

Homogeneous modeling for laminar flows in structured catalysts: CO₂ methanation

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Over the recent years, research and industrial design and prototyping for reduction of CO₂ emissions has been focused on the production of Hydrogen (H₂) using surplus electricity generated by renewables. This is a way to store energy for later use within a CO₂-free power cycle. An alternative is to stop the release of new CO₂, while conserving its current levels, e.g., by direct conversion of CO₂ into CH₄. Moreover, the conversion of H₂ into CH₄ offers convenience in terms of transportation and storage. These are all possibilities offered by the catalytic hydrogenation of CO₂, which has seen moderate research in the last years [1]. Typically, catalytic reactions occur under conditions characterized by very low flow Reynolds numbers. At the macroscale, this manifests as a laminar flow, dominated by viscosity and linear advection. However, at the microchannel (pore) scale, the influence of inertial forces, involving nonlinear advection, can be significant [2]. In order to account for these effects, it is necessary to carry out Direct Numerical Simulations (DNS) of heterogeneous structured catalysts [3]. Unfortunately, these simulations are prohibitively expensive in real-world applications for the current accelerated design and prototyping demands. To this extent, the focus of this contribution is on the formulation of homogeneous flow models relying on the volume-averaging theory (VAT). By anticipating the dominant operational regime based on the dimensionless parameters characterizing the flow, it becomes feasible to establish a hierarchy of models, akin to the catalogue of empirical phenomenological relations routinely employed in process engineering, e.g., in the evaluation and design of heat exchangers [4]. It is imperative that the proposed models adhere to first principles rather than relying on observational statistics. To this end, expertise for the formulation of numerical reduced order turbulent flow models for hydrocarbon combustion [5], and for the design and prototyping of homogeneous structured catalysts [6], has been combined to formulate 0-D and 3-D homogeneous models for reacting porous flow in structured catalysts. In a first step, a 0-D model has been

formulated and validated in a steam reforming application. **Figure 1** shows the model results for a honeycomb-like structured catalyst. The figure shows the outlet chemical species concentrations (on dry basis) as a function of the catalyst inlet flow temperature. The model was applied in order to replicate the experimental results from [7], demonstrating reasonable agreement that underscores the dominant effect of chemical kinetics in the flow.

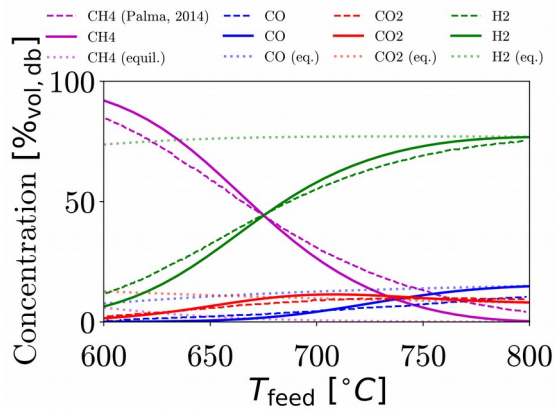


Figure 1: 0-D model results

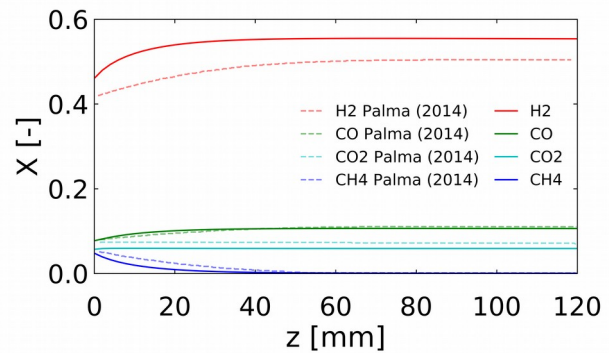


Figure 2: 3-D homogeneous model

Subsequently, a 3-D homogeneous model was developed and compared to the 3-D simulation results in [7]. The reference numerical simulations incorporate the detailed geometry of the honeycomb catalyst, while the proposed model simply considers a homogeneous fluid-solid phase, i.e., a one-equation model in the context of VAT. **Figure 2** shows the results for the species concentrations along the reactor length, exhibiting reasonable agreement with the detailed catalyst topology-simulated data from [7]. Preliminary findings suggest that, despite the general limitations of homogeneous reactor models stemming from inadequate physical representation, they remain valuable and computationally efficient. In the extended discussion, additional results of the proposed models will be presented for a CO₂ methanation process occurring on the irregular gyroid-like structured catalyst presented in [6].

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