“A CFD-based study of steady-state multiplicity in a gas-solid vortex reactor for oxidative coupling of methane”

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The ideal reactor for oxidative coupling of methane (OCM) has two key characteristics: 1) limited species backmixing to maximize the selectivity towards intermediate C_2 products, and 2) sufficient thermal backmixing to allow steady-state multiplicity and autothermal operation. Non-reactive computational fluid dynamic (CFD) simulations have already shown that the first of these characteristics can be obtained in a gas-solid vortex reactor (GSVR). Whether the GSVR also exhibits the second key characteristic, i.e., sufficient thermal backmixing, is less straightforward to verify. A reactive Euler-Euler CFD methodology is developed for this purpose, allowing to simulate the GSVR while using detailed microkinetic models for both the homogeneous gas phase and heterogeneous surface chemistry. Simulations of a 2D adiabatic GSVR for OCM are performed, for inlet temperatures ranging from 873 K to 1198 K, in 25 K increments, while fixing the inlet composition, mass flow rate, catalyst mass and total pressure. The possibility for steady-state multiplicity is assessed by numerically igniting some of the non-ignited steady states, and evaluating whether or not these cases reach a new steady state on the ignited branch. Thanks to steady-state multiplicity, a CH_4 conversion of 40 % (see Figure 1) and C_2 selectivity of about 75 % (incl. 53 % C_2H_2) can be obtained on the ignited branch, for an inlet temperature of merely 873 K.

Figure 1: Conversion of CH_4 and O_2 in a GSVR for OCM, clearly showing steady-state multiplicity. ($Sr-La/SiC$ catalyst, $P = 2$ bar, $m_{ca}/F_{V(NTP),CH_4}^0 = 3.6$ kg.s/Nm³, $CH_4:O_2=4$)