
<td>kg s$^{-1}$</td>
<td>0.605 \times 10$^{-4}$</td>
</tr>
<tr>
<td>Dry matter content</td>
<td>$DM_{sol}$</td>
<td>kg kg$^{-1}$</td>
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</tr>
<tr>
<td>Coating solution temperature</td>
<td>$T_{sol}$</td>
<td>°C</td>
<td>20</td>
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<tr>
<td>Coating solution viscosity</td>
<td>$\mu_{sol}$</td>
<td>Pa s</td>
<td>1.002 \times 10$^{-3}$</td>
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<tr>
<td>Coating solution surface tension</td>
<td>$\gamma_{sol}$</td>
<td>N m$^{-1}$</td>
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</tr>
<tr>
<td>Particle/coating solution contact angle</td>
<td>$\theta_{dr/p}$</td>
<td>°</td>
<td>14</td>
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<tr>
<td><strong>Atomisation air properties</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Atomisation air mass flow rate</td>
<td>$G_{at}$</td>
<td>kg s$^{-1}$</td>
<td>1.45 \times 10$^{-3}$</td>
</tr>
<tr>
<td>Atomisation air pressure</td>
<td>$P_{at}$</td>
<td>bar</td>
<td>1.5</td>
</tr>
<tr>
<td>Atomisation air temperature</td>
<td>$T_{at}$</td>
<td>°C</td>
<td>20</td>
</tr>
<tr>
<td>Atomisation air relative humidity</td>
<td>$\Phi_{at}$</td>
<td>-</td>
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<tr>
<td>Shape factor of atomisation air jet</td>
<td>$s_{at}$</td>
<td>m$^{2/3}$</td>
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### Table 5.4. Model and operational parameters of the simulation (continued).

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<thead>
<tr>
<th>Variable</th>
<th>Symbol</th>
<th>Unit</th>
<th>Value</th>
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<tbody>
<tr>
<td><strong>External air (atmospheric) conditions</strong></td>
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<td></td>
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<tr>
<td>External air temperature</td>
<td>$T_e$</td>
<td>°C</td>
<td>20</td>
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<tr>
<td>External air relative humidity</td>
<td>$\varphi_e$</td>
<td>-</td>
<td>0.6</td>
</tr>
<tr>
<td>Atmospheric pressure</td>
<td>$P_e$</td>
<td>Pa</td>
<td>101325</td>
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<tr>
<td><strong>Other parameters</strong></td>
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<tr>
<td>Reactor wall thickness</td>
<td>$d_w$</td>
<td>m</td>
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</tr>
<tr>
<td>Reactor wall thermal conductivity</td>
<td>$\lambda_w$</td>
<td>Wm$^{-1}$K$^{-1}$</td>
<td>14.6</td>
</tr>
<tr>
<td>Reactor wall emittance for far-infrared radiation</td>
<td>$\varepsilon'_w$</td>
<td>-</td>
<td>0.16</td>
</tr>
<tr>
<td>Reactor wall specific heat</td>
<td>$C_{p,w}$</td>
<td>J kg$^{-1}$K$^{-1}$</td>
<td>500</td>
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<tr>
<td>Reactor wall density</td>
<td>$\rho_w$</td>
<td>kg m$^{-3}$</td>
<td>7850</td>
</tr>
<tr>
<td>Wall mixing constant</td>
<td>$\kappa_w$</td>
<td>-</td>
<td>0.05</td>
</tr>
<tr>
<td>Bed voidage at minimum fluidisation velocity</td>
<td>$\varepsilon_{mf}$</td>
<td>-</td>
<td>0.39</td>
</tr>
<tr>
<td>Bubble frequency</td>
<td>$f_{bu}$</td>
<td>Hz</td>
<td>8900</td>
</tr>
<tr>
<td><strong>Simulation (main model) parameters</strong></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>Simulated time</td>
<td>$t_{sim}$</td>
<td>s</td>
<td>500</td>
</tr>
<tr>
<td>Simulation time step</td>
<td>$\Delta t_{sim}$</td>
<td>s</td>
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</tr>
<tr>
<td><strong>Simulation (droplet submodel) parameters</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum simulation time (single trajectory)</td>
<td>$t_{dr,sim}$</td>
<td>s</td>
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</tr>
<tr>
<td>Maximum time step*</td>
<td>Max($\Delta t_{dr,sim}$)</td>
<td>s</td>
<td>$2.5 \times 10^{-5}$</td>
</tr>
<tr>
<td>Minimum time step*</td>
<td>Min($\Delta t_{dr,sim}$)</td>
<td>s</td>
<td>$1.33 \times 10^{-6}$</td>
</tr>
<tr>
<td>Maximum tolerable droplet temperature difference*</td>
<td>Max($\Delta T_{de}$)</td>
<td>°C</td>
<td>1.0</td>
</tr>
<tr>
<td>Minimum tolerable droplet temperature difference*</td>
<td>Min($\Delta T_{de}$)</td>
<td>°C</td>
<td>$1.0 \times 10^{-4}$</td>
</tr>
<tr>
<td>Maximum tolerable droplet displacement*</td>
<td>Max($\Delta x_{de}$)</td>
<td>m</td>
<td>$2.0 \times 10^{-4}$</td>
</tr>
<tr>
<td>Minimum tolerable droplet displacement*</td>
<td>Min($\Delta x_{de}$)</td>
<td>m</td>
<td>$5.0 \times 10^{-4}$</td>
</tr>
</tbody>
</table>

**Remarks:**

* Relating to the adaptive time stepping algorithm in the simulation of the droplet submodel. These parameters are obtained through trial-and-error, where values are selected yielding sufficiently fast calculations, while the rejection due to uncontrollable oscillations is kept below 5%.

** $T_{a,in} = 51.6 \, ^\circ C$ was used instead of $T_{a,in} = 50 \, ^\circ C$ to allow comparison with experimentally recorded bed temperatures and humidities. In the experimental set-up, as described in Chapter 4, the fluidisation air’s temperature – as measured in the plenum below the gas distributor - was equal to 51.6 °C when the set point temperature was set to 50 °C on the PID-controller (reproducible systematic error in the temperature controller).
Figure 5.13. Model-predicted outlet air temperature ($T_{a,\text{out}}$ in °C) as a function of time. Initial particle and air temperature were set to $T_{a,\text{in}} = 51.6$ °C.

Considering the small diameter of the core particles, the drying rate – which is expressed as the amount of water evaporated per unit of time, per mass unit of core particles – of the wetted particles was relatively fast. Consequently, significantly higher air humidities and lower temperatures were predicted in the control volumes where both droplets and particles coexist (region below the nozzle) compared to the control volumes containing only particles (Figures 5.14a and 5.14b). When the model-predicted air temperatures were compared with the particle temperatures (Figure 5.14c), it was observed that the low air temperature region, corresponding to the spraying region, extended less axially in the bed, but more radially towards the reactor wall. This can be explained by the fact that particles in a conically shaped fluidised bed have a circular spout-like movement: in the centre particles are lifted upward and travel counter-current with the sprayed droplets while near the reactor wall, particles fall back towards the air distributor.

In Chapters 3 and 4, the fluidised bed coating model has proven to reliably predict the overall thermodynamic operation conditions (such as outlet air properties or average bed temperature). However, when comparing the experimentally measured axial temperature profiles with the model-predicted ones (Section 4.5), some difficulties were encountered. More specifically, the model-predicted axial temperature gradients were more linear in nature than the measured temperature gradients. Based on the model results presented in Figure 5.14, the radial air temperature and humidity gradients appear to be more pronounced than the axial gradients. The lack of radial gradients in temperature and humidity of both phases in the 1D model also explains the difficulties encountered in Section 4.5 when comparing the 1D model-predicted results against the experimentally measured temperature distribution.
Modelling the dynamic two-dimensional axisymmetric temperature and humidity distributions

Figure 5.14. Model-predicted results of a steady-state fluidised bed of 2 kg glass beads with 3.63 g/min water spraying rate. The results show the 2D spatial distribution inside a vertical half-plane (nozzle at \( r = 0, h = 0.16 \)) of air temperature (a), air relative humidity (b) and particle temperature (c).

Figure 5.15a and b show the results of the measured steady-state air temperature and air relative humidity using a scanning probe, but without processing into 1D axial profiles. The
same conditions apply as those used in the model-predicted results as summarised in Table 5.4. Considering the humidity (Figure 5.15b), both experimental and model-predicted distributions roughly correspond, although higher air humidity was predicted on the central axis. Also, the model-predicted high air humidity region tends to protrude deeper into the fluidised bed. When considering the temperature distribution (Figure 5.15a), the low temperature region below the nozzle could not be distinguished. Instead, a more constant vertical temperature gradient, regardless of the radial position, was observed. One possible explanation for the absence of this low temperature region in the measured profiles is that the used temperature probe in the scanning probe had a non-negligible thermal inertia and therefore, had a rather slow responsiveness to changing air temperatures.

**Figure 5.15.** Experimental results of a steady-state fluidised bed of 2 kg glass beads with 3.63 g/min water spraying rate. The results show the 2D spatial distribution, interpolated from a 10×10 grid, inside a vertical half-plane (nozzle at \( r = 0 \) m, \( h = 0.16 \) m) of air temperature (a) and air relative humidity (b).
5.6. Conclusions

In this chapter, a modelling framework was provided for the top-spray fluidised bed coating process based on a 2D axisymmetric discretisation of the bed. The aim of this model was to provide a solution to some important physical phenomena which were absent or neglected in the current 1D model, including structured particle movement, radial temperature and humidity gradients, and a correct shape of the coating region of the fluidised bed.

The required calibration data – for both particle and flow patterns – was provided using a simplified Langrangian model and the theoretical gas profiles produced by a free jet, respectively. In future studies, these models could be replaced within the current fluidised bed coating model framework by more complete and coupled Eulerian (gas flow) and Langrangian (solids flow) models.

Preliminary results using the presented model, combined with the experimentally measured air temperature and humidity spatial distributions inside the fluidised bed during coating, have proven that the presented model is quite reliable in predicted both axial and radial gradients of thermodynamic properties of liquid-sprayed fluidised beds.
Modelling the dynamic two-dimensional axisymmetric temperature and humidity distributions

Modelling heat and mass transfer in fluidised bed coating processes
6. General conclusions

Microencapsulation of food ingredients and additives enables the food technologist to select products with improved or totally new properties. Microencapsulation, or the process of applying polymer walls or coatings to either solid, liquid or even gaseous materials, could be achieved with varying techniques, including fluidised bed coating. Although fluidised bed coating has its roots in the pharmaceutical industry, it has found widespread use, including in the food industry. Although both industries share the techniques for the production of microencapsulated ingredients, there is a large difference in the economics of their markets. Whereas the pharmaceutical industry is characterised by low volume production and high profit margins, the food industry has to handle high capacity, low profit margin products. Therefore, the food technologist is obliged to cut production costs and to further explore opportunities to optimise the coating processes in fluidised bed.

To optimise the fluidised bed coating process, process models should be considered an important tool. First of all, there is currently a lack of knowledge on how all the different heat and mass transfer processes interact in combination with the apparent erratic behaviour of the bubbling fluidised bed. Furthermore, the quantitative link between the fluidised bed coating mechanisms and the occurrence of side-effects and/or the quality of the encapsulated product is not yet fully understood. Process models could not only help in clarifying these ‘missing links’, but could also be applied in advanced process control strategies or in scale-up studies.

A brief overview has been given covering the existing models applicable in fluidised bed coating. Current models for coating could be classified among three classes: black-box, white-box and grey-box models. This model classification is based on the extent to which physical phenomena are incorporated into the model, as opposed to relying on empirical equations that match the process input/output data. However, given the complex nature of fluidised beds, pure white-box models have to be considered hypothetical. Even the most elaborate models, such as hybrid eulerian-langrangian models still rely on empirical equations.

In a first approach, a one-dimensional model was developed in which the fluidised bed reactor was horizontally descretised into several layers or control volumes. Whereas existing lumped-region models make the distinction between 2 (spraying and non-spraying) to 3 (spraying, drying and heat transfer) perfectly mixed regions (or layers) in the liquid-sprayed fluidised bed, the proposed model consisted of a significantly higher number of layers (>10) to allow improved implementation of the axial particle dispersion behaviour into the model. In each layer, the heat and mass balances for the gas phase were constructed, along with the population balance for the particles (or solid phase). The particles were characterised by four independent variables, namely temperature, surface moisture content, coating mass content and the location (layer) in which the particle resides. Furthermore, the model is based on the
empirical equations for heat and mass transfer between the solid and gas phases, as well as particle exchange (mixing) in bubbling fluidised beds, and takes heat losses into account.

