Bridges in modelling and simulation of steam cracking: from fossil to renewable feedstock and from molecule to furnace

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66th Canadian Chemical Engineering Conference “Sustainability & Prosperity”
October 16-19 (2016) Québec City (Canada)
From molecule to industrial plant

Reaction engineering

Process level

Molecular level
Steam cracking: from fossil to renewables

Crude oil
- Naphtha
- Light gasoil
- Crude oil

Natural gas
- Natural gas liquids
- Gas-condensates

Bio-based feeds
- Hydrode-oxygenated FAME

Steam cracking
- Ethene
- Propene
- Butadiene
- Aromatics

Consumer goods from chemical industry

atozforex.com; pnnl.org; districtenergy.org; scade.fr; schmidt-clemens.de; Linde Group
Steam cracking: Ghent University history

- 1974: Pilot plant starts operation
- 1976: First simulations of steam cracking
- 1978: First kinetic of coke formation
- 2001: Implementation of GCxGC
- 2004: Largely expanded kinetic of network
- 2008: TLE model
- 2010: Pilot plant revamped
- 2015: Kinetic expansion; Firebox and convection models
- 2016: COILSIM3D, continuous expansion of kinetic network...

COILSIM1D's GUI implementation
Steam cracking: hot section

Preheat feed and other utility streams

Feed
Dilution steam

5% 700K

FPH
ECO
SSH
HTC

50%

800K

1050–1150 K

1450 K

45%

Rapidly quenching of reactor effluent

A: Radiation section
B: TLE
C: Steam drum
D: Convection section

Endothermic process 1050–1150 K

COILSIM1D
The advanced simulation and optimization software for the ethylene industry
Reaction and reactor model

Analytical techniques

GC×GC

Feedstock properties

Simulation model

Reactant geometry
Operating conditions

Product specs

Modeling

Microkinetic model

Reactor model

Detailed reactor model

Detailed product composition

Reaction and reactor model


Toraman, H.E. et al., Journal of Chromatography A, 1460, 135-146, 2016
Outline

• Introduction
• Feedstock
• Kinetics
• Reactor
• Process
• Conclusions
Outline

• Introduction
• Feedstock: characterization
• Kinetics
• Reactor
• Process
• Conclusions
On-line GC×GC

GC×GC chromatogram: 2 parts
- Conventional 1D part → C₄-
- Comprehensive 2D part → C₅+

SIMCO: ANN or Shannon entropy

**Analytical techniques**
- Standard methods
- GC×GC

**Goal**
- Reactor geometry
- Operating conditions
- Feedstock properties
- Simulation model
- Product specs

**Feedstock reconstruction**
- SIMCO

**Microkinetic model**
- Detailed feedstock composition
- Fundamental reactor model

**Reactor model**
- Detailed product composition
Maximization of Shannon Entropy

Feedstock properties
- Average molecular weight
- Elemental composition
- Specific density
- Global PINA analysis
- Boiling point data (e.g. D2887 simdist)
- Aromatic Sulfur

Detailed composition
- Species identity
- Mole or mass fractions

\[
\text{MAX} \quad S(y_i) = - \sum_{i=1}^{N_M} y_i \ln(y_i) \quad \text{with} \quad \sum_{i=1}^{N_M} y_i = 1
\]

Constraints from mixing rules (example):

\[
\frac{1}{d_{\exp}} = \frac{\sum_{i=1}^{N_M} y_i M_{W_i}}{d_i \sum_{i=1}^{N_M} y_i M_{W_i}}
\]

More than 100 unknown mole fractions

S.P. Pyl et al., AIChE Journal, 56, 12, 3174-3188, 2010
SIMCO results: Hydrocarbons

- **n-paraffins**
  - Mass fraction (wt%) vs Carbon number

- **i-paraffins**
  - Mass fraction (wt%) vs Carbon number

- **Naphthenes**
  - Mass fraction (wt%) vs Carbon number

- **Aromatics**
  - Mass fraction (wt%) vs Carbon number
Outline

• Introduction
• Feedstock: renewables
• Kinetics
• Reactor
• Process
• Conclusions
Thermochemical conversion of biomass

