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Citation for published version (APA):
Claassen, C. M. Y., Tadayon Mousavi, S., Wintjens, S., Baltussen, M. W., Peters, E. A. J. F., & Kuipers, J. A. M. (2023). *Comparison of multicomponent diffusion models in single catalytic particles and packed bed reactors*. Abstract from 11th International Conference on Multiphase Flow, Kobe, Japan.

Document status and date:
Published: 02/04/2023

Document Version:
Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
• The final author version and the galley proof are versions of the publication after peer review.
• The final published version features the final layout of the paper including the volume, issue and page numbers.

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Comparison of multicomponent diffusion models in single catalytic particles and packed bed reactors

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Keywords: Multicomponent, Diffusion model, Packed bed

Abstract

With the advances in computational power, it becomes increasingly feasible to model systems closer to their real conditions. Simplifications like constant physical properties or dilute systems are no longer a necessity. Instead, multicomponent modeling approaches are arising. One topic of interest is the diffusion model in multicomponent mixtures. A simplified diffusion mechanism can be assumed that takes the flux of component *i* just as a function of the gradient in mass/mole fraction of component *i*. But more advanced diffusion models could also be considered where the flux of component *i* depends on the gradient of all components. This latter case is more computationally expensive as it results in a coupled system. In this work, we investigate when a simplified model (Fick Wilke) suffices and when a more rigorous model (Maxwell-Stefan) is strictly required to obtain accurate results. The considered systems are single catalytic particles as well as full packed bed systems, where also hydrodynamics play an important role.

Introduction

Packed bed reactors are a commonly found reactor type in the chemical industry, pharmaceutical and biotechnological industry. To model these systems accurately we require a multicomponent modeling approach that describes the time evolution of mixture composition. A changing mixture composition does not only affect the local physical properties but also the diffusive fluxes. Simplified diffusion models, e.g. Fick's law, assume the only driving force for the flux is the component's mass or molar gradient. More rigorous models, e.g. the Maxwell-Stefan equations, also take into the gradient of the other components by considering the molar friction between all components. The latter is significantly more computationally expensive as it leads to a strongly coupled set of species conservation equations.

In this work, the simplified diffusion models will be compared with the more rigorous models to examine in which case a simplified model might be accurate enough and hence favorable with respect to computational power. This comparison will be done on the scale of a single particle and on the scale of a packed bed for various reactive systems.

Numerical Methods

The generic fluid and solid phase governing equation for the mass fraction of species i (ω_i) is:

$$\frac{\partial}{\partial t}(\rho\omega_i) + \nabla\cdot(\rho\vec{v}\omega_i) - \nabla\cdot\left(\sum_j \Gamma_{ij}\nabla\omega_i\right) = r_i \qquad (1)$$

In the fluid phase the reaction term (r_i) is zero, whereas in the solid phase the velocity in the convective term ($\nabla\cdot(\rho\vec{v}\omega_i)$) is zero or follows from Darcy's law. The diffusive term is given in a generic way. In the case of Fick diffusion, the Γ matrix is diagonal as given in Eq. 2.

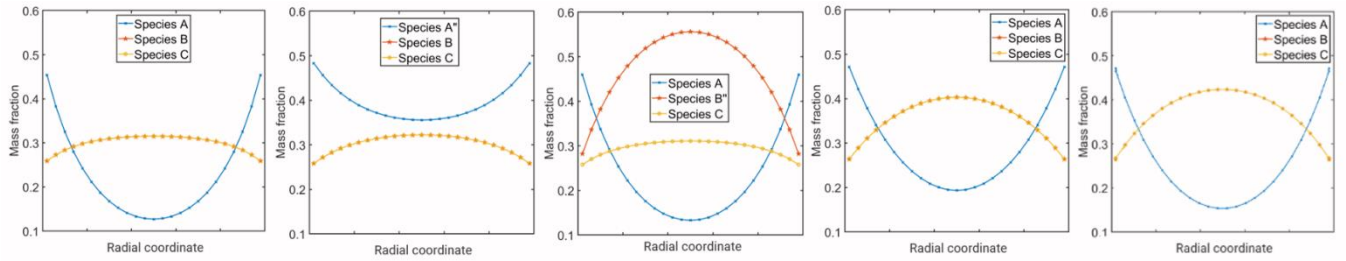
$$\Gamma_{ij} = \begin{cases} \rho D_{im} & \text{if } i = j \\ 0 & \text{if } i \neq j \end{cases} \qquad (2)$$

D_{im} is the mixture diffusivity of component i which is a function of composition, molar masses and binary diffusion coefficients according to the Wilke formula (Solsvik & Jakobsen, 2013).