Once the proposed model has been experimentally validated using steady state measurements of the outlet air temperature during batch coating processes, performed in a Glatt GPCG-1 unit and was found to achieve similar or even improved accuracy compared to existing black-box thermodynamic models for batch fluidised bed coating, the model was used to assess the impact of different input variables on the dynamics of the coating process. The variables, having the largest effect on the fluidised bed thermodynamic properties were found to be, in descending order: the inlet air temperature, the inlet air flow rate and the spraying rate. The effect of these variables is clearly visible in the outlet air thermodynamic properties and in both the thermal and vaporisation efficiency.

Because of their impact on the coating thermodynamics, these variables are commonly used in process control strategies. Although the spraying rate does not necessarily have the highest effect for the thermodynamic operation point of the fluidised bed, it is one of the variables of choice in terms of controllability and responsiveness in a real fluidised bed coating system as opposed to control by means of the inlet air temperature, which has a slower response time, or by means of the inlet air flow rate, which is constrained due to its relationship with the bed’s fluidisation characteristics.

Furthermore, the particle exchange rate and the relative size of the spraying (coating) region were found to play a major role in determining the particle property distributions. For instance, increasing the particle exchange rate or increasing the size of the spraying region with regard to the total bed volume, resulted in a more narrow coating mass distribution. However, these variables have no impact whatsoever on the outlet air properties, but do alter the temperature and humidity gradients in the bed. Hence, these variables could be referred to as ‘internal’ variables: they are hard to measure in-situ, but have a strong influence in determining coating mass uniformity and particle moisture content distribution – and consequently, the agglomeration tendency of the liquid-sprayed fluidised bed.

Internal variables such as the particle exchange rate and the size of the spraying region are controllable, but only to a certain extent. The problem in adjusting the internal variables is how they are restricted by their relationship to the bed’s thermodynamics. For instance, the particle exchange rate or the bed’s mixing behaviour could be adjusted by the fluidisation air flow rate. But at the same time, changing the air flow rate also impacts the evaporative capacity of the bed. On the other hand, modifying the reactor geometry by the addition of baffles or draft tubes appears to be a plausible strategy in controlling the particle exchange rate without affecting the bed’s thermodynamic operation point. However, the introduction of additional reactor complexity neutralises one of the main benefits of top-spray coating,
namely that of the simple reactor configuration, which allows much larger reactor volumes compared to the inherently more complex Wurster or tangential coaters.

The modelling results also revealed that the use of pneumatic nozzles in the coating process results in the introduction of additional (compressed) air into the bed. Consequently, the impact atomisation air properties on the fluidised bed’s thermodynamics could not be neglected, especially in small coating units, where the ratio of fluidisation air to atomisation air is relatively small.

Although the proposed model proved to predict the overall thermodynamic behaviour of the fluidised bed coating it also had some disadvantages. First of all, the model required prior knowledge of the size of the spraying (coating) region in the bed. Sensitivity analysis revealed that this variable is of paramount importance to the particle-related properties. Furthermore, in dynamic coating processes, it is quite likely that the size of the spraying region is not constant. Another shortcoming of the proposed model was the assumption that the coating solution was uniformly divided over all particles residing in the spraying region of the bed. In reality however, the coating solution is discontinuously being distributed through droplet-particles adhesion. Whether the droplet concentration in the spray decreases linear (as assumed in the proposed model) or non-linear is unknown.

Therefore, an extension to the current model was proposed which included the addition of the third phase – more specifically, the droplet phase – to the model. By introducing the additional heat and mass transfers between the droplet phase on the one hand, and the gas and solid phases on the other hand, the migratory behaviour of the spray could be simulated. Furthermore, by extending the model with the droplet phase, the premature droplet evaporation occurring between droplet production at the nozzle and impact on the fluidised particles could also be quantified.

The extended model was validated in two ways: First, the model-predicted temperature and humidity profiles of the bed’s air were compared with experimentally retrieved thermodynamic profiles during steady state batch fluidised bed coating. To that end, a purpose-built fluidised bed unit was used, combined with a moving probe measurement system. Comparing model-predicted and experimental air temperature profiles gave rise to mixed results. Usually, the outlet air thermodynamic properties were correctly predicted, but, depending on the inlet air temperature and fluidisation gas flow rate, the temperature gradient inside the bed was often difficult to predict. Subsequent analysis revealed that this problem was both related to the measurement method in multiphase flows or to difficulties in estimating the particle exchange rates, which were still required as model input variables. Second, spray drying losses were compared against values found in literature. Even though effects such as attrition were still not integrated into the extended model, the model was found to be capable of predicted spray drying losses.
The extended model also allowed performing sensitivity analysis with regard to the occurrence of spray drying losses, the size of the spraying region in the bed and the coating layer growth rate distribution. Within the group of variables studied, the position of the nozzle above the bed, the temperature of the coating solution, the inlet air flow rate, the atomisation air temperature, the spraying rate and the viscosity of the coating solution were found to be the most influencing variables on the occurrence of premature droplet evaporation. The same variables, excluding the inlet air flow rate, were also found to have a major impact on the size of the spraying region. One important conclusion from the sensitivity analysis is the fact that many of the involved microprocesses potentially counteract each other. For instance, increasing the atomisation air pressure yields smaller droplets which should result in larger spray drying losses, but at the same time, the temperature in the spraying region is reduced due to the mixing of process air with larger volumes of atomisation air, resulting in a reduced drying rate.

Finally, an attempt was made to further improve the model as one of the main drawbacks in both the basic and the extended one-dimensional models is the way the fluidised bed is divided into horizontal layers or control volumes. Because radial temperature or humidity gradients could not be modelled using the one-dimensional model, a framework was provided to model the coating process based on a two-dimensional axisymmetric discretisation of the bed. Besides radial temperature and/or concentration gradients, ordered particle movement and a more correct (jet-like or cone-like) shape of the spraying (or coating) region could be included in the model as opposed to the existing one-dimensional model. However, moving from one to two-dimensional models requires additional calibration data, including both the particle and gas flow patterns.

Preliminary results using two simple submodels to provide the necessary calibration data, were compared with the experimentally measured air temperature and humidity spatial distributions inside the fluidised bed during steady state coating. The results have shown that the presented model is quite reliable in predicting both axial and radial gradients of the thermodynamic properties of liquid-sprayed fluidised beds and that the difficulties encountered in validating the one-dimensional model using the experimental temperature distributions were probably the result of neglecting radial temperature and humidity gradients.

To conclude, throughout this research work, a new modelling methodology has been presented whose main benefit – compared to existing coating models – is the scale and the level of detail included in the models. To be more specific, the proposed models are able to combine macro-scale thermodynamic bed behaviour, including temperature and humidity distributions with the behaviour of the particles to be coated, which were modelled as a population in which properties such as coating mass, moisture content and temperature are statistically distributed. It is exactly this connection between macro-scale bed behaviour and
micro-scale particle behaviour that allows these models to be used in a number of applications.

First of all, once the two-dimensional axisymmetric models are coupled with more elaborate models to provide the necessary calibration data – such as fluid dynamic models to calculate the flow field of the fluidising gas or discrete element models to provide particle trajectories and mixing behaviour – the proposed two-dimensional model is suitable to study the effect of different reactor geometries on yield and process efficiency. Second, it is well known that fluidised bed coating processes do not scale linearly. Although the proposed models have not been tested in their ability to be extrapolated to larger reactor geometries, the grey-box nature and the degree to which actual physical phenomena – as opposed to existing black-box models – should allow the models proposed in this dissertation to be applicable in future scale-up studies. Finally, these models also could provide a basis for advanced process control strategies. The major obstacle in applying these models for process control is their numerical complexity which does not allow the simulation to be calculated in real-time during a single coating process. Perhaps future research into model simplification or into the development of more sophisticated numerical solving schemes – as opposed to the presented solving method which is quite ‘brute force’ in its use of computational resources – could allow future use of these models in fluidised bed coating process control with the ultimate aim of increasing process efficiency and reducing operational costs.
Modelling heat and mass transfer in fluidised bed coating processes
Samenvatting

Micro-encapsulatie van levensmiddelen- en -additieven stelt de voedings-technoloog in staat te kiezen uit een breed scala van producten met verbeterde of zelfs nieuwe eigenschappen in vergelijking met niet-geïncapsuleerde ingrediënten of additieven. Micro-encapsulatie kan gedefinieerd worden als een proces waarbij vaste, vloeibare of zelfs gasvormige bestanddelen voorzien worden van een omhullende polymeerlaag. Verscheidene technieken waren reeds ontwikkeld voor het verwezenlijken van micro-encapsulatie, inclusief wervelbedomhulling. Hoewel wervelbedomhulling zijn oorsprong kent in de farmaceutische industrie, is de techniek vandaag de dag wijdverspreid, tevens in de voedingsindustrie. Hoewel de farmaceutische en de voedingsindustrie dezelfde technieken delen voor de productie van omhulde ingrediënten en actieve componenten, is er een groot verschil in de economie van de afzetmarkten die ze van producten voorzien. Terwijl in de farmaceutische industrie de productie gekenmerkt wordt door een beperkte capaciteit en de producten kunnen afgezet worden met hoge winstmarges, worden levensmiddelen gekenmerkt door hoge productievolumes, terwijl de winstmarges beduidend lager liggen. Omwille van die reden zal de voedingstechnoloog verplicht zijn om productiekosten te drukken en verder nieuwe mogelijkheden moeten aanboren voor verdere optimalisatie van het wervelbed-omhullingsproces.

De ontwikkeling van modellen voor het voorspellen van het thermodynamische gedrag kan beschouwd worden als een belangrijk instrument in het optimaliseren – en uiteindelijk, het onderdrukken van de kosten – van wervelbedomhullingsprocessen. Wanneer het omhullingsprocess, uitgevoerd in een wervelbed, in detail wordt bestudeerd, stelt men vast dat het basisprincipe bestaat uit een aantal complexe warmte- en massaoverdrachtsprocessen tussen een drietal fasen bestaande uit een vloeistoffase (de vernevelde oplossing van het wandmateriaal), een vaste fase (het poeder) en een gasfase (opgewarmde lucht). Wanneer daarenboven rekening wordt gehouden met het schijnbaar onvoorspelbaar gedrag van wervelbedden, komt men tot het besluit dat er momenteel een gebrek is aan kennis die de verschillende deelprocessen met elkaar verbindt. Hoewel de kennis van ieder afzonderlijk deelproces (druppelvorming, botsing van druppels en poederdeeltjes, warmteoverdracht, enz...) reeds behoorlijk uitgebreid is, is de relatie tussen de samenwerking van deze deelprocessen en het uiteindelijke optreden van ongewenste nevenreacties, zoals sproeidrogen of agglomeratie, en de relatie met de kwaliteit van de gevormde omhulde materialen, verre van opgehelderd. Het opstellen van procesmodellen kunnen een bijdrage leveren in het ophelderen van deze 'missing links’. Bovendien kunnen modellen gebruikt worden in geavanceerde strategieën voor processturing en in het opschalen van het omhullingsproces van labo- tot industriële grootte.
Een kort overzicht werd geschetst van de bestaande en gedocumenteerde modellen toepasbaar voor wervelbedomhulling. Naargelang de mate waarin de fysische mechanismen van het eigenlijke proces worden opgenomen in het model en de mate waarin gebruik gemaakt wordt van empirische correlaties, wordt een onderscheid gemaakt tussen zgn. black-box, white-box en grey-box modellen. Het bestaan van white-box modellen, specifiek voor omhullingsprocessen in wervelbed, is in feite puur hypothetisch. Zelfs de meest uitgebreide modellen, zoals de hybride Euler-Lagrange modellen, moeten nog steeds gebruik maken van, zij het beperkt, empirische vergelijkingen.

In een eerste benadering, werd een één-dimensionaal model ontwikkeld waarin het wervelbed werd opgedeeld in verscheidene lagen of controlevolumes. In bestaande modellen wordt meestal de opdeling van het bed gemaakt in ofwel twee (een sproei en een niet-sproei zone), ofwel drie (een sproei-, droog- en warmteoverdrachtszone) ideaal gemengde lagen. In het voorgestelde model wordt de opsplittings verder uitgewerkt naar een beduidend groter aantal lagen (> 10) met het doel de mengeigenschappen en de deeltjesdispersie in het wervelbed beter te kunnen integreren in het model. In iedere laag werden de balansen voor het behoud van warmte en massa opgesteld, alsook de populatiebalans voor de deeltjes (de vaste fase). De deeltjes werden gekarakteriseerd door vier onafhankelijke veranderlijken: de deeltjestemperatuur, de vochtconcentratie aan het oppervlak, de concentratie aan wandmateriaal en de locatie (laag of controlevolume) van de deeltje. Bovendien is het model gebaseerd op empirische correlaties die aangetroffen werden in de literatuur voor de beschrijving van warmte- en massaoverdrachtsnelheden tussen de gas- en vaste fase in een wervelbed, alsmede ook voor de beschrijving van deeltjesdispersie. Tenslotte werden ook de warmteverliezen geïntegreerd in het model.