GAS

CO

H₂O

C₂H₄

NH₃

H₂

CO₂

BIO-OIL

CH₄

H₂O

C₂H₄

HCN

CHAR

5wt% Torrefaction
35wt% Slow pyrolysis
13wt% Fast pyrolysis
85wt% Gasification

20wt%
30wt%
75wt%
5wt%

75wt%
35wt%
12wt%
10wt%

LIGNOCELLULOSIC BIOMASS

Model components for biomass pyrolysis

Study molecules with structural moieties found in biomass

For example:
Gamma-Valero Lactone (GVL) pyrolysis

Reaction families
1) Scission
2) Hydrogen abstraction
3) β-scission/addition
4) Concerted ring opening

De Bruycker, R. et al., Combustion and Flame, 164, 183-200, 2016
Outline

• Introduction
• Feedstock
• Kinetics
• Reactor
• Process
• Conclusions
CRACKSIM: steam cracking kinetics

**Goal**
- Reactor geometry
- Operating conditions

**Simulation model**

**Feedstock properties** → **Simulation model** → **Product specs**

**Modeling**
- Microkinetic model: CRACKSIM
- Reactor model

**Fundamental reactor model**

**Detailed feedstock composition** → **Fundamental reactor model** → **Detailed product composition**
Outline

• Introduction
• Feedstock
• Kinetics: reaction network
• Reactor
• Process
• Conclusions
Families of elementary reactions

Bond dissociation and radical recombination

\[ R_1 - R_2 \rightleftharpoons R_1^\cdot + R_2^\cdot \]

Hydrogen abstraction (inter- and intramolecular)

\[ R_1 - H + R_2^\cdot \rightleftharpoons R_1^\cdot + R_2 - H \]

Radical addition and β-scission (inter- and intramolecular)

\[ R_1^\cdot + R_2 - R_3 \rightleftharpoons R_1 - R_2 - R_3^\cdot \]
Decomposition scheme: n-hexane

R

R\H

\beta\ species
PSSA to $\mu$ radicals
μ and β networks: CRACKSIM

CRACKSIM

β network
Bi- and monomolecular reactions for β radicals
\[
\begin{align*}
R_1-R_2 & \overset{\text{••}}{\rightleftharpoons} R_1 + R_2 \\
R_1-H + R_2 & \overset{\text{••}}{\rightleftharpoons} R_1 + R_2-H \\
R_1 + R_2=R_3 & \overset{\text{••}}{\rightleftharpoons} R_1-R_2-R_3
\end{align*}
\]
1324 reversible reactions
51 molecules
43 radicals

μ network
Monomolecular reactions for μ radicals
\[
\begin{align*}
R_1-R_2 & \rightarrow R_1 + R_2 \\
R_2-R_1-H & \rightleftharpoons H-R_2-R_1 \\
R_1 + R_2=R_3 & \rightarrow R_1-R_2-R_3 \\
R_1-R_2=R_3 & \rightleftharpoons R_1\underset{R_2}{\longrightarrow}R_3
\end{align*}
\]
13584 schemes
676 molecules
Experimental yields during steam cracking of bio-derived hydrocarbons
Feedstock composition: $\text{MW}_{\text{average}} = 230\text{g/mol}$ 51wt% normal alkanes – 49wt% branched alkanes
$F_{\text{HC},0} = 0.04\text{ g/s}, F_{\text{H}_2\text{O},0} = 0.02\text{ g/s}, P = 0.17\text{ MPa}$

