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Numerical Simulation of Two-Phase Flow and Interfacial Species Transfer in Structured Packings

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This study presents the derivation and validation of a model that permits simulation of interfacial species transfer in rectification and absorption processes in the context of the algebraic Volume-of-Fluid (VoF) approach. The model is based on the Continuous Species Transfer (CST) method introduced by Marschall et al. (2012) and Deising et al. (2016), which is generalised in such a way that not only absorption but also rectification can be simulated, and is therefore termed Generalised Continuous Species Transfer (GCST) model. After briefly setting out the motivation of this generalisation, the derivation of the model is outlined, which is followed by its validation.

1. Introduction

To optimise the separation performance of structured packings in rectification columns, a thorough understanding of the hydrodynamics as well as the transport processes inside these column internals is vital. Conversely, this means that only simulations comprising both phenomena allow a comprehensive analysis of the process. However, before merging both fields, each of them is looked at individually.

In doing so, the hydrodynamics was investigated exclusively in a first step and the results achieved are presented in Hill et al. (2016). The case of single-phase gaseous flow was analysed, focusing on the visualisation and understanding of the transient flow structures occurring at higher flow rates. Moreover, a two-phase flow simulation was conducted to identify the local distribution of the liquid phase with the results compared to experimental data. Several other groups also investigated the hydrodynamics in structured packings, three of which are named in the following. Raynal and Royon-Lebeaud (2007) used a two-step approach. In a first step the liquid hold-up and the interface velocity was determined by a 2D-Simulation. In the following, single-phase simulations were performed in a periodic geometry. Thereby, the results obtained during the previous step were indirectly used by setting moving walls as boundary conditions and by adapting the calculation of the F-factor. Focusing on the multi-phase flow in commercial structured packings, Sebastia-Saez et al. (2015) investigated the dependency of the interfacial area and the liquid hold-up on the liquid load. Furthermore, visualisations of the transient flow development were presented and computed irrigated and dry pressure drops were compared to experimental results. Kenig (2017) used a periodic element to efficiently simulate the gas flow, allowing for an evaluation of the pressure drop as a function of the F-factor.

In this paper, the second step towards the simulation of separation processes inside structured packings is described, namely the development of a model to consistently incorporate the interfacial species transfer into the two-phase simulation. Several approaches for modelling species transfer can be found in the literature. One of them is the previously mentioned CST model, which uses a single-field formulation to describe the interfacial species transfer of a dilute species, based on the algebraic VoF method. To obtain a consistent single-field formulation, the conditional volume-averaging technique is applied to the local instantaneous transport equation of the volumetric molar concentration

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$$\partial_t c_i + \nabla \cdot (c_i \boldsymbol{u}) = \nabla \cdot (D \nabla c_i), \tag{1}$$

valid in both bulk phases Ω_1 and Ω_2, including the interfacial transmission condition

$$-(D\nabla c_i)_{\Sigma,1} \cdot \boldsymbol{n}_{\Sigma,1} - (D\nabla c_i)_{\Sigma,2} \cdot \boldsymbol{n}_{\Sigma,2} = 0 \tag{2}$$

and the jump condition

$$(c_i)_{\Sigma,1} + (c_i)_{\Sigma,2} = (c_i)_{\Sigma,1}\left(1 - \frac{1}{H}\right) \Leftrightarrow H = \frac{(c_i)_{\Sigma,1}}{(c_i)_{\Sigma,2}}. \tag{3}$$

Here, H denotes the Henry coefficient. Eq(3) corresponds to Henry's law and is used to close the system of equations. The derivation and formulation of the single-field equation is described in detail in Deising et al. (2016).

2. Motivation

As mentioned above, the CST model is developed to simulate the interfacial species transport of a dilute species and it is therefore well suited to the investigation of absorption processes. However, the research subject of this project is the optimisation of rectification columns, which necessitates an adaption of the model. Firstly, instead of using the molar concentration as transport variable, it is beneficial to use the molar fraction. In the absorption case where the considered species is assumed to be dilute, the molar concentrations in both phases are of similar magnitude. However, the components to be separated in rectification processes are usually the main components of the mixture, entailing molar concentrations that differ in their orders of magnitude. To avoid the resulting local jump over several orders of magnitude, which is prone to numerical instabilities, the molar concentration is selected as transport variable since it is limited between the values of zero and one. The correlation between both variables is given by

$$c_i = c x_i, \tag{4}$$

with c denoting the total molar concentration of the mixture. The corresponding local instantaneous transport equation reads

$$\partial_t c x_i + \nabla \cdot (c x_i \boldsymbol{u}) = \nabla \cdot (c D \nabla x_i) \tag{5}$$

and can be transformed into Eq(1) by using Eq(4) and assuming the molar concentration c to be constant. Analogously, the reformulated transmission condition