Het voorgestelde model werd proefondervindelijk gevalideerd, gebruik makende van de luchttemperatuur van de uitlaat van een Glatt GPCG-1 laboratorium wervelbedeenheid. Er werd tot de bevinding gekomen dat het voorgestelde model gelijk of zelfs beter presteerde dan de bestaande black-box modellen wat betreft de accuraatheid. Vervolgens werd het effect van verandering in een aantal van de ingangsvariabelen op het voorspelde thermodynamisch gedrag van het wervelbed, bestudeerd. De variabelen die schijnbaar de grootste invloed hadden op het thermodynamisch gedrag van het bed waren, in volgorde van afnemend belang, de inlaatluchttemperatuur, de vochtconcentratie aan het oppervlak, de concentratie aan wandmateriaal en de locatie van de deeltjes. Bovendien is het model gebaseerd op empirische correlaties die aangetroffen werden in de literatuur voor de beschrijving van warmte- en massaoverdrachtsnelheden tussen de gas- en vaste fase in een wervelbed.

Omwille van hun invloed op het thermodynamisch gedrag van het wervelbed, worden deze variabelen geregeld gebruikt in strategieën tot processsturing. Hoewel het pompdebiet niet onmiddellijk de grootste invloed heeft, blijkt het één van de uitgesproken variabelen te zijn in de keuze van regelvariabele. De redenen hiertoe zijn het gemak van instelbaarheid en de
snelle respons van het wervelbed op veranderingen in het pompdebiet en dit in tegenstelling tot de inlaatluchttemperatuur, die een veel tragere responstijd heeft, of de toevoersnelheid van de fluidisatielucht, die beperkt instelbaar is omwille van het verband met de fluidisatie-eigenschappen van het bed.

Bovendien werd tot de bevinding gekomen dat de uitwisselingssnelheid van de deeltjes en de verhouding van het volume van de sproeizone tot het volume van het bed een belangrijke rol spelen in het bepalen van de eigenschappen die gerelateerd zijn met de deeltjes. Bijvoorbeeld, het verhogen van de uitwisselingssnelheid van de deeltjes of het vergroten van de sproeizone (in vergelijking met het volume van het wervelbed) gaf aanleiding tot een nauwere verdeling van het wandmateriaal over de deeltjespopulatie. Niettegenstaande deze variabelen geen enkele invloed vertonen op de thermodynamische eigenschappen van de uitlaatlucht, hebben ze een invloed op de temperatuurs- en luchtvochtigheidsgradienten in het bed. Omwille van deze redenen worden deze variabelen aangeduid als ‘interne’ variabelen: ze zijn behoorlijk moeilijk om te meten of te kwantificeren, maar hebben een sterke invloed in het bepalen van de uniformiteit van de verdeling van het wandmateriaal of de neiging tot het agglomereren van de bevochtigde deeltjes.

De inwendige variabelen zijn maar tot op zekere hoogte te sturen. Het probleem situeert zich in het feit dat de interne variabelen, zoals deeltjesuitwisselingssnelheden en de relatieve grootte van de sproeizone, tevens verband houden met het thermodynamische gedrag van het wervelbed. Bijvoorbeeld, uitwisseling van deeltjes kan verhoogd worden door de toevoer van fluidisatielucht te verhogen. Tegelijkertijd, echter, zal hiermee ook de droogcapaciteit van het bed toenemen waardoor ongewenst sproeidrogen van de coatingvloeistof kan optreden. Een andere mogelijkheid om de uitwisselingssnelheden van de deeltjes te doen toenemen of om de beweging van deeltjes op een beter geordende manier tot stand te brengen, is het aanpassen van de reactorgeometrie met tussenschotten of kanalen. Echter, de toename in complexiteit van de reactorgeometrie doet een van de belangrijkste voordelen van top-spray reactoren – in vergelijking met reactoren van het Wurster of tangentiële type – teniet, met name dat zijn simpele constructie toelaat veel grotere reactorvolumes te ontwerpen zodoende de kosten te drukken.

Uit de simulatieresultaten kwam het belang van het gebruik van pneumatische spuitkoppen voor het vernevelen van de coatingvloeistof tot uitting. De afgifte van koudere en drogere perslucht in het bed resulteerde in een niet te verwaarlozen effect op de temperatuurs- en vochtverdeling in het wervelbed. Dit effect kwam des te meer tot uiting in laboratorium schaal wervelbedeenheden, waar de verhouding van de toevoer van atomisatielucht tot de toevoer van fluidisatielucht betrekkelijk groot is.

Hoewel het voorgestelde model bewezen heeft om het globale thermodynamische gedrag van een wervelbedomhullingsproces accuraat te voorspellen, zijn er ook een aantal nadelen.
verboden aan het model. Ten eerste, vereist het model een opgegeven waarde voor de volumeverhouding van de sproeizone tot het feitelijke bed. In de eerdere gevoeligheidsanalyse werd aangetoond dat dit een essentiële factor is in het bepalen van de deeltjes-gerelateerde eigenschappen. Bovendien werd in het bestaande model verondersteld dat deze volumeverhouding constant is gedurende het omhullingsproces. Echter, wijzigingen in de droogcapaciteit van het bed of in de samenstelling van de coatingvloeistof kunnen aanleiding geven tot gewijzigde grootte van de sproeizone gedurende het omhullingsproces. Ten tweede werd verondersteld dat de coatingvloeistof uniform over ieder deeltje dat zich in de sproeizone bevindt, verdeeld wordt. In de praktijk is er eerder een discontinue verdeling van de sproeivloeistof onder de vorm van druppels. Het is echter nog de vraag of de druppelconcentratie in de nevel al dan niet lineair afneemt naarmate de sproeinevel dieper in het bed doordringt.

Om deze nadelen van het bestaande model te verhelpen, werd een uitbreiding van het model voorgesteld. Deze uitbreiding bestond voornamelijk uit de toevoeging van de nevel als een volwaardige derde fase in het model. Door het toevoegen van warmte- en massaoverdracht tussen de vloeistoffase (nevel) enerzijds, en de gas- en vaste fase anderzijds, is het mogelijk om de migratie van de gesproeide nevel en de adsorptie aan de gefluidiseerde deeltjes te modelleren. Een bijkomend voordeel van dit type van uitgebreid model, is dat tevens de spreiing van de druppels met coatingvloeistof tijdens hun traject van de spuitkop naar het oppervlak van de gefluidiseerde deeltjes – mee voorspeld kan worden.

Het uitgebreide model werd op twee manieren experimenteel gevalideerd: Ten eerste werd een vergelijking gemaakt tussen de één-dimensionale temperatuursprofielen enerzijds afkomstig uit experimentele metingen, anderzijds bekomen door voorspelling met behulp van het model. Om de experimentele meting van temperatuurs- en vochtdistributies in een wervelbed mogelijk te maken, werd een aparte pilootschaal wervelbedinstallatie ontworpen. Deze werd voorzien van een beweegbare probe die de lokale luchttemperatuur en – vochtigheid kon meten in het bed. De vergelijking van de gemeten met de voorspelde luchttemperatuursprofielen bleek niet altijd zonder problemen. In de meeste gevallen werd een correcte uitlaatluchttemperatuur voorspeld, maar in een aantal gevallen – afhankelijk van het gekozen debiet en de temperatuur van de fluidisatielucht – werd de temperatuursgradient nogal foutief voorspeld. Verdere analyse toonde aan dat een aantal problemen aan de basis lagen: enerzijds bleek het moeilijk om uitsluitend de luchttemperatuur te meten in wervelbedden waar tevens een vloeistof werd neergetikt, zonder interferentie te hebben van de deeltjes en de rondzwevende druppels in de eigenlijke luchttemperatuursmeting. Anderzijds bleek het moeilijk om op een correcte manier de deeltjesuitwisselingssnelheden te berekenen voor het aanvangen van de simulatie. Zoals reeds gebleken is, heeft het menggedrag van de deeltjes in het wervelbed een belangrijk effect op de temperatuursgradienten in het bed. Een tweede validatiemethode bestond in het kwantitief vergelijken van de gemodelleerde
sproeidroogverliezen met experimentele waarden uit de literatuur. Hier bleek een goede overeenkomst te bestaan tussen model en experiment.

Ook op het uitgebreide model werd een gevoeligheidsanalyse uitgevoerd, ditmaal met betrekking tot het optreden van sproeidroogverliezen, de grootte van de sproeizone en de groeisnelheid van de afgezette laag van wandmateriaal tijdens het proces. De gevoeligheidsanalyse leverde als resultaat op dat de volgende ingangsvariabelen, in afnemend belang, significant bijdragen tot ongewenst sproeidrogen van de coatingvloeistof: de hoogte van de spuitkop boven het bed, de temperatuur waarmee de coatingvloeistof verneveld wordt in de reactor, het toevoerdebiet van de fluidisatielucht, het pompdebiet van de coatingvloeistof en tenslotte, de viscositeit van de coatingvloeistof. Dezelfde variabelen, behalve het toevoerdebiet van de fluidisatie, bleken in sterke mate de grootte van de sproeizone te bepalen in het wervelbed. Eén belangrijk besluit dat werd getrokken uit de gevoeligheidsanalyse is dat veel van de betrokken deelprocessen mekaar kunnen tegenwerken bij wijziging van de ingangsvariabelen, waardoor de relatie tussen ingangsvariabelen en processparameters enerzijds, en de uitgangsvariabelen anderzijds, niet-lineair is.

In het laatste deel werd een poging ondernomen om het bestaande (uitgebreide) model nog verder te verbeteren door de omvorming van een één-dimensionaal naar een axisymmetrisch twee-dimensionaal model. Eén van de problemen geassocieerd met één-dimensionale modellen is dat de radiale temperatuurs- of concentratiegradienten worden verwaarloosd. Bovendien, hebben studies reeds aangetoond dat in kegelvormige wervelbedden, de beweging van deeltjes niet volledig willekeurig is, zoals verondersteld in diffusie, maar eerder circulair. Het toevoegen van een deze geordende deeltjesbeweging vereist de overgang naar modellen met een hogere dimensie (>1). Tenslotte, wordt in het één-dimensionale model, de sproeizone gemodelleerd als één of meerder vlakke lagen. In werkelijkheid zal het spuitpatroon kegelvormig zijn, hetgeen niet geïmplementeerd kan worden in een één-dimensionaal model.


Om te besluiten, werd er in dit onderzoekswerk een nieuwe modelleringsmethode voorgesteld met als grootste voordeel – ten opzichte van bestaande modellen voor wervelbedomhulling –
de schaal en het niveau aan detail dat aan het model werd toegevoegd. Om meer specifiek te zijn, de voorgestelde model zijn in staat om het macroschaal thermodynamisch gedrag van het wervelbed (met onder andere, de temperatuurs- en vochtverdeling in het wervelbed) te combineren met het gedrag van de gesuspendeerde deeltjes. De partikels werden gemodelleerd als een populatie waarin eigenschappen zoals hoeveelheid aan coating, vochtconcentratie en temperatuur werden voorgesteld als statische distributies. Het is precies deze verbinding tussen de macroschaal van het wervelbed en de microschaal van de deeltjes dat toelaat om deze voorgestelde modellen te gebruiken in aantal toepassingen.