---
calculated
Network generators

RING Daoutidis Minneapolis
Genesys PRIM(-O) ReNGeP LCT, Ghent, Belgium
Rain Ugi Munich, Germany
REACTION Blurock Lund, Sweden
RING Lederer Prague, Czech Republic
CASB Porollo Josjkar-Ola, Russia
MAMOX(+) Mavrovouniotis Evanston
COMGEN Truong Salt Lake City
MECHGEN Valdes-Perez Pittsburgh
MECHEM Green Cambridge
EXGAS Battin-Leclerc Nancy, France
RMG Green Cambridge
NetGen Broadbelt Delaware
NetGen Ranzi Milan, Italy
KING Di Maio Cosenza, Italy
GRACE, Yoneda KUCRS, Miyoshi Tokyo, Japan
MAMOX(++) Ranzi Milan, Italy
RING Lederer Prague, Czech Republic
CASB Porollo Josjkar-Ola, Russia
MAMOX(++) Ranzi Milan, Italy
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CASB Porollo Josjkar-Ola, Russia
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CASB Porollo Josjkar-Ola, Russia
MAMOX(++) Ranzi Milan, Italy
A new program for kinetic model construction

GENESYS - GENERation [of reacting] SYStems

Genesys: Kinetic model construction using chemo-informatics
Vandewiele, N.M.; Van Geem, K.M.; Reyniers, M.-F.; Marin, G.B.
Chemical Engineering Journal, 207-208, 526-538, 2012

Graph theory yields powerful algorithms

Pattern Recognition

Reactant -> product

Molecule Identifiers

“Kinetics of Chemical Reactions : Decoding Complexity”
G.B. Marin and G.S. Yablonsky, Wiley-VCH Verlag, 446 pages, 2011
Outline

• Introduction
• Feedstock
• Kinetics: ab initio
• Reactor
• Process
• Conclusions
Objective: data base

Reactor simulation for hydrocarbon radical chemistry

- Reactor model
- Solver
- Reaction network
  - Kinetic and thermodynamic data

Develop a consistent data set based on ab initio calculations using group additivity

Van de Vijver, R. et al., Chemical Engineering Journal, 278, 385–393, 2015
Benson’s group additive method

Benson group

X: Central atom
Valence ≥ 2
{ C, C_d, O, CO, CCO, C•, C_d• }

A, B, C, D: Ligand

Notation: X-(A)(B)(C)(D)

Group additivity for thermochemistry

\[ f = \sum_i GAV_f \text{ (group}_i) + \sum_j NNI_j \]

\[ f = \Delta_f H^\circ, S^\circ_{\text{int}}, C_p^\circ \]

\[ S^\circ_{\text{int}} = S^\circ + R \ln\left( \frac{\sigma}{n_{\text{opt}}} \right) \]

Group additive values (GAV)

Corrections for non-nearest-neighbor interactions (NNI)

✓ hydrogen bonds
✓ gauche interactions
✓ other interactions
From small to large species with group additivity

2-methoxy-2-methylbut-3-enoic acid

Atoms in large molecules

Additivity

Atoms in small molecules with similar surroundings

**Group additivity**

**Additive groups**

- $C_d-(H)_2$
- $C_d-(C)(H)$
- $C-(C)(C_d)(O)(CO)$
- $O-(C)_2$
- $C-(O)(H)_3$
- $C-(C)(H)_3$
- $CO-(C)(O)$
- $O-(CO)(H)$
Outline

• Introduction
• Feedstock
• Kinetics: thermo first
• Reactor
• Processen
• Conclusions

Thermo e.g. for oxygenates: GAV data base

Database of thermodynamic data, $\Delta_f H^\circ$, $S^\circ$ and $C_p^\circ$ (300 K-1500 K)
- 450 oxygenate compounds
- CBS-QB3 methodology
- 1D-HR approximation for all internal rotors

- 157 GAVs using Benson’s GA method
- 26 NNI corrections (mainly hydrogen bonds)
- 77 HBIs for the thermochemistry of radicals

2-hydroxy-2-methyl-propanal

NNI8 Hbr_H-O-C-CO
Outline

• Introduction
• Feedstock
• Kinetics: rate coefficients
• Reactor
• Process
• Conclusions
Kinetics: computational approach

Conventional Transition State Theory (high pressure limit)

\[ A + B \overset{[AB]^+}{\rightleftharpoons} C \]

Transition state

\[ k_\infty(T) = \kappa(T) \frac{k_B T}{h} \frac{q^+}{q_A q_B} V_m e^{-\frac{\Delta E_0}{RT}} \]

Electronic barrier \( \Delta E_0 \)
The CBS-QB3 ab initio method is used.