For Maxwell-Stefan diffusion, the Γ matrix contains off-diagonal items. In this work the formulation of Peerenboom et al. (2011) is used.

Between the two phases a continuity of fluxes as given in Eq. 3, is assumed and enforced using the immersed boundary method implementation as presented by Chandra et al. (2020) and Deen et al. (2012) but extended to be applicable to multicomponent diffusion models.

$$\left(\rho\vec{v}\omega_i - \textstyle\sum_j \Gamma_{ij}\nabla\omega_i\right)_s \cdot \vec{n} = \left(\rho\vec{v}\omega_i - \textstyle\sum_j \Gamma_{ij}\nabla\omega_i\right)_f \cdot \vec{n} \qquad (3)$$



(a) Model 1 (b) Model 2 (c) Model 3 (d) Model 4 (e) Model 5

Figure 1: Mass fraction profiles for the decomposition reaction. If in the legend a species has ", that species is not resolved but follows from $\omega_i = 1 - \sum_{i \neq j} \omega_j$.

Results and Discussion

The different diffusion models are first compared in a simplified system where we only consider a single catalytic particle without convection, but with constant bulk conditions. Next to that we consider pseudo kinetics, i.e. not a real chemical reactive system. The pseudo reactions that are considered are a decomposition reaction that produces two isomers and a dehydrogenation reaction.

For the decomposition reaction five different diffusion models are considered. Model 1 is a traditional Fick Wilke as given in equation (2) which is solved for all three components. Model 2 and 3 are similar but only resolve 2 components. Model 4 uses a flux correction to ensure the diffusive fluxes sum to zero as proposed by Sutton & Gnoffo (1998). Finally model 5 uses the Maxwell-Stefan model. Model 4 and 5 are solved for all components.

The resulting steady state profiles inside a single particle are given in Fig. 1. Model 1 proves to be inaccurate as a close look reveals it to have an almost 25% mass loss in the particle center. Model 2 and 3 also seem to be unreliable as the resulting profiles are highly dependent on which component is not resolved. Model 4 and 5 both treat all components symmetrically and are both inherently mass conservative. The resulting profiles are comparable. Due to the flux correction the Γ matrix of model 4 is no longer a purely diagonal matrix and hence the computational favorability of Fick over Maxwell-Stefan is only marginal.

Fig. 2 shows the steady state results of the dehydrogenation reaction for model 2, 4 and 5. In this case all models result in matching profiles which would mean model 2 is favorable since it takes only 40% of the time that model 4 and 5 require.

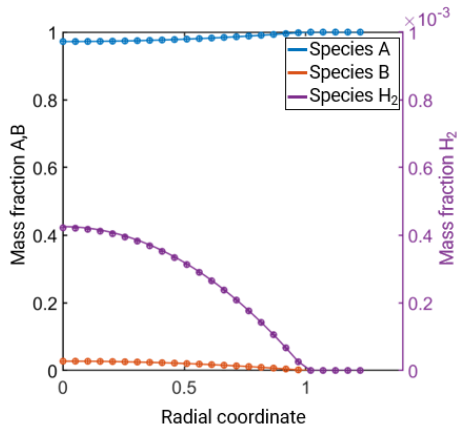


Figure 2: Mass fraction profiles for the dehydrogenation reaction. ∘ model 2, + model 4, − model 5.

How these differences between models affect the transport on packed bed level will be reported during the conference; as well as applications to real reactive systems.

Conclusion

In this study different multicomponent diffusion models have been compared on particle scale. It is shown that for certain systems the conventional Fick Wilke model does not yield trustworthy results whereas for others it performs comparable to more detailed diffusion models but using only a fraction of the computational cost. For packed bed simulations this reduced computational cost can be significant.

Acknowledgments

This work was supported by the Netherlands Organization for Scientific Research (NWO) TOP grant. First principles based multi - scale modeling of transport in reactive three phase flows/, Grant/Award Number: 716.014.001

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