Ten eerste, eens de tweedimensionale modellen gekoppeld worden met meer uitgediepte modellen om te voorzien in de noodzakelijke calibratiegegevens – zoals de numerieke stromingsmodellen voor het berekenen van de luchtstromingen en ‘discrete element’-modellen om de trajectoriën van de deeltjes te berekenen – zijn deze tweedimensionale modellen geschikt om het effect van verschillende reactorgeometrieën op procesopbrengst en efficiëntie te bestuderen. Ten tweede, het is welbekend dat wervelbedprocessen niet lineair opschalen. Hoewel de voorgestelde modellen niet werden getest in hun vermogen om te worden geëxtrapoleerd naar grotere reactorgeometrieën, zouden ze moeten toegepast kunnen worden in opschalingsstudies gezien de mate waarmee fysische fenomenen die optreden tijdens wervelbedomhulling in rekening werden gebracht. Ten slotte kunnen deze modellen tevens de basis vormen voor het uitwerken van geavanceerde strategieën in procesregeling. Het grootste probleem van deze modellen is de huidige lange rekentijd waardoor deze niet geschikt zijn om te worden berekend in ‘real-time’ gedurende een omhullingproces. Vandaar dat toekomstig onderzoek naar de ontwikkeling van meer gesofisticeerde rekenschema’s – in tegenstelling tot de voorgestelde rekenmethode die vooral gesteund is op het benutten van ‘brute kracht’ van de huidige computerhardware – een mogelijkheid biedt om deze modellen toe te passen in het sturen van wervelbedomhullingsprocessen om zo uiteindelijk de procesefficiëntie te verbeteren en de werkingskosten te verlagen.
7. Nomenclature

\( A, A' \) Surface area, m\(^2\)

\( a \) Acceleration, m s\(^{-2}\)

\( a' \) Constant used in the calculation of droplet impingement efficiency

\( a'' \) Constant used in the empirical Nusselt or Reynolds number correlations

\( Ar \) Archimedes number, dimensionless

\( B' \) Birth process

\( b' \) Constant used in the calculation of droplet impingement efficiency

\( Bi \) Biot number, dimensionless

\( C \) Concentration, kg m\(^{-3}\)

\( c \) Number of coating control volumes, dimensionless

\( C_D \) Drag force coefficient (corrected for fluidised beds), dimensionless

\( C_D' \) Drag force coefficient (standard), dimensionless

\( C_p \) Specific heat at constant pressure, J kg\(^{-1}\)K\(^{-1}\)

\( D \) Diffusion or dispersion coefficient in, m\(^2\)s\(^{-1}\)

\( D' \) Decay process

\( d \) Diameter or thickness, m

\( d' \) Characteristic dimension, m

\( DM \) Spraying solution dry matter content, dimensionless

\( E \) Elasticity modulus, Pa

\( E(\ ) \) Particle enumerator operator

\( e \) Unit vector

\( F \) Force (N)

\( f \) Frequency, Hz
\( G \) Air mass flow, kg dry air s\(^{-1}\)

\( g \) Gravitational constant, = 9.81 m s\(^{-2}\)

\( Gr \) Grasshof number, dimensionless

\( h, h' \) Height, m

\( I \) Number of cycles, dimensionless

\( J \) Droplet phase mass flow rate, kg s\(^{-1}\)

\( k \) Linear spring coefficient, N m\(^{-1}\)

\( M \) Mass, kg

\( m \) Correction factor for the drag force coefficient, dimensionless

\( m' \) Mass flux, kg m\(^{-2}\)s\(^{-1}\)

\( m'' \) Constant used in the empirical Nusselt number correlations

\( m_k(\cdot) \) 2D axisymmetric model specific operator, returns whether gas transport is inbound (0) or outbound (1) at the \( k^{th} \) boundary surface of \( S(i, j) \)

\( \dot{M} \) Mass flow rate, kg s\(^{-1}\)

\( MW \) Molecular weight, kg mol\(^{-1}\)

\( N \ or \ N' \) Number of particles (in the overall model and the Langrangian submodel, respectively)

\( n_k(\cdot) \) 2D axisymmetric model specific operator, returns the index of the control volume adjacent to the \( k^{th} \) boundary surface of \( S(i, j) \)

\( n \) Number of control volumes

\( Nu \) Nusselt number, dimensionless

\( O(\cdot) \) Algorithm complexity

\( P \) Pressure, Pa

\( P(\cdot) \) Probability

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\( p \) Population density function, dimensionless

\( Pe \) Peclet number, dimensionless

\( Pr \) Prandtl number, dimensionless

\( Q \) Heat, J

\( q \) Heat flux, W m\(^{-2}\)

\( R \) Thermal resistance, m\(^2\)K W\(^{-1}\)

\( R' \) Thermal resistance (external), m\(^2\)K W\(^{-1}\)

\( R^* \) Universal gas constant, = 8.314 J mol\(^{-1}\)K\(^{-1}\)

\( r \) Particle transfer rate, s\(^{-1}\)

\( r_c \) Droplet collection rate, kg solution kg core\(^{1}\)s\(^{-1}\)

\( r_c^* \) Droplet collection rate at reactor wall, kg solution m\(^2\)s\(^{-1}\)

\( r_d \) Drying rate, kg water kg particle\(^{-1}\)s\(^{-1}\)

\( r_d^* \) Droplet drying rate, kg water s\(^{-1}\) kg droplet\(^{-1}\)

\( r_d^* \) Droplet drying rate at reactor wall, kg water m\(^2\)s\(^{-1}\)

\( Re \) Reynolds number, dimensionless

\( S \) Control volume

\( s \) Shape factor, dimensionless

\( s(\ ) \) Standard deviation

\( Sc \) Schmidt number, dimensionless

\( Sh \) Sherwood number, dimensionless

\( SSR \) Sum of squared residuals

\( St \) Stokes number, dimensionless

\( T \) Temperature, K

\( t, t' \) Time, s

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$U (0,1)$ Random number, uniformly distributed between 0 and 1

$V$ Volume, $m^3$

$v$ Linear velocity, $m s^{-1}$

$W$ Particle moisture content, $kg$ water $kg$ particle$^{-1}$

$We$ Weber number, dimensionless

$X$ Absolute humidity, $kg$ water $kg$ dry air$^{-1}$

$x$ Internal coordinate in population balance equation, or rectangular $x$-coordinate

$Y$ Particle coating mass, $kg$ DM $kg$ particle$^{-1}$

$y$ Rectangular $y$-coordinate

$Z$ Integer number

$z$ Rectangular $z$-coordinate

$z'$ Binary value

**Greek symbols**

$\alpha$ Convective heat transfer coefficient, $W m^{-2}K^{-1}$

$\alpha'$ Mass transfer coefficient, $m s^{-1}$

$\beta$ Volume expansion coefficient, $K^{-1}$

$\gamma$ Surface tension, $N m^{-1}$

$\delta$ Coating growth rate, $m s^{-1}$

$\delta'$ Laminar boundary layer thickness, $m$

$\Delta t, \Delta t'$ Time step, $s$

$\varepsilon$ Porosity or voidage, dimensionless

$\varepsilon'$ Emissivity, dimensionless

*Modelling heat and mass transfer in fluidised bed coating processes*
Nomenclature

\begin{align*}
\zeta & \quad \text{Droplet collection efficiency, dimensionless} \\
\eta & \quad \text{Efficiency} \\
\theta & \quad \text{Contact angle} \\
\iota & \quad \text{Droplet adhesion probability, dimensionless} \\
\kappa & \quad \text{Mixing constant} \\
\lambda & \quad \text{Heat transfer coefficient, W m}^{-1}\text{K}^{-1} \\
\lambda' & \quad \text{Heat transfer coefficient for stagnant gas in the emulsion phase, W m}^{-1}\text{K}^{-1} \\
\Lambda(\cdot) & \quad \text{Control volume membership} \\
\mu & \quad \text{Viscosity, Pa s} \\
\mu' & \quad \text{Viscosity at particle surface, Pa s} \\
\xi & \quad \text{Dimensionless radial coordinate} \\
\pi & \quad \text{Constant, } = 3.1415926535 \\
\rho & \quad \text{Mass density, kg m}^{-3} \\
\varsigma & \quad \text{Adhesion probability} \\
\sigma & \quad \text{Stefan-Boltzmann constant, } = 5.669 \times 10^{-8} \text{ W m}^{-2}\text{K}^{-4} \\
\tau & \quad \text{Equivalent gas film thickness around particle, dimensionless} \\
\upsilon & \quad \text{Contact angle} \\
\varphi & \quad \text{Relative humidity, dimensionless} \\
\Phi & \quad \text{Heat transfer rate, J s}^{-1} \\
\Phi' & \quad \text{Mass transfer rate, Kg s}^{-1} \\
\chi & \quad \text{Impingement efficiency, dimensionless} \\
\psi & \quad \text{Sphericity, dimensionless} \\
\omega & \quad \text{Input variable (sensitivity analysis)} \\
\Omega & \quad \text{Output variable (sensitivity analysis)}
\end{align*}
Superscripts and subscripts

a  air
at atomisation air
aw air at the wall region
ax axial
b bottom
bed bed
bu bubble
bouy bouyancy
c coating
coal coalescence
circ circulation
cond conduction
conv convection
ct contact
d drag
dmd dry matter to disperse
dp droplet phase
dr single droplet
drc single droplet at the moment of particle-droplet collision
e external
eq equivalent
f fluidum
g gas distributor
<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>eq</td>
<td>equivalent</td>
</tr>
<tr>
<td>exp</td>
<td>experimental</td>
</tr>
<tr>
<td>ew</td>
<td>particle gas film at wall region</td>
</tr>
<tr>
<td>im</td>
<td>impact</td>
</tr>
<tr>
<td>in</td>
<td>input</td>
</tr>
<tr>
<td>lat</td>
<td>latent heat of evaporation</td>
</tr>
<tr>
<td>loss</td>
<td>loss</td>
</tr>
<tr>
<td>mb</td>
<td>minimum bubbling</td>
</tr>
<tr>
<td>mf</td>
<td>minimum fluidisation</td>
</tr>
<tr>
<td>N</td>
<td>normal</td>
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<tr>
<td>or</td>
<td>orifice</td>
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<tr>
<td>out</td>
<td>output</td>
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<tr>
<td>p</td>
<td>particle</td>
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<td>particle emulsion packet</td>
</tr>
<tr>
<td>pcd</td>
<td>particle conduction</td>
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<td>particle population</td>
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<td>reactor</td>
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<td>rad</td>
<td>radiation</td>
</tr>
<tr>
<td>rd</td>
<td>radial</td>
</tr>
<tr>
<td>real</td>
<td>referring to real (physical) system being simulated</td>
</tr>
<tr>
<td>sat</td>
<td>saturated</td>
</tr>
<tr>
<td>sd</td>
<td>spray dried</td>
</tr>
<tr>
<td>sim</td>
<td>simulated</td>
</tr>
<tr>
<td>sol</td>
<td>coating solution</td>
</tr>
<tr>
<td>surf</td>
<td>surface</td>
</tr>
</tbody>
</table>
T  tangential

t  top

te  terminal

th  thermal

tu  turbine or centrifugal fan

v  water vapour

vap  vaporisation

w  wall
8. References


References


Modelling heat and mass transfer in fluidised bed coating processes


References


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References


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References


Modelling heat and mass transfer in fluidised bed coating processes
References


*Modelling heat and mass transfer in fluidised bed coating processes*
Modelling heat and mass transfer in fluidised bed coating processes
Appendix A. Random number generator selection

A.1. Commonly used types pseudo-random number generator algorithms

A.1.1. Linear congruential generators

Probably the most commonly used random number generators are linear congruential generators (or LCGs) (Knuth, 1997). This class of algorithms is based on the following integer recursion (Vattulainen et al., 1995; Knuth 1997):

\[ Z_{i+1} = (aZ_i + b) \mod m \]  

(A.1)

The LCG, as shown in Eq. (A.1) is characterised by 4 parameters,

- The modulus, \( m > 0 \)
- The multiplier, \( a \) with \( 0 \leq a < m \)
- The increment, \( b \) with \( 0 \leq b < m \)
- The starting value or seed, \( Z_0 \) with \( 0 \leq Z_0 < m \)

The recursion in Eq. (A.1) produces a sequence of integer numbers, \( Z_i \) in the interval \([0, m]\). Uniformly distributed random numbers in the interval \([0, 1]\) are obtained by the normalisation (Hellekalek, 1998):

\[ x_i = \frac{Z_i}{m} \]  

(A.2)

The maximum length of the period in the sequence of pseudo-random numbers, generated by a LGC, is equal to \( m \). It is also important to note that only few combinations of the parameters \((m, a, b, Z_0)\) yield uniformly distributed pseudo-random numbers. For instance, the RNG implemented in the Ansi-C programming language or \texttt{RAND} uses the parameter combination \((2^{31}, 1103515245, 12345, 12345)\) (Hellekalek, 1998) and the Visual Basic 6.0 RNG or \texttt{RND} uses the combination \((2^{24}, 1140671485, 12820163, 327680)\) (Microsoft knowledge base article Q231847, 2004). This implies that the \texttt{RND} algorithm has a rather short period of \(2^{24} - 2 = 16.7 \times 10^6\). Compared to the average ‘consumption’ of random numbers in a typical simulation being higher with 2 to 3 magnitude orders, this standard available RNG in Visual Basic proved inadequate.
A.1.2. Multiple recursive congruential generators and Fibonacci-type generators

One way to increase the period of the linear congruential algorithm, is to increase the order of the linear recursion and thus, the multiple recursive congruential generator (or MRG) is obtained (Vattulainen et al., 1995; Hellekalek, 1998):

\[ Z_i = (a_1 Z_{i-1} + a_2 Z_{i-2} + \ldots + a_p Z_{i-p}) \mod m \]  

(A.3)

Again normalisation is required, as in Eq. (A.2) to obtain random numbers in the unit interval [0,1]. The maximum period of this generator is \( m^p - 1 \), which is significantly longer compared to a simple LCG. However, the calculation of higher order MRGs, as shown in Eq. (A.3), also results in longer calculation times compared to simple LCGs. Consequently, MRGs are less attractive for large-scale Monte Carlo simulations. One of the most simple forms of the MRG is when \( p = 2 \) and \( a_1 = a_2 = 1 \), resulting in

\[ Z_i = (Z_{i-1} + Z_{i-2}) \mod m \]  

(A.4)

Equation (A.4) is a special type of MRG, called a Fibonacci generator. However, in most cases, Fibonacci generators do not use the two previous samples \( (Z_{i-1} \text{ and } Z_{i-2}) \) to generate \( Z_i \) but use older samples to improve RNG quality as the pseudo-random numbers generated by the recursion in Eq. (A.4) are considered to be deficient in quality. Hence this class is called lagged Fibonacci generators (or simply LFG):

\[ Z_i = (Z_{i-r} + Z_{i-s}) \mod m \text{ for } r \leq s \]  

(A.5)

If the modulus \( m \) could be represented as \( m = 2^b \), then the maximum attainable period with appropriate selection of the parameters \( r \) and \( s \) in a LFG-type generator is equal to \( 2^{b-1} (2^r - 1) \) (Mascagni and Srinivasan, 2004).