Partition functions \( q \)
- Ideal gas approximation
- Hindered Rotor (1D-HR)

Tunneling coefficient \( \kappa \)
- Eckart

Reactants → Reaction coordinate → Products

\[ \Delta^\pm E_{\text{electronic}} \]
Group additivity for kinetics: data base of $\Delta G_{AV}^o$

Transition state for hydrogen abstraction

Arrhenius equation

$$k = A e^{-\frac{E_a}{RT}}$$

Number of single events

$$n_e = \frac{n_{opt, \delta}}{\prod_j \sigma_j}$$

Group additivity for Arrhenius parameters

**Primary**

$$E_a(T) = E_{a, ref}(T) + \sum_{i=1}^{2} \Delta G_{AV}^o_{E_a}(C_i) + \sum_{i=1}^{3} \Delta G_{AV}^o_{E_a}(X_i) + \sum_{i=1}^{3} \Delta G_{AV}^o_{E_a}(Y_i) + \Delta E_{E_a, res}$$

$$\log \tilde{A}(T) = \log \tilde{A}_{ref}(T) + \sum_{i=1}^{2} \Delta G_{AV}^o_{\log \tilde{A}}(C_i) + \sum_{i=1}^{3} \Delta G_{AV}^o_{\log \tilde{A}}(X_i) + \sum_{i=1}^{3} \Delta G_{AV}^o_{\log \tilde{A}}(Y_i) + \Delta \log \tilde{A}_{res}$$

**Secondary**

**Tertiary**

Proposed by Saeys et al. for activation energies (*AIChE J.*, 2004, 50 (2), 426-444.)

Extended by Sabbe et al. for pre-exponential factors (*Phys. Chem. Chem. Phys.*, 2010, 12, 1278-1298)
Kinetics: data base of $\Delta G^o_{AV}$

Hydrogen abstraction (ethyl + ethenyl methylether)

$$E_a(T) = E_{a, ref}(T) + \Delta G^o_{E_a}(C_1) + \Delta G^o_{E_a}(C_2) + \Delta G^o_{E_a}(X_1) + \Delta G^o_{E_a}(Y_1) + \Delta E_{a, res}$$

$E_{a, GA} = 62.6 \text{ kJmol}^{-1}$

$E_{a, \text{ ab initio}} = 62.0 \text{ kJmol}^{-1}$

$\log(A_{GA}/\text{m}^3 \text{ mol}^{-1} \text{ s}^{-1}) = 5.095$

$\log(A_{\text{ab initio}}) = 5.203$
Kinetics: validation of group additivity

Outline

• Introduction
• Feedstock
• Kinetics
• Reactor
• Process
• Conclusions
Outline

• Introduction
• Feedstock
• Kinetics
• **Reactor : pilot scale**
• Process
• Conclusions
Steam Cracking Pilot Plant

High temperature sampling system

Gas-Fired Furnace + Reactor

Online Analysis Section

Control “Room”

H₂O

HC Feed

HC Feed H

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Pilot results: $C_1/C_2/C_3/C_4$

<table>
<thead>
<tr>
<th>Process conditions</th>
<th></th>
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</thead>
<tbody>
<tr>
<td>Feed</td>
<td>3 wt% $C_1$</td>
</tr>
<tr>
<td></td>
<td>67 wt% $C_2$</td>
</tr>
<tr>
<td></td>
<td>22 wt% $C_3$</td>
</tr>
<tr>
<td></td>
<td>8 wt% $C_4$</td>
</tr>
<tr>
<td>COT</td>
<td>1005-1119 K</td>
</tr>
<tr>
<td>COP</td>
<td>0.152-0.157 MPa</td>
</tr>
<tr>
<td>Steam dilution</td>
<td>0 kg/kg</td>
</tr>
</tbody>
</table>

