

# Homogeneous modeling for laminar flows in structured catalysts: $\text{CO}_2$ methanation

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## Summary

We discuss the fluid mechanics modeling of structured catalysts for use on carbon dioxide ( $\text{CO}_2$ ) methanation. Various zero-dimensional (0-D) and three-dimensional (3-D) homogeneous models are evaluated in order to allow better informed modeling decisions.

## $\text{CO}_2$ methanation process overview

Parallel to the partial decarbonization of the energy industry by the shift to green hydrogen  $\text{H}_2$  production and utilization, a theoretical net-zero  $\text{CO}_2$  energy cycle can also reutilize  $\text{CO}_2$  by transforming it back to natural gas (methane,  $\text{CH}_4$ ). The rationale is the transport and storage of excess  $\text{H}_2$  generated during times of surplus renewable energy production. In order for the reaction to proceed with a measurable reaction rate, catalysts are required due to otherwise hindered reaction kinetics. Structured catalysts seek to balance the active surface area required for optimum heat transfer and reaction chemistry, with the flow-through output [1].

## Homogeneous modeling rationale

We favour homogeneous modeling strategies due to the complexities of large scale  $\text{H}_2$  production, either related to turbulence, or complex porous structures. Both situations may hinder the application of otherwise more reliable direct numerical simulations (DNS).

## Zero-dimensional (0-D) models

We formulate 0-D models for both  $\text{CO}_2$  methanation and steam-reforming applications (methanation reaction counterpart, used for the production of  $\text{H}_2$  from  $\text{CH}_4$ ). These models are obtained by volume-averaging theory (VAT), see also [2]. For non-adiabatic catalysts, we resort to the utilization of heat transfer models, based on the reactor transverse Péclet number  $\text{Pe}_T$ , as a way to include the effects of convective and conductive heat transfer [3, 4]. Figure 1(a) shows model results in terms of  $\text{CO}_2$  conversion for the methanation process in [5]. Since the overall porosity of the catalyst  $\epsilon$  can be used as a design variable in the representative elementary volume (REV), we are free to evaluate both the honeycomb parallel-channels (PC-HC) and the 3-D gyroid (G-3D) structured catalysts in [5]. We also evaluate the consequences of utilizing chemical kinetics mechanisms available in the literature, which include both (bulk) gas kinetics [6], as well as Ni-based catalyst surface kinetics [7]. These results are shown in Figure 1(b). Figure 2(a) shows the results for feed-temperature-dependent outlet concentrations for a steam-reforming catalyst, such as that in [8].

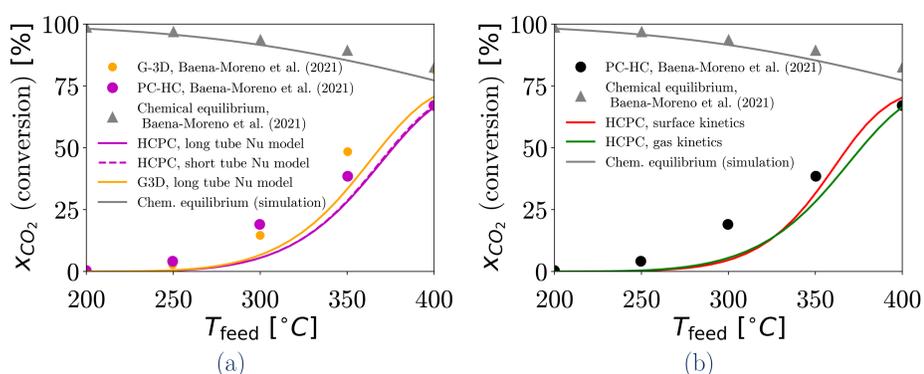


Figure 1: 0-D homogeneous  $\text{CO}_2$  methanation reactor. (a) Effect of heat transfer models and reactor geometries. (b) Effect of chemical kinetics.

## Three-dimensional (3-D) models

Using VAT, we also formulate 3-D homogeneous models. The generalization of the 0-D models to 3-D allows the inclusion of anisotropic heat convection and diffusion effects, as well as anisotropic chemical species convection and diffusion, on top of differential diffusion effects.