A.1.3. Feedback shift register (FSR) generators

A third important class of random number generators is the group of feedback shift register generators (FSR) or also called Tausworthe generators. As opposed to the previously discussed classes of RNGs, the FSR generators operate using binary variables or digits. The generalised FSR algorithm (or GFSR) is based on the theory of primitive trinomials of the form \( z^n + z^m + 1 \). Given such a primitive trinomial and \( p \) binary digits from \( z'_0 \) to \( z'_{p-1} \), a binary shift register sequence can be generated by the following recursion (Makino and Miyamura, 1995; Vattulainen et al., 1995):

\[ z'_i = z'_{i+p} \oplus z'_{i+p+q} \]  

(A.6)
In Equation (A.6), the symbol $\oplus$ represents the bitwise exclusive-or (XOR) operator, which returns a binary ‘1’ only if one of both binary inputs is equal to 1. Mathematically, the XOR operator is equivalent to addition modulo 2:

$$z'_i \oplus z'_j = (z'_i + z'_j) \mod 2 \tag{A.7}$$

The longest attainable period of this generator is equal to $2^p - 1$ and can only be achieved when $z'^n + z'^n + 1$ is a primitive trinomial, and if this trinomial divides $x^n - 1$ for $n = 2^p - 1$, for the smallest value of $n$ (Vattulainen et al., 1995). These conditions could be met by choosing $n$ to be a Mersenne prime, a Mersenne prime being a prime that could be written in the form:

$$n = 2^p - 1 \tag{A.8}$$

In order to generate 32-bit integers, the recursion in Eq. (A.6) is parallelised into vectors comprising 32 digits, which are being updated in parallel each time a new vector (or 32-bit integer) is generated (Thisted, 1988).

To further increase the quality of GFSR generators, a modified recursion of Eq. (A.6) was proposed by Matsumoto and Kurita (1992):

$$z'_{i+q} = z'_{i+p} \oplus z'_i A \tag{A.9}$$

In Eq. (A.9), $A$ represents a $w \times w$ matrix with binary entries. This type of generator is also called twisted GFSR (or tGFSR). The improvement of adding the ‘twist’ (or the multiplication with the binary matrix $A$) lies in the longest attainable period of $2^w p - 1$ instead of $2^p - 1$ for non-twisted GFSRs. One recent and highly promising variant of tGFSR generators is the Mersenne Twister RNG, proposed by Matsumoto and Nishimura (1998). This generator has an extremely long period of $2^{19937}-1$, which is a Mersenne prime.

### A.1.4. Other generators

In order to mitigate structural flaws found in pseudo-random number generators, two or more RNGs from different classes could be combined. Combination of different RNGs might also increase its period. Typical examples of combined RNG include Marsaglia’s KISS with a period of $2^{123}$ (Marsaglia, 2003) and recently Marsaglia proposed the generator MWC1038, which has a period of $3056868392^{3216} - 1$ (Thisted, 2005). Other techniques to improve RNG quality (for instance, removing the serial correlation between two consecutive samples) include the application of shuffling algorithms to the output of a RNG algorithm, such as the Bays-Durham shuffle algorithm (Klimasaukas, 2002).
A.2. Testing the pseudo-random number generator

A.2.1. Introduction

An important common characteristic of these different classes of pseudo-random number generators, is that these algorithms are all based on rather simple recursive equations with carefully selected parameters. The question that one now has to ask is whether these ‘hidden’ correlations affect the statistical properties of the RNG’s output and ultimately, whether the inherent structural flaws of deterministic RNGs might affect the results in a Monte Carlo simulation. To measure the quality of RNGs, different statistical tests have been developed. The more tests a proposed can pass, the higher the confidence that the algorithm is appropriate to be used in Monte Carlo simulations, but absolute certainty that the RNG won’t fail cannot be given. (L’Ecuyer, 2001). These statistical tests could be grouped into the following categories (Klimasauskas, 2002):

A.2.2. Simple statistical tests (Knuth, 1997)

Simple statistical tests verify whether the generated sequence of pseudo-random numbers is uniformly distributed in the unit interval \([0,1]\). Failing these tests simply invalidates the proposed algorithm as a RNG. Standard tests to analyse the distribution of a RNG against a theoretical (i.e. uniform) distribution include the Kolmogorov-Smirnov test and \(\chi^2\)-test. Another important test is the serial correlation test where the uniformity of \(n\)-tuples, \((x_{ni}, x_{ni+1}, \ldots, x_{ni+n-1})\) is tested using a \(\chi^2\)-test.

A.2.3. Advanced statistical tests

These tests verify more advanced features of the statistical properties of the generated pseudo-random numbers of a particular RNG. An example of an advanced statistical test is the entropy test. This test analyses the compression ratio that can be obtained from a generated sequence of random numbers using for instance, the Lempel-Ziff-Welch data compression algorithm. If the generated sequence of pseudo-random numbers contains a recurring pattern, then a higher compression ratio will be achieved. Ideally, the pseudo-random number sequence is uncompressible.

A.2.4. Basic simulation-bases tests

These tests simulate a particular system, where the outcome is known, and the deviation from what is expected, is measured. Numerous tests have been developed, especially by authors such as Knuth (1997), Marsaglia (1985, 1993), Hellekalek (1998) and L’Ecuyer (2001), including:
• **Birthday spacing test:** It can be proven that for a group of \( n \) persons, the distribution of the number of days between two successive birthdays follows a Poisson distribution. To test the RNG, the random numbers are used as birthdays of \( n \) people and the distribution of the gap (or birthday spacing) size can be tested against the Poisson distribution using the \( \chi^2 \)-test.

• **Poker test:** The dealing of cards from a deck is simulated using the pseudo-random numbers to pick the cards. The distribution of different poker hands, such as three-of-a-kind, full house, etc. is tested against the theoretical distribution using the \( \chi^2 \)-test.

• **Run test:** This test counts the length and distribution of monotone (continuous ascending or continuous descending) sections in a sequence of pseudo-random numbers, which should follow a specific distribution.

• **Monte Carlo integration of the number Pi:** Remarkably, uniformly generated random numbers could be used to calculate the value of the number Pi. Suppose couples of random numbers \((x_i, x_{i+1})\) with \( x_i \in [0,1] \), are used as coordinates of points as shown in Figure A.1. Then all points with \( x_i^2 + x_{i+1}^2 \leq 1 \) lie inside first quadrant of the unit circle, which has a surface of \( \pi/4 \). The total surface of the square in which all points are generated equals 1. By counting all points within the quadrant of the unit circle and dividing by the total number of generated points, the value of \( \pi/4 \) is approached if a sufficiently large number of points is generated and, if the values of \( x_i \) are truly uniformly distributed. An example of calculating Pi using the Mersenne Twister is given in Figure A.2.

![Figure A.1.](image)

**Figure A.1.** Calculating the value of Pi is based on counting the number of random points that are within the unit circle compared to the total number of points.
Figure A.2. The output in the test to calculate the number Pi using the Mersenne Twister as source of random numbers.

These are only a few examples of tests which fall in this category. Marsaglia (1995) standardised and bundled 12 different assays into a testing, which became known under the name ‘Diehard’. Ever since, the Diehard testing suit has become the de-facto standard for testing and certifying new RNG algorithms (Narasimhan, 2001; Tan, 2002; Srinivasan et al., 2003).

A.2.5. Application-specific tests

Complete Monte-Carlo models, whose exact results are known analytically, could also be used as a means of testing RNG algorithms (Srinivasan et al., 2003). Examples include the Ising-model, a simple lattice spin model and the random walk model (Kang, 2005).
Appendix B. Simulation procedures and implementation

B.1. Introduction

B.1.1. Programming environment

The entire model was programmed in Visual Basic 6, developed by Microsoft (US). This particular programming environment was chosen because it integrates a language which is widely used and considered to be easy-to-learn compared with traditional programming languages such as C or C++. Standard built-in features such as graphic user interface components provide VB6 an environment suitable for rapid application development. However, one of the serious drawbacks of Visual Basic is the overall low performance of the compiled code. This problem is especially visible when running simulations which take up more than 24 hours to complete on non-optimised code. In order to alleviate this problem, part of the code, more specifically, the numerically intensive code blocks such as the functions to recalculate thermodynamic particle properties, were ported to C++ code and compiled into a dynamic link library (DLL) using Visual C++ 6 (Microsoft, US). These DLL-embedded functions and procedures could then be called by the main simulation VB6-written program, allowing code execution at much higher speeds.

B.1.2. Structure of the simulation program

The simulation code, which is described in the following section, was implemented as a part of a software package which had a complete custom designed GUI (graphic user interface). Multiple models, with partial sharing of the simulation code base, could be implemented. Besides the necessary simulation procedures, the software package contained procedures and modules which provide the following functionality:

- Model selection, variable and parameter input modules
- Command line interface with scripting ability
- Material property database
- Saving or restoring a particular model configuration
- Automated simulation crash recovery
- Simulation debugging mode
- Simulation results database
- Simulation data post processing tools and advanced spreadsheet macros (i.e. for automated processing of data in sensitivity analysis)
Focusing on the actual simulation procedure, Figure B.1 gives an overview of the different steps involved from the moment the user activates the simulation by clicking the 'start'-button until the simulation ends successfully or is terminated unexpectedly, i.e. in case of oscillations or fatal errors due to inappropriate selection of the model and simulation variables.

![Figure B.1. Overview of the main simulation procedure](image)

The following groups of procedures are executed in a single simulation run:

- **Prepare simulation environment**: This group contains five different preparatory procedures. First, a procedure checks whether the entered values for the model parameters and variables fit into an acceptable range. Second, possible presence of overflow errors in the simulation of the particle exchange is analysed. This check includes verifying the continuity equation (3.44) for each control volume $S_i$. Also, the total number of particles that could be exchanged with the neighbouring control volumes $S_{i-1}$ and $S_{i+1}$ cannot be larger than the number of particles contained within $S_i$, or the control volume would be exhausted (or overloaded) during the execution of the exchange algorithm. The number of exchanged particles from $S_i$ toward $S_{i+1}$ and $S_{i-1}$ can be calculated using Eqs. (3.42) and (3.43) and has to lie between the following boundaries to avoid overflow errors:

$$1 \leq N_{i \rightarrow i+1,i-1} = \text{int} \left( \Delta t_{\text{sim}} N_{\text{sim}} \left( r_i + r_{i-1} \right) \right) \leq \frac{N_{\text{sim}}}{n}$$  \hspace{1cm} (B.1)
From Eq. (B.1) it is obvious that overflow errors could be avoided by appropriate selection of the number of simulated particles, \( N_{\text{sim}} \), and the simulation time step, \( \Delta t_{\text{sim}} \).

The third procedure allocates the necessary memory space for all control-volume and particle-related variables and the fourth procedure initialised the files for data output and checks for file-related errors (file already existing, file already in use by other programs, ...). The final procedure initialises the random number generator (Mersenne Twister RNG) by providing an appropriate seed value to the RNG algorithm (see also Section B.2.1).

- **Initialise simulation:** In these procedures, the simulated population is generated and the initial particle-related properties \( (d_p, T_p, W_p, Y_p) \) are attributed. Also, the thermodynamic properties of the wall and the gas-phase in each control volume are initialised. Finally, some bed-related properties such as bed voidage, \( \varepsilon_{\text{bed}} \), bubble voidage, \( \varepsilon_{\text{bu}} \), bubble frequency, \( f_{\text{bu}} \) and bubble diameter, \( d_{\text{bu}} \) are calculated, which are required for the heat loss calculations.