$$-(cD\nabla x_i)_{\Sigma,1} \cdot \boldsymbol{n}_{\Sigma,1} - (cD\nabla x_i)_{\Sigma,2} \cdot \boldsymbol{n}_{\Sigma,2} = 0 \tag{6}$$

can also be linked to Eq(2). As only binary mixtures are considered in this work, the diffusive coefficient is equal for both components, so that the subscripts are omitted:

$$D_{ij} = D_{ji} = D \tag{7}$$

The second adjustment affects the closure assumption made in the CST model. Since the CST model only covers the transport of a dilute species, Henry's law as written in Eq(3) is a valid choice to close the system of equations. In rectification processes, however, the transported species could represent any proportion of the mixture and therefore also the closure assumption must be valid for any composition of the mixture. Out of thermodynamic considerations for a binary mixture a non-linear correlation between the equilibrium molar fraction $(x_i)_{\Sigma,1}$ and $(x_i)_{\Sigma,2}$ can be derived (Hausen and Linde, 1985):

$$(x_i)_{\Sigma,2} = \frac{A_{ij}(x_i)_{\Sigma,1}}{1 + \left(A_{ij} - 1\right)(x_i)_{\Sigma,1}}, \tag{8}$$

where A_{ij} denotes the relative volatility. In Figure 1, the graphical representation of both closure assumptions is displayed. It can be seen that for small molar fractions, and therefore in the validity range of the CST model, both correlations are identical, which is due to the fact that Henry's law is a simplified version of Eq(8) for dilute species. Hence, the GCST model represents a true generalisation of the CST model.

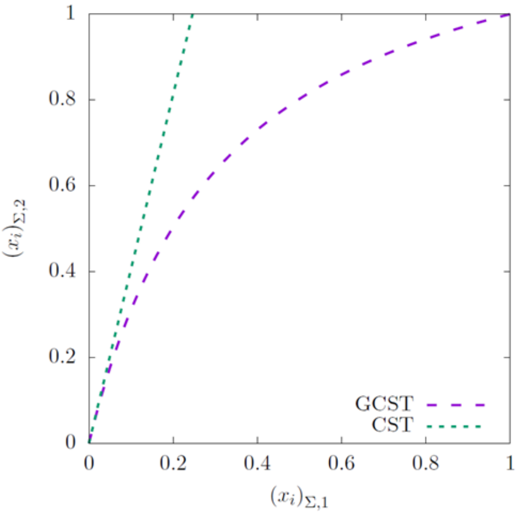

Figure 1: Closure assumption of equilibrium mole fractions presumed in GCST and CST model for $A_{ij} = 4.2$

3. Limitations of the GCST model

The model is limited to species transfer, which inherently implies a passive scalar and thus does not affect the hydrodynamics. This leads to some restrictions on possible scenarios to be simulated. In the case of absorption, the considered species needs to be dilute ensuring that the volumetric effect due to phase-change is negligible. In case of rectification this assumption generally does not hold true. However, provided the mixture consists of two components, the decoupling of hydrodynamics and species transfer is still valid if all of the following conditions are met:

- The molar heats of evaporation of components are in the same order of magnitude
- Molar masses of components are almost identical
- Mixing enthalpies are negligible
- The column is adiabatic

In addition, concerning the simulation of rectification processes, the model is only valid for binary mixtures and therefore not applicable for multicomponent mixtures.

4. Derivation of the GCST model

To arrive at a single-field formulation, the conditional volume-averaging technique is applied to the local instantaneous transport equation Eq(5) and the transmission condition Eq(6). The concept of conditional volume averaging (CVA), described by Drew and Passman (1999) and Jakobsen (2008), consists of two steps. Firstly, the equation is multiplied by a phase indicator function χ to extend its range of validity to the whole computational domain. In the case of phase 1 the phase indicator function would read:

$$\chi_1(t) = \begin{cases} 1 & \text{in } \Omega_1(t) \\ 0 & \text{otherwise} \end{cases} \tag{9}$$

Secondly, the conditioned equation is volume averaged, which leads to a mathematical form suitable for consistent discretisation using the finite volume method. Performing both steps on Eq(5) yields for phase 1

$$\overline{\chi_1 \partial_t c x_i} + \overline{\chi_1 \nabla \cdot (c x_i \boldsymbol{u})} = \overline{\chi_1 \nabla \cdot (c D \nabla x_i)} \tag{10}$$

and for phase 2

$$\overline{\chi_2 \partial_t c x_i} + \overline{\chi_2 \nabla \cdot (c x_i \boldsymbol{u})} = \overline{\chi_2 \nabla \cdot (c D \nabla x_i)}, \tag{11}$$

respectively. In combination with the also conditioned and averaged transmission condition Eq(6) and by applying the rules of CVA-theory, the following single-field equation can be derived:

$$\partial_t \left(\left(\alpha_1 \overline{c}^1 + \alpha_2