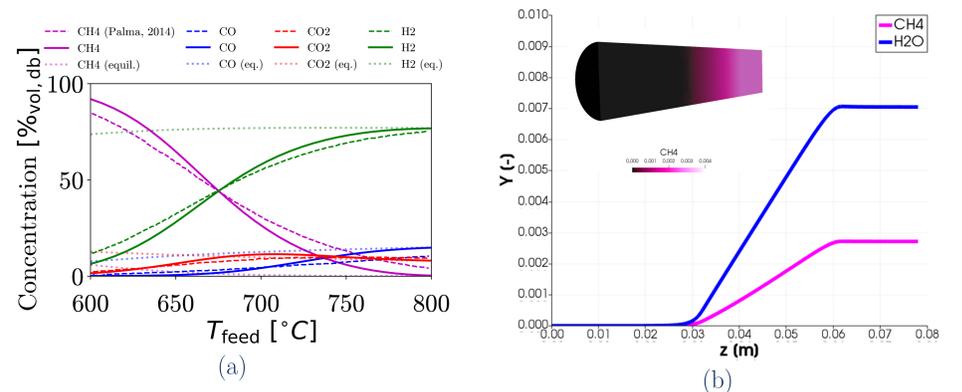


Figure 2: (a) Outlet concentrations in 0-D homogeneous steam-reforming reactor. (b)  $\text{CH}_4$  and  $\text{H}_2\text{O}$  mass fractions in a 3-D homogeneous reactor.

Figure 2(b) shows the 3-D model extension of the  $\text{CO}_2$  methanation process in [5], showing the streamwise-dependent centerline mass-fractions of  $\text{CH}_4$  and  $\text{H}_2\text{O}$ . The key issue in the formulation of the 3-D model is the adequate representation of hydrodynamic and thermodynamic total dispersion tensors. In order to facilitate an adequate modeling of the tensors, we perform detailed 3-D simulations. Figure 3(a) and (b) shows  $\text{H}_2$  mass fractions and streamwise velocity values within both the detailed and homogeneous 3-D models for the steam-reforming case in [8]. Note that the simulated geometry is shown in Figure 3(c). Fully resolved 3-D DNS of Navier-Stokes exceed our current computational capabilities, reason for which we simulate the detailed catalysts as implicit large eddy simulations (LES) with a deliberate choice of no sub-grid stress modeling. Finally, Figure 3(d) shows the homogeneous model mole fractions throughout the streamwise coordinate of the monolith.

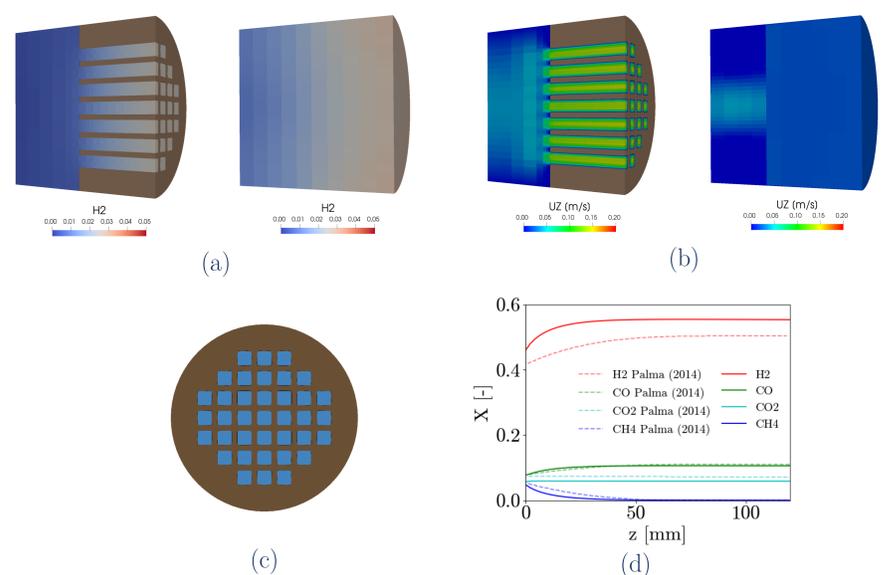


Figure 3: Comparison between 3-D detailed and homogeneous model in (a) and (b). A cross-sectional view of the geometry is shown in (c), while (d) shows the streamwise concentrations in the homogeneous model.

## Conclusions and future work

Despite the general limitations of homogeneous reactor models stemming from inadequate physical representation, these remain valuable and computationally efficient. Future work will focus on the formulation of the hydrodynamic and thermodynamic total dispersion tensors based on the detailed model results.

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