- **Main simulation loop:** Contains all model calculations, full details on these model-specific simulation procedures are given in Sections B.2 and B.3.

- **Data collection and saving:** As the dynamic processes are being simulated, huge amounts of data are generated and saved to disk. The following data is collected during simulation: dynamic air temperatures \( (T_{a,i}) \), air humidities \( (\varphi_{a,i} \text{ and } X_{a,i}) \), all dynamic air-particle heat and mass transfer components per control volume and heat loss per control volume, including all heat loss components as shown in Figure 3.6.

- **Termination procedures:** When an error occurs during the preparatory stage or during the actual simulation, the code execution path is rerouted to the termination procedure. This procedure includes closing all file access and reporting the error to the user. If the simulation is successful, then a new entry is added to the internal simulation database, which helps in keeping track of all performed simulations and the variables combinations used in each simulation.

**B.2. One-dimensional, basic (Chapter 3) fluidised bed coating model**

**B.2.1. Detailed description of the simulation routine**

The core of the simulation program is the main simulation loop, which is executed for each time step \( \Delta t_{\text{sim}} \); a graphical representation is given in Figure B.2. One of the first procedures to be executed within this loop, is the *input variable controller*, which allows the study of how the system (being the modelled fluidised bed coating process) responds when input variables or parameters are modified during the process (such as an increase in spraying rate, or varying
temperature/humidity in the inlet air). Table B.1 gives an overview of the different input types that can be generated by the input variable controller.

The next step is the procedure responsible for the particle exchange between the different control volumes. The principles of particle exchange in a finite population have already been discussed in Section 3.4.3.2. The basic idea is that particle are tagged at random and reallocated to neighbouring control volumes according to the particle exchange rate, \( r_i \). The practical implementation of this algorithm is given in Figure B.3.

**Table B.1.** Different input types handled by the input variable controller

<table>
<thead>
<tr>
<th>Type</th>
<th>Graph</th>
<th>Equation</th>
</tr>
</thead>
</table>
| 1. Step input | ![Graph](image1.png) | \[
x_i(t) = \begin{cases} 
0 & t < t_0 \\
= & a & t \geq t_0
\end{cases}
\]
| 2. Ramp input | ![Graph](image2.png) | \[
x_i(t) = \begin{cases} 
0 & t < t_0 \\
= & a(t - t_0)/(t_1 - t_0) & t_0 \leq t \leq t_1 \\
= & a & t > t_1
\end{cases}
\]
| 3. Frequency input | ![Graph](image3.png) | \[
x_i(t) = \begin{cases} 
0 & t < t_0 \\
= & a \sin(\omega t + \theta) & t_0 \leq t \leq t_1 \\
= & 0 & t > t_1
\end{cases}
\]
| 4. Linear combination of (1), (2) or (3) | | \[
x(t) = \sum_{i=1}^{k} b_i x_i(t)
\]
Figure B.2. Overview of the main simulation routine

The principle at the basis of this algorithm in Figure B.3 is that first, $n$ vectors $P_i$, composed out of the particle indices (each simulated particle has a unique index) contained within the control volume $S_i$, are constructed. The number of particles that have to be selected for
exchange was calculated using Eqs. (3.42) and (3.43). Particles are randomly sampled from the vector \( P_i \) using a RNG which generated samples in the unit interval \([0, 1]\). Once a particle has been selected for transfer, its index is removed from \( P_i \). The resulting gap in \( P_i \) is then removed by concatenating the two formed parts of \( P_i \) resulting in a new vector \( P_i \) reduced by one element and the next particle can be sampled.

One of the issues with the exchange algorithm is that its execution is significantly slowed down by the vector concatenations. Therefore, a modification to the original exchange algorithm was proposed, as shown in Figure B.4. In this modified algorithm, the vector \( P_i \) is not updated between two subsequent selection steps. As a result, collisions occur, meaning that the same particle could be selected more than once. Therefore, when a collision is detected, the algorithm tries a new selection until a particle has been found that has not yet been selected before. This modified algorithm is faster, especially when exchanging a small number of particles, compared to the number of available particles in \( S_i \). When a larger fraction of particles from \( S_i \) has to be selected, the algorithm slows down significantly due to the ever increasing number of collisions in the selection procedure. Consequently, for high values of \( r_i \) – or high values of \( \Delta t_{sim} \) – the non-modified exchange algorithm is preferred.

As already demonstrated, the Mersenne Twister RNG was preferred over Visual Basic’s internal RNG due to its superior quality and very long period (cfr. Section 3.4.3.3 and Appendix B). The source code of the Mersenne Twister has been made freely available on the internet under the BSD license (Matsumoto and Nishimura, 2004). The source code was compiled into a dynamic link library (DLL) using Visual C++ 6.0 (Microsoft, US), which enabled the use of the C-compiled functions within applications written in other programming languages such as Visual Basic 6.0 (Microsoft, US). The Mersenne Twister is composed out of two parts:

- First, the generator is seeded (or initialised) using the function `init_genrand(seed)` in which the seed is a 32-bit unsigned integer value. Usually the computer system time, using the `timer` function, or the CPU cycle counter, using the `queryperformancecounter` function (in MS Windows’ `kernel32.dll` library) are used to provide the seed value. The CPU cycle counter is 64-bit register that counts each clock cycle – on a 1 GHz processor, there are \(10^9\) clock cycles per second – from the moment the CPU was powered on. Consequently the CPU cycle counter can be considered a valid source of entropy or randomness to give the initial value or seed in the pseudo-random number generation.

- Next, pseudo-random numbers, within the unit interval \([0, 1]\) are generated using the function `genrand_real2()`. Note that the Mersenne Twister also includes functions to generate pseudo-random numbers between \([0, 1]\) or \([0, 1]\).
Figure B.3. The particle exchange procedure in the computer simulation program

Figure B.4. The particle exchange procedure, modified for execution speed for low values of $N_{\text{sim}}r_i\Delta t_{\text{sim}}$

Modelling heat and mass transfer in fluidised bed coating processes
The third step in the simulation consists of all calculations on the particle-level including the particle heat balances, using Eqs. (3.32) and (3.33); the particle moisture balances, based on Eqs. (3.36) and (3.37) and the particle coating mass balances, using Eqs. (3.34) and (3.35). When calculating the drying rate $r_D$, two limiting factors are needed to be taken into consideration (see Figure B.5): the available moisture content on the particles and, in case of condensation (negative $r_D$), the available moisture content in the gas (air) phase, or:

$$\min (r_D) = \frac{nM_{a,i}X_{a,i}}{N_{\text{bed}}\Delta t_{\text{sim}}}$$

$$\max (r_D) = W_p$$

(B.2)

For the part of the heat losses that have to be substracted from the particles (see also Eq. (3.32)), the calculated heat loss from the previous iteration is used (from $t - \Delta t_{\text{sim}}$).

**Figure B.5.** Drying rate calculation procedure of a single particle

The fourth step in the simulation procedure performs all thermodynamic calculations on the control volume’s air and wall level including the heat loss calculation, wall element heat balance, fluidisation air heat balance, using Eqs. (3.45) and (3.46) and air mass balance using Eqs. (3.47) and (3.48). Also a procedure has been included to check whether the dynamic simulation exhibits oscillatory behaviour, for instance due to the selection of an inappropriate time step $\Delta t_{\text{sim}}$. Finally, the exhaust air thermodynamic properties are calculated. Note that the exhaust air is defined as the air that is leaving the fluidised bed at the bed/headsapce interface. In case of a one-dimensional model, the thermodynamic properties of the exhaust air are the same of those of the air in the highest positioned control volume, $S_n$.
B.3. One-dimensional, droplet phase extended (Chapter 4) fluidised bed coating model

B.3.1. Merging the droplet model with the discretised, combined population balance and thermodynamic model

B.3.1.1. Introduction

As already stated in the Section 4.3.3, the droplet submodel has been designed to work in tandem with the main model whereby the axial temperature and humidity profiles (at any given time \( t \)) of both the gas and solid phases are given as input to the droplet submodel. The droplet submodel, in turn, provides the droplet-related interaction terms as input to the main model at time \( t + \Delta t_{\text{sim}} \) (cfr. Figure 4.8). Figure B.6 provides a more detailed description of the implementation of the droplet submodel and the subsequent processing of the droplet-related output into the main model simulation procedure as described in Chapter 3.

Instead of representing the solid phase by means of its control-volume averaged properties, such as the average particle temperature, \( T_{p,i} \), the average coating mass content, \( Y_{p,i} \), and the average moisture content, \( W_{p,i} \), the solid phase was modelled as a finite population of \( N_{\text{sim}} \) particles. Consequently, the solid phase heat and mass balances, being Eqs. (4.7), (4.11) and (4.13), were replaced by the individual particle heat and mass balances. In Equations (4.7) and (4.11), the coating solution is distributed continuously, expressed by means of the droplet collection rate variable, \( r_{C} \). In the population balance model, using a finite population (or subset) of \( N_{\text{sim}} \) particles, coating solution deposition was no longer continuous. Instead, given the droplet collection efficiency, the number of droplets to disperse per control was calculated and randomly distributed among the particles in that control volume.

In the following Section, more detail is provided concerning how the droplet submodel-generated data is processed into the main model and how the main model’s algorithm for coating liquid distribution was modified.
Figure B.6. Lay-out of the main simulation algorithm and the implementation of the droplet submodel.

B.3.1.2. Intermediary processing of droplet submodel-generated data

The implementation of the droplet phase within the discretised model, described in Section 3.4, is fairly different from the description of the continuous droplet-phase extended model in Section 4.2. The majority of droplet-related interaction terms in the heat and mass balances as
described in 4.2, are entirely pre-computed within the droplet submodel. Each time the droplet submodel is executed, the trajectories and accompanying heat and mass transfers are repeatedly calculated for a pre-set number of droplets, \( N_{\text{dr,sim}} \).

The model-predicted data, which includes droplet trajectories and the droplet/gas heat and mass transfers of \( N_{\text{dr,sim}} \) droplets is further processed into the following outputs:

- By counting the successful droplet-particle adhesions occurring in each control volume of the \( N_{\text{dr,sim}} \) simulated droplets, the overall collection efficiency per control volume, \( \zeta_{\text{dp,i}} \), could be calculated.
- The droplet-air heat transfer term \( \alpha_{\text{dp,i}} A_{\text{dp,i}} \left( T_{\text{a,i}} - T_{\text{dp,i}} \right) \) and the droplet-air mass transfer term \( r_{\text{D,i}}^* M_{\text{dp,i}} \) to be used in the heat and mass balances in Eqs. (4.8), (4.9) and (4.14). These terms are calculated using the cumulative droplet/gas phase heat and mass exchanges of \( N_{\text{dr,sim}} \) droplets. However, due to computational limitations, the number of simulated droplets per second of simulation time, is actually much smaller than the number of droplets produced per second in a (real) physical system,

\[
N_{\text{dr,real}} = \frac{6 M_{\text{sol}}}{\pi \rho_{\text{sol}} d_{\text{dr,nox}}^3}
\]  

(B.3)

Therefore, the cumulative heat- and mass transfers of \( N_{\text{dr,sim}} \) droplets have to be multiplied with a scaling factor,

\[
\alpha_{\text{dp,i}} A_{\text{dp,i}} \left( T_{\text{a,i}} - T_{\text{dp,i}} \right) = \frac{N_{\text{dr,real}}}{N_{\text{dr,sim}}} \sum_{j=1}^{N_{\text{dr.sim}}} \alpha_{\text{dr,j}} A_{\text{dr,j}} \left( T_{\text{a,j}} - T_{\text{dr,j}} \right) \]  

(B.4)

\[
r_{\text{D,i}}^* M_{\text{dp,i}} = \frac{N_{\text{dr,real}}}{N_{\text{dr,sim}}} \sum_{j=1}^{N_{\text{dr.sim}}} r_{\text{D,j}}^* M_{\text{dr,j}} \]  

(B.5)

Note, that in Eqs (B.11) and (B.12), \( T_{\text{dr,j}} \) represents the temperature of the \( j^{\text{th}} \) individual droplet, while \( T_{\text{dp,i}} \) stands for the droplet phase temperature in the \( i^{\text{th}} \) control volume.

- Per control volume, the droplet temperature \( \left( T_{\text{dr,c,i}} \right) \), droplet dry matter content \( \left( DM_{\text{dr,c,i}} \right) \) and droplet diameter \( \left( d_{\text{dr,c,i}} \right) \) distributions at the moment of successful droplet-particle adhesion are calculated based on the collision data of \( N_{\text{dr,sim}} \) simulated droplets.
- The spray drying rate, \( \dot{M}_{\text{sd,i}} \), and the rate at which droplets adhere to the inner reactor wall, \( r_{\text{c,i}}^* A_{\text{w,i}} \).
B.3.1.3. Deposition of coating solution

For the simulation of the coating liquid dispersion (as droplets) onto the simulated particle population, the following procedure was used, which was executed for each time step $\Delta t_{\text{sim}}$.