Without a single adjusted parameter

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66th Canadian Chemical Engineering Conference “Sustainability & Prosperity” October 16-19 (2016) Québec City (Canada)
• Introduction
• Feedstock
• Kinetics
• Reactor: 3D alternatives
• Process
• Conclusions
Deposition of a carbon layer on the reactor surface

- Thermal efficiency
- Product selectivity
- Decoking procedures

Estimated annual cost to industry: $ 2 billion

Mitigation by
- Feed additives
- Metallurgy & surface technology
- 3D reactor technology

Coke formation: 3D reactor technologies

Cokes formed here
T ↑  coking rate ↑↑

- ~ 15 °C
- ~600°C

Reduce convective heat resistance
Increase surface area
Better mixing

*Borealis.com, kubota.com, Technip.com*
“There ain’t no such thing as a free lunch”

3D reactor technology

Flow

Chemistry

3D

CRACKSIM

Increased heat transfer

Reduced coking rate

Longer run length

More capacity

Quantify coking & selectivity effect

Economics

Increased friction

Higher average reactor pressure

Selectivity ↓

* Milton Friedman - TANSTAAFL
Helicoidal finned tubes

Radiant coils in Borealis Furnace, KBR

Computational domain: 1 fin with periodic boundaries
Outline

- Introduction
- Feedstock
- Kinetics
- Reactor: CFD models
- Process
- Conclusions
Resolving turbulence

**Reynolds-Averaged Navier-Stokes (RANS)**
Single model for all scales, additional equations to provide closures

**Large Eddy Simulation (LES)**
Resolve relevant energy containing scales, model the smaller energy dissipating eddies

**Direct Numerical Solution (DNS)**
Fully resolve all time and length scales
Radial temperature profiles

Bare

More uniform gas temperature
Lower metal temperature

Bare

Optimized

Straight

Helix

SmallFins

1170
1203
1235
1268
1300
K
Axial wall temperature and coking profile

- 50°C
- 49%

Cracking reaction model
- 26 components
- 13 radical species
- 212 elementary reactions

Coke model

S. Wauters and G.B. Marin, IEC Research, 41, 2379-2391, 2002
S. Wauters and G.B. Marin; Chemical Engineering Journal, 82, 267-279, 2001
Effect on start of run yields

-0.7 wt% 
+0.3 wt%

Radical reaction model
26 components
13 radical species
212 elementary reactions
Outline

• Introduction
• Feedstock
• Kinetics
• Reactor: “run length”
• Process
• Conclusions
Run length simulation

Increasing run length

SOR 48h 96h

Axial position [m]

Heat flux [W m\(^{-2}\)]
Millisecound propane cracker

- Feedstock: 118.5 kg/h propane
- Propane conversion: 80.15 % (± 0.05%)
- Steam dilution: 0.326 kg/kg
- CIT: 903.7 °C
- COP: 170 kPa

Different geometries simulated
- Same reactor volume
- Same axial length
- Same minimal wall thickness
Non-uniform coke layer growth

SOR (0 hrs)

Fin

c-Rib

SOR

48 hrs

10 days
Tube Metal Temperature

Thermal resistance coke layer ➔ Max. TMT increases
Cross-sectional flow area decreases due to coke

Pressure drop increases
Outline

• Introduction
• Feedstock
• Kinetics
• Reactor
• Process
• Conclusions
Outline

• Introduction
• Feedstock
• Kinetics
• Reactor
• Process: fire box
• Conclusions
Coupled reactor-furnace simulation

