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Modeling of 3D Catalyst Pellets using Multicomponent Diffusion Models

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Highlights

- Theoretical study of multicomponent diffusion on 1D, 2D and 3D examples.
- Full coupling of mass, heat and momentum transport including Knudsen and viscous fluxes.
- Solution of reaction-diffusion problem in 3D catalyst shapes.
- Optimizing pellet geometry for carbon dioxide methanation.

1. Introduction

Theoretical studies of diffusive transport and reaction in porous media have been in the focus of chemical reaction engineering for almost one century. Maxwell-Stefan-based models are used to combine multicomponent transport of bulk, Knudsen, and surface diffusion together with complex reaction kinetics. In a recent study [1] we demonstrated the binary-friction model (BFM) [2] to be the most promising approach of this kind. Within this work the BFM is applied to carbon dioxide methanation [3] as diffusion-reaction problem. Starting with basic 1D geometries, the catalyst shapes are successively extended to 2D and actual 3D cases.

2. Methods

For a consistent description, simultaneous solution of governing equations for the transport of all species *i*, heat and total mass are essential. Molar fluxes $\vec{N}$ are implicitly given via the Maxwell-Stefan transport model. Neglecting surface diffusion as well as thermophoresis (Soret effect) the BFM can be written as [4]

$$-c_t \nabla x_i - \frac{x_i}{RT}\nabla p = \sum_{j=1}^{n_{species}} \frac{x_j \vec{N}_i - x_i \vec{N}_j}{D_{ij}^{eff}} + \frac{\vec{N}_i}{D_{i,Kn}^{eff} + \frac{B_0}{\kappa_i}}. \quad (1)$$

Transport coefficients are the Maxwell-Stefan diffusion binary coefficient D_{ij}^{eff}, the Knudsen diffusion coefficient $D_{i,Kn}^{eff}$ and the permeability of the porous catalyst B_0. The parameter κ_i denotes fractional viscous contributions of component *i*. By applying the gradient field of total pressure *p* and molar fractions x_i Eq. (1) can be rearranged to matrix form

$$\vec{N}_i = -c_t \boldsymbol{C}^{-1}\left(\nabla x_i - \frac{x_i}{RT}\nabla p\right), \quad (2)$$

where the entries of the transport matrix $\boldsymbol{C}$ are easily derived by comparing Eq. (1) with (2).

Two software packages are applied for the solution. For one and two dimensional problems the coupled differential equations are transferred to dimensional form and discretized via orthogonal collocation on finite elements using a constrained trust-region-reflective algorithm in MATLAB®. The coefficient form partial differential equation module in COMSOL Multiphysics® is used to effectively simulate the 3D shapes. Herein, a tetrahedral mesh of variable size is solved via the direct MUMPS solver.

3. Results and discussion

As a first step to modeling of multiple dimensions, the 1D MATLAB model is extend by using an axial coordinate. Figure 1 demonstrates the consistency in both dimensions. With increased *d:h*-ratio the 2D-case approaches the 1D-solution. Furthermore, distinct difference between a simple 1D case and a 2D simulation - allowing variable *d:h*-ratios - becomes apparent.

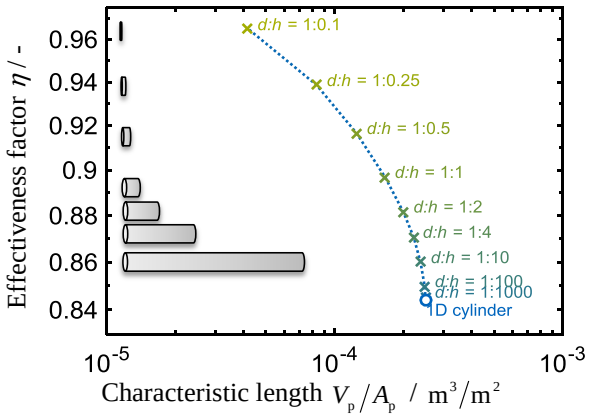

Figure 1: Proof of consistency for 1D and 2D cylinder model with varied diameter to height ratios *d:h*, simulated with MATLAB®.

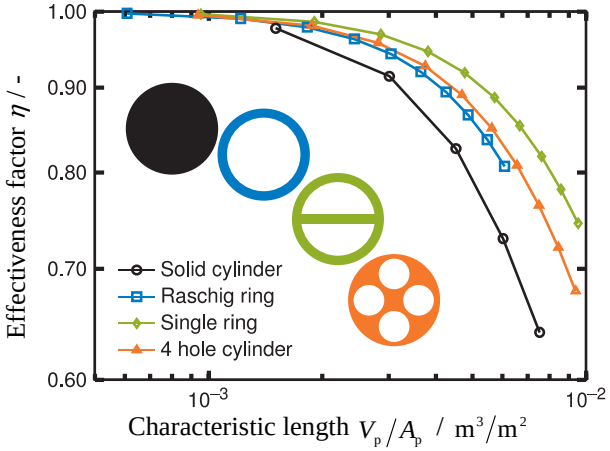

Figure 2: Evaluation of different catalyst shapes for carbon dioxide methanation, simulated with COMSOL Multiphysics® (*d:h* = 1).

Due to the high level of coupling between heat and mass transport, a sophisticated numerical methodology is essential. To achieve this for 3D pellet shapes the governing equations together with the multicomponent diffusion model are transferred to COMSOL Multiphysics. Figure 2 illustrates the dependency of the catalyst effectiveness factor η for exemplary geometries. With an increasing volume-to-surface ratios V_p/A_p the values of η are decreasing differently fast for the four shapes. However, this characteristic length is somewhat arbitrary and could be substituted with diameters of spheres with equal volume or surface. From an industrial point of view, the utilization of one (kilo)gram catalyst material is crucial. Another important aspect is catalyst stability, which among other things is strongly correlated to the exposed temperatures. To illustrated the influence of the 3D geometry on the evolution of temperature peaks inside a catalyst pellet Figure 3 depicts the temperature profile of a '4 hole cylinder' pellet.

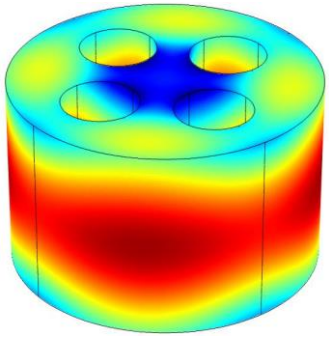

Figure 3: Exemplary temperature profile of a 4 hole cylinder.

4. Conclusions

Multicomponent diffusion is successfully applied to the reaction-diffusion problem of carbon dioxide methanation on 1D, 2D and 3D catalyst shapes. The latter is the basis for a novel, diffusion-based route in optimization of catalyst geometries.

References

[1] P.J. Donaubauer, O. Hinrichsen, 2017 (*submitted to Chem. Eng. J.*).
[2] P.J.A.M. Kerkhof, Chem. Eng. J. 64 (1996), 319–343.
[3] F. Koschany, D. Schlereth, O. Hinrichsen, Appl. Catal., B. 181 (2016), 504–516
[4] P. Capek, A. Seidel-Morgenstern, Appl. Catal., A 211 (2001), 227–237.

Keywords
Multicomponent Diffusion; Multidimensional Design; Catalyst Shape Optimization; Carbon Dioxide Methanation