The procedure is detailed in Figure B.7:

First, the total mass of dry matter (coating polymer) to be dispersed during a single time step $\Delta t_{\text{sim}}$ onto the simulated particle population is calculated based on Eq. (B.6):

$$M_{\text{dmd}} = \Delta t_{\text{sim}} \frac{N_{\text{sim}}}{N_{\text{bed}}} \left( M_{\text{sol}} D M_{\text{sol}} - \sum_{i=1}^{n} r_{C,i} A_{w,i} - \sum_{i=1}^{n} M_{\text{sol},i} \right)$$  \hspace{1cm} (B.6)

In Eq. (B.6) the term $N_{\text{sim}}/N_{\text{bed}}$ is the scaling factor between the simulated population ($N_{\text{sim}}$ particles) and the actual number of particles in the physical system being simulated ($N_{\text{bed}}$ particles). The loss terms in Eq. (B.6), being the spray drying loss and droplet wall adhesion, are supplied by the droplet submodel.

![Figure B.7. Detailed description of the working principle of the coating solution deposition algorithm (main model).](image)

Next, the discontinuous deposition of coating solution onto the particles is simulated. The amount of dry matter that has to be dispersed onto the particles per control volume during each time step $\Delta t_{\text{sim}}$ equals,
\[ M_{\text{dmd},i} = M_{\text{dmd}} \frac{\zeta_{\text{dr},i}}{\sum_{i=1}^{n} \zeta_{\text{dr},i}} \]  

(B.7)

The droplet collection efficiency per control volume, \( \zeta_{\text{dp},i} \) in Eq. (B.7), is one of the outputs resulting from intermediary processing of the raw submodel data. Based on the calculated coating mass to disperse, \( M_{\text{dmd},i} \), and the distributions of the droplet dry matter content and diameter at the moment of particle/droplet collision, the number of droplets to be dispersed among the particles in each control volume \( S_i \) could be calculated. Each drop impacting onto a particle in \( S_i \) is attributed a temperature, \( T_{\text{dec}} \), a diameter, \( d_{\text{dec}} \), and dry matter content, \( DM_{\text{dec}} \), according to the submodel-predicted droplet properties at the moment of droplet/particle adhesion, per control volume. Finally, the droplets’ dry matter and solvent (water) were then added to randomly selected particles from each control volume \( S_i \).

**B.3.2. The droplet submodel**

An overview of the simulation procedure of the droplet submodel is presented in Figure B.8.

---

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The simulation procedure consisted of the following parts:

- As stated in the model assumptions, the droplet submodel is two-dimensional axisymmetric. To simulate the (un)succesful collision between the travelling droplets and the fluidised particles, the particles somehow have to be introduced as discrete entities into the submodel. To that end, prior to the droplet trajectory simulation, particles were distributed throughout the two-dimensional plane in which droplet movement was simulated. The location of the particles reflects the time-averaged particle spatial distribution or time-averaged bed voidage distribution. Although the momentary bed voidage distribution is heterogeneous due to the presence of bubbles inside the bed, it was assumed that the time-averaged bed voidage distribution could be considered uniform.

Consequently, the particles were uniformly distributed between $h = 0$ and $h = h_{\text{bed}}$ in the reactor domain using the Mersenne twister RNG to provide the axial and radial particle coordinates. The total number of particles that had to be distributed along the reactor domain in order to simulate the solid phase with a preset voidage ($\varepsilon_{\text{bed}}$) was calculated as,

\[
N_{\text{planar}} = \frac{6(1-\varepsilon_{\text{bed}})h_{\text{bed}}}{\pi d_p^2} \left( d_b + \frac{(d_t-d_b)}{2(h_1-h_2)}h_{\text{bed}} \right)
\]

(B.8)

In Eq. (B.8), the two-dimensional plane onto which both particles and droplet were modelled, was considered a slice of the fluidised bed with a finite thickness of $d_p$ (Figure B.9). Approaching the modelled domain as a slice, rather than a (mathematical) 2D plane allowed to attribute it a volume and consequently, allowed to accurately calculate the number of particles contained within it ($N_{\text{planar}}$). Using typical process variables, as described in Table 3.1 and the dimensions of the Glatt GPCG-1 for the reactor domain, the number of particles to distribute over the modelled 2D plane typically ranged between 40000 to 60000.

Although each particle has the diameter $d_p$, the particle’s centre point does not necessarily lie in the modelled 2D plane, as shown in Figure B.9. Therefore, in the collision detection algorithm, the particle diameter intersecting the 2D plane, $d_{p,\text{planar}}$, had to be used instead of $d_p$; all other calculations, including the Reynolds and Stokes number are based on $d_p$.

Assuming that the particles are uniformly distributed, the intersecting diameter of the $N_{\text{planar}}$ particles was calculated using Eq. (B.9), where $U(0,1)$ represents uniformly random numbers in the unit interval $[0,1]$. 

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$$d_{p,\text{planar}} = d_p \sqrt{1 - (U(0,1))^2}$$

(B.9)

**Figure B.9.** Detail of the reconstruction of the spatial particle distribution inside the bed

The bulk of the simulation procedure, being the complete calculation of a single droplet trajectory, was repeated for $N_{\text{dr,sim}}$ droplets. This part was split up into the droplet initialisation procedure and the so-called ‘droplet main loop’. Initialising included attributing the initial droplet temperature, $T_{\text{dr,nox}}$ (which was equal to $T_{\text{sol}}$), the initial droplet dry matter content, $DM_{\text{dr,nox}}$ (equal to $DM_{\text{sol}}$) and the initial droplet size, $d_{\text{dr,nox}}$, calculated using the binary nozzle droplet size models of Lefebvre (1989). The ‘droplet main loop’ consisted of the actual droplet trajectory and droplet/gas phase heat and mass transfer calculations. The following steps were repeatedly executed within this ‘droplet main loop’:

- Calculation of the droplet displacement within the time interval $\Delta t_{\text{dr,sim}}$ using the force balance as described in Eq. (4.20). The equation for radial and axial droplet velocity and position are given in Eqs. (B.10) – (B.13). Note that $\Delta t_{\text{dr,sim}}$ is the time step in the simulation of the droplet path while $\Delta t_{\text{sim}}$ is the time step used in the main model.

$$v_{\text{dr,ax}}(t + \Delta t_{\text{dr,sim}}) = v_{\text{dr,ax}}(t) + (\ddot{a}_{\text{dr}} \cdot \hat{e}_{\text{ax}}) \Delta t_{\text{dr,sim}}$$

(B.10)

$$v_{\text{dr,rd}}(t + \Delta t_{\text{dr,sim}}) = v_{\text{dr,rd}}(t) + (\ddot{a}_{\text{dr}} \cdot \hat{e}_{\text{rd}}) \Delta t_{\text{dr,sim}}$$

(B.11)

$$h'_{\text{dr}}(t + \Delta t_{\text{dr,sim}}) = h'_{\text{dr}}(t) + v_{\text{dr,ax}} \Delta t_{\text{dr,sim}}$$

(B.12)

$$x_{\text{dr}}(t + \Delta t_{\text{dr,sim}}) = x_{\text{dr}}(t) + v_{\text{dr,rd}} \Delta t_{\text{dr,sim}}$$

(B.13)

- Next, a procedure checked whether the droplet had crossed the boundaries of the modelled reactor geometry (wall impact) in its displacement between $t$ and $t + \Delta t_{\text{dr,sim}}$. If wall impact was detected, the droplet main loop was terminated and droplet properties, such as $d_{\text{dr}}$ and the control volume into which the droplet underwent wall impact, were registered to calculate $r^w_{i,j}$. 

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• Third, the droplet/gas phase heat and mass transfers during the time interval between \( t \) and \( t + \Delta t_{dr,sim} \) are calculated.

• Once the moisture loss due to droplet/gas phase mass transfer was known, the droplet dry matter content, \( DM_{dr} \), and droplet diameter, \( d_{dr} \), were recalculated using Eqs. (4.31) and (4.32).

• The fifth step in the ‘droplet main loop’ was the particle/droplet collision detection algorithm. This algorithm analyses whether particle/droplet collision occurred in the time interval between \( t \) and \( t + \Delta t_{dr,sim} \) without further calculating the droplet impingement efficiency nor adhesion probability (which is reserved for the next step). To reduce the numerical intensity of the collision detection algorithm – as there were normally \( N_{planar} \) particles to verify – the algorithm used a moving boundary box, as shown in Figure B.10. All particles having their centre point within the moving boundary box were selected as collision candidates. After selecting the collision candidates – and hereby significantly reducing the number of particles to consider in the collision algorithm – each candidate was analysed for possible intersection on the line between the droplet midpoints at time \( t \) and \( t + \Delta t_{dr,sim} \) with the edge of the candidate particle.

![Figure B.10. The moving boundary box in the droplet/particle collision detection algorithm.](image)

• If a droplet/particle collision had been detected in the previous step, the droplet collection efficiency, \( \zeta_{dr} \), had to be calculated according to the equations in Section 1.3.2.3 and Eqs. (4.36) and (4.37). Using a random generated number from the unit interval \([0,1]\), droplet adhesion was successful if \( U(0,1) < \zeta_{dr} \). Successful droplet adhesion was followed by termination of the ‘droplet main loop’ and registration of \( T_{drc} \), \( d_{drc} \) and \( DM_{drc} \) to construct the droplet collision property distributions \((T_{drc,i}; d_{drc,i})\).
and $DM_\text{trc,i}$ per control volume after completing the simulation of the $N_\text{dr.sim}$ droplet trajectories.

- The droplet trajectory simulation continued if no successful adhesion occurred. One final step in the ‘main droplet loop’ consisted of verifying whether the droplet dry matter content was still lower than 1. Otherwise, the droplet would have been spray-dried and consequently, the simulation of the droplet trajectory was terminated.

- The initial droplet velocity was high, generally above 300 m s$^{-1}$ as can be calculated using Eq. (4.19) and thus requiring very small time steps, usually between 0.1 and 1 µs. However, in the fluidised bed, droplets rapidly decelerate to velocities between 1 and 10 m s$^{-1}$ which no longer requires the use of the very small time step, $\Delta t_\text{dr.sim}$. To reduce the number of simulation steps, an adaptive time stepping algorithm was implemented.

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**Figure B.11.** Adaptive time stepping algorithm including oscillation detection procedure

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The adaptive time stepping algorithm used both the droplet displacement, $\Delta x_{dr}$, and temperature change, $\Delta T_{dr}$, as criteria to modify the time step, $\Delta t_{dr,sim}$. The reason to include the droplet temperature in the adaptive time stepping, was that, once the droplet had decelerated, increasing the simulation time step solely based on the displacement will likely yield a time step being too large for thermodynamic phenomena (i.e. heat transfer, evaporation) resulting in unstable oscillations for droplet temperature, dry matter content and diameter properties throughout the simulation.

An overview of the adaptive time stepping algorithm is given in Figure B.11. First, if the droplet temperature change $\Delta T_{dr}$ between $t - \Delta t_{dr,sim}$ and $t$ was outside the (user-defined) interval $[\min(\Delta T_{dr}), \max(\Delta T_{dr})]$, then $\Delta t_{dr,sim}$ was recalculated so that $\Delta T_{dr}$ was repositioned within this interval (the modified time step is denoted as $\Delta t'_{dr,sim}$). Next, the droplet displacement $\Delta x_{dr}$ between $t$ and $t + \Delta t_{dr,sim}$ was estimated based on the droplet velocities at time $t$. Again, if $\Delta x_{dr}$ lied outside a user-defined interval $[\min(\Delta x_{dr}), \max(\Delta x_{dr})]$, the time step (denoted as $\Delta t''_{dr,sim}$) was recalculated. The smallest of both time steps, $\Delta t'_{dr,sim}$ and $\Delta t''_{dr,sim}$, was then selected. A final check was then applied to verify whether the newly proposed $\Delta t_{dr,sim}$ stayed within a user-defined interval, which functioned as a final safeguard to avoid excessively large or excessively small time steps.