External coil temperature

Reactor (COILSIM1D)

convergence

Furnace simulation

Heat flux to reactors

Hu, G. et al., Industrial & Engineering Chemistry Research, 54 (9), 2453-2465, 2015
Ultra Selective Conversion (USC)

- 100% floor burner
- Fuel composition mol%: CH$_4$(89%)-H$_2$(11%)
- U coil
- Feedstock: Naphtha

Coupled modeling

- 3D CFD furnace model
- 1D reactor model (COILSIM1D)
- Detailed cracking kinetics (CRACKSIM)

Zhang, Y. et al., AIChE Journal 61 (3), 936-954, 2015
Detailed : long flame burners

detailed

simplified
**Flue gas: velocity and concentration fields**

- Detailed case
  - Stronger turbulence
  - Faster reaction

Methane mole fraction

- Detailed
- Simplified
Tube wall temperature field: local hot spots

This information is key for control.
Outline

• Introduction
• Feedstock
• Kinetics
• Reactor
  • Process: convection section
• Conclusions
Steam cracker convection section

Schematic of convection section

Flue gas out ~ 700K

Feed
Evaporator
Steam super heater
Mixture overheater-1
Mixture overheater-2
Flue gas in ~ 1400 K

Mixing nozzle
To radiation section

Heavy feed

Feed

Nozzle

Flue gas in ~ 1400 K

Steam

Feed-steam mixture overheater-1

Evaporator

Feed

Mahulkar, A.V. et al., Chemical Engineering Science, 110, 31-43, 2014
Gas condensate: multicomponent mixture

- Wider stick regime as compared to that in single component droplet regime map
- For splash and limited splash, the no. of daughter droplets formed is greater than predicted by correlations available in literature

Mahulkar, A.V. et al., Chemical Engineering Science, 130, 275-289, 2015
Outline

- Introduction
- Feedstock
- Kinetics
- Reactor
  - Process: hot section
- Conclusions
The COILSIM1D package

COILSIM1D

Transfer Line Exchanger

Reactor Coil

Convection section

Wall Burners

Firebox

Long Flame Burners
The COILSIM1D package
Outline

- Introduction
- Feedstock
- Kinetics
- Reactor
- Process
- Conclusions
Conclusions

- Shannon Entropy maximization to reconstruct feedstocks in terms required for a microkinetic model
- Consistent data set for thermochemistry and kinetics of hydrocarbon and oxygenates radical chemistry
- Ab initio simulation of steam cracking of C$_2$/C$_3$/C$_4$ and oxygenates
- Compatibility of renewable feedstocks with existing plants
- Emergence of 3D reactor technologies based on CFD
- Integration of reactor/convection section/furnace integration of computational chemistry methods with engineering tools at larger time and length scales and experimental validation provides a powerful tool for the optimization and/or design of industrial units
Acknowledgments

The Long Term Structural Methusalem Funding

Long Term Structural Methusalem Funding of the Flemish Government
Acknowledgments

• Profs. Marie-Françoise Reyniers, Geraldine Heyndericks, Maarten Sabbe, Tony Arts, Feng Qian

• Drs. Steven Pyl, Amit Mahulkar, Nick Vandewiele, Ruben Debruycker, Carl Schietekat, Thomas Dijkmans, Paschalis Paraskevas, Marco Djokic, Andres Munoz

• PhD students David Van Cauwenberghe, Pieter Reyniers, Brigitte Devocht, Ruben Van de Vijver, Laurien Vandewalle, Nenad Ristic, Alper Ince, Ezgi Toraman, Yu Zhang, Marco Virgilio
• **SFT**: Swirl Flow Tube, a tube with a helicoidal centerline. The helix amplitude is smaller than or equal to the tube radius.
• **Swirl flow**: a whirling or eddying flow of fluid.
• **Swirl number**: ratio of tangential over axial momentum transfer
• **Wall shear stress**: component of stress parallel with the wall. It is the product of the viscosity and the derivative of axial velocity with respect to the radial coordinate.