It is important to note that simulation tests, despite all the adaptive time stepping algorithms and their safeguards, have shown to produce uncontrollable oscillatory behaviour in some droplet trajectories (usually 0.5 % of $N_{dr,sim}$). Deactivating the adaptive time stepping algorithm avoids the problem of oscillation – thus confirming that the algorithm in Figure B.11 is not absolutely safe – but comes with a heavy calculation time penalty. Therefore the inclusion of an oscillation detection procedure proved to be of vital importance when using adaptive time stepping. If excessive oscillation is detected, the ‘droplet main loop’ is preventively terminated, reinitialised and restarted, usually solving the problem of oscillatory behaviour.
Appendix C. Simplified 2D Langrangian model for the prediction of particle exchange rates

C.1. Model description

C.1.1. Motion equation

In particle trajectory models, also known as Langrangian models, the motion of each out of a population of particles is described by an ordinary differential equation. The particle velocity and the corresponding particle trajectory are calculated by integrating this motion equation of each particle (Fan and Zhu, 1998).

The particle movement was described by the Newton equation which includes all interacting forces exerted on the particle along its trajectory. These forces could be categorised among three groups: fluid-particle interaction forces, inter-particle interaction forces and external field forces.

The particle-fluid interaction forces include drag, buoyancy, lift forces, Basset force, Saffman force, virtual mass force and the Magnus effect (Finlay, 2001). However, in case the particle density is much larger than the fluid density ($\rho_p >> \rho_a$), only the drag force is significantly larger than all other fluid-particle interaction forces, resulting in a simplified analysis of the particle-fluid force balance (Crowe et al. 1997).

The group of inter-particle interaction forces consists of the short-ranged van der Waals, electrostatic and collision (or contact) forces of which only the contact force had to be taken into account. Finally, external long-range external fields, including gravitational, electric and magnetic fields contribute to the particle motion (Fan and Zhu, 1998).

By identifying the relevant and present forces in each of these three different categories, the following motion (Newton) equation was obtained (Limtrakul et al., 2003; Tatemoto et al., 2004):

$$M_p \frac{d\vec{v}_p}{dt} = \vec{F}_d + \vec{F}_{ct} - M_p \vec{g}$$  \hspace{1cm} (C.1)

C.1.2. Collision and contact forces

In Equation (C.1) is $\vec{v}_p$ the velocity vector of the particle, $\vec{F}_{ct}$ the contact force, which consists of a tangential ($\vec{F}_T$) and a normal component ($\vec{F}_N$), as shown in Figure C.1. Because the
particles were represented as points, only the normal contact forces were taken into account, which were modelled as a linear spring, without dissipation, connecting the two colliding particles.

\[ \text{Figure C.1.} \quad \text{The normal} \ (F_N) \text{ and tangential} \ (F_T) \text{ contact forces during particle collision (a)} \]

\[ \text{and the method of applying the (normal) contact force into the model.} \]

The spring coefficient \( k \) for the quantification of the normal contact force was calculated using the equation (Tatemoto et al., 2004):

\[ k = \frac{4}{3} E_{\text{eq}} \sqrt{\frac{d_{p,\text{eq}}}{2}} \quad \text{(C.2)} \]

Where \( d_{p,\text{eq}} \) is the equivalent particle diameter and \( E_{\text{eq}} \) is the equivalent elasticity modulus for a collision between two particles \( i \) and \( j \). The equivalent particle diameter was calculated as:

\[ d_{p,\text{eq}} = \left( \frac{1}{d_{p,i}} + \frac{1}{d_{p,j}} \right)^{-1} \quad \text{(C.3)} \]

For monodisperse particle populations, Eq. (C.3) is further simplified to

\[ d_{p,i} = d_{p,j} = \frac{d_{p,\text{eq}}}{2} \quad \text{(C.4)} \]

In Eq. (C.2), the equivalent elasticity modulus was given by:

\[ E_{\text{eq}} = \left( \frac{1 - v_i^2}{E_i} + \frac{1 - v_j^2}{E_j} \right)^{-1} \quad \text{(C.5)} \]
For a collision of a sphere with a wall, the same relation for $E_{\text{eq}}$ applies, but the equivalent diameter is now given by $d_{p,\text{eq}} = d_p$.

### C.1.3. Drag forces

To solve the motion (or Langrangian) equation of the particle, the dynamic behaviour of the gas flow needs to be predetermined. As a first and simplified approach, the motion of the fluid (gas) – in the absence of the gas jet produced by the pneumatic nozzle – was assumed to be comparable to laminar ($Re_{a,\text{in}} \leq 2300$, or using the reactor dimensions listed in Table 5.2, $G_{a,\text{in}} \leq 0.011 \text{ kg s}^{-1}$) or turbulent ($Re_{a,\text{in}} > 2300$, or $G_{a,\text{in}} > 0.011 \text{ kg s}^{-1}$) pipe flow inside the reactor, the gas being incompressible and inviscid. In reality, the motion of the fluid and the particles are coupled; however, introducing this coupling or mutual interaction into the model would inevitably result in increased model complexity necessitating the use of iterative solving schemes.

The drag force $F_d$ exerted by the fluid upon the particles depends on the void fraction (and is given by (Limtrakul et al., 2003; Tatemoto et al., 2004):

$$F_d = \frac{\beta' d_p^3}{6\pi(1-\epsilon)} \left( \bar{v}_p - \bar{v}_a \right) \quad \text{(C.6)}$$

with

$$\beta' = \frac{\mu_a (1-\epsilon) \left[ 150(1-\epsilon) + 1.75 Re_p \right]}{\epsilon d_p^2} \quad (\epsilon \leq 0.8) \quad \text{(C.7)}$$

$$\beta' = \frac{3}{4} C_D \frac{\mu_a (1-\epsilon) \epsilon^{-2.7}}{d_p^2} Re_p \quad (\epsilon > 0.8) \quad \text{(C.8)}$$

In Eqs. (C.7) and (C.8) is $\epsilon$ the local bed voidage fraction where the particle is residing at time $t$, while $C_D$ is the drag force coefficient on each particle, given by

$$C_D = \frac{24}{Re_p} \left( 1 + 0.15 Re_p^{0.687} \right) \quad (Re_p \leq 1000) \quad \text{(C.9)}$$

$$C_D = 0.43 \quad (Re_p > 1000) \quad \text{(C.10)}$$

Where the dimensionless Reynolds number of the particle, $Re_p$, is equal to:

$$Re_p = \frac{\rho_a \epsilon d_p |\bar{v}_p - \bar{v}_a|}{\mu_a} \quad \text{(C.11)}$$
C.2. Calculation procedure

Prior to the simulation of the particle trajectories, the number of particles that have to be modelled is calculated using the following equation:

\[
N'_{\text{planar}} = \frac{6(1-\varepsilon_{\text{bed}})h_{\text{bed}}}{\pi d^2_p} \left( d_b + \frac{(d_i - d_b)}{2(h_i - h_b)}h_{\text{bed}} \right)
\]  
(C.12)

The calculation of the particle trajectories is straightforward: For each particle, a motion equation – Eq (C.1) – which is an ordinary differential equation (ODE) has to be solved. During each time step \(\Delta t'_{\text{sim}}\), this ODE is solved for each particle in the model:

\[
M_p \ddot{\mathbf{r}}_p = \mathbf{F}_d + \mathbf{F}_{\text{ct}} - M_p \ddot{\mathbf{g}}
\]  
(C.13)

\[
v_{p,ax}(t' + \Delta t'_{\text{sim}}) = v_{p,ax}(t') + (\ddot{\mathbf{a}}_p \cdot \mathbf{e}_{ax}) \Delta t'_{\text{sim}}
\]  
(C.14)

\[
v_{p,rd}(t' + \Delta t'_{\text{sim}}) = v_{p,rd}(t') + (\ddot{\mathbf{a}}_p \cdot \mathbf{e}_{rd}) \Delta t'_{\text{sim}}
\]  
(C.15)

\[
h_p(t' + \Delta t'_{\text{sim}}) = h_p(t') + v_{p,rd} \Delta t'_{\text{sim}}
\]  
(C.16)

\[
x_p(t' + \Delta t'_{\text{sim}}) = x_p(t') + v_{p,rd} \Delta t'_{\text{sim}}
\]  
(C.17)

Finally, the particle exchange rates, \(n_k(i, j)\) in Hz, were calculated by counting the number of particle crossings per unit time through the boundary surfaces of each control volume \(S(i, j)\) and by dividing by the number of simulated particles, \(N'_{\text{sim}}\).
Appendix D. Estimation of the gas flow rates in the fluidised bed

D.1. Description

As stated by various authors, e.g. Donadono et al. (1980), Becher and Schlünder (1997), Zank et al. (2001) and Ariyapadi et al. (2003, 2005) the gas jet formed by the release of compressed air by the pneumatic nozzle, submerged in a fluidised bed, shows many similarities to a free axisymmetric jet.

The gas velocity profiles of a free axisymmetric jet have been mathematically derived from the mass and momentum conservation equations by Schlichting et al. (2004) and are given in Eqs. (D.1) and (D.2). It is important to note that this approach assumes a gas jet suspended in an infinitely large fluidum. In reality, the effect of the reactor wall and the gas distributor at the bottom of the reactor on the gas flow pattern inside cannot be neglected.

\[
\frac{v_{a,ax}(h',\xi)-v_{a,f}}{v_{a,ax}(h',0)-v_{a,f}} = \frac{1}{\left(1+0.25\xi^2\right)^\frac{3}{2}} \text{ for } 0 \leq h' \leq h_{noz} \tag{D.1}
\]

\[
v_{a,rd}(h',\xi) = \frac{1}{2} \frac{\partial v_{a,ax}(h',0)}{\partial h'} (\xi - 0.25\xi^3) \left(1+0.25\xi^2\right)^\frac{3}{2} \text{ for } 0 \leq h' \leq h_{noz} \tag{D.2}
\]

In Eqs. (D.1) is \(v_{a,f}\) the fluidisation gas velocity and \(\xi\) is the dimensionless radial coordinate with

\[
\xi = 2 - \frac{x}{x_{at}(h')} \tag{D.3}
\]

With \(r\) the radial distance to the nozzle, \(h'\) the axial distance to the nozzle (\(h' = h_{noz} - h\)) and \(x_{at}(h')\) the jet radius at axial distance \(h'\) from the nozzle. According to Schlichting et al. (2004), the jet radius is defined as the radial distance from the jet’s symmetry axis where the radial gas velocity \(v_{at,rad}\) equals zero. In the case of axisymmetric jets, the following applies:

\[
x_{at}(h') \sim \frac{1}{h'^{\frac{1}{3}}} x_{at}(h') \tag{D.4}
\]

The proportionality between \(x_{at}(h')\) and \(h'^{\frac{1}{3}}\) is given by the so-called jet shape factor, \(s_{at}\) which was already discussed and experimentally derived in Section 4.6.3.1.
Assuming a constant air density in the fluidised bed, \( \rho_a \), the different mass flows in and out of a single control volume, whose shape was detailed in Figure 5.2, are calculated as

\[
G_a(i, j) = 2\pi \rho_a \int_{d_s(i+1, j)/2}^{d_s(i+1, j)/2} v_{a,x}(h', \xi) \, dx
\]

With \( h' = h_{noc} - h_x (i+1, j) \) and \( \xi = \frac{2x}{x_a(h')} \) \hspace{1cm} (D.5)

\[
G_a(i, j) = 2\pi \rho_a \left( \int_{d_s(i, j)/2}^{d_s(i+1, j)/2} v_{a,x}(h', \xi) \, dx + \int_{h_x(i, j)/2}^{h_x(i+1, j)/2} v_{a,yd}(h', \xi) \, dh \right)
\]

With \( h' = h_{noc} - (h_x(i+1, j) - h_x(i, j)) \frac{d_y(i+1, j) - h_x}{d_y(i+1, j) - d_y(i, j)} \) and \( \xi = \frac{2x}{x_a(h')} \) \hspace{1cm} (D.6)

The calculation of \( G_a,3 \) and \( G_a,4 \) is similar to \( G_a,1 \) and \( G_a,2 \), respectively. The only difference being that the index \( i+1 \) is replaced by \( i \) in Eq. (D.5) for \( G_a,3 \); and the index \( j \) by \( j-1 \) in Eq. (D.6) to retrieve \( G_a,4 \).
Curriculum Vitae

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Education

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1996-2001: Bio-engineer, option Chemistry, with great honour,
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1990-1996: General secondary education
Math (8h) – Sciences
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Professional activities

09/2005 – …
Function: Academic assistant
Faculty of Bioscience Engineering, Ghent University
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09/2001 – 08/2005
Function: BOF – mandated research assistant
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Workshops


Scientific publications (as accepted or published on 06/06/06)

A1 (Articles in journals from ISI - Web of Science):


A2 (Articles in international journals with peer review, non-ISI):


A3 (Articles in national journals with peer review):


Active participation to symposia, congresses or workshops with oral presentation


**Active participation to symposia, congresses or workshops with poster presentation**


**Contributions to oral presentations or posters**


**Scientific awards**

Best paper award, 3rd International Conference on Simulation in the Food Industry (FOODSIM), 16-18 June 2004, Wageningen, The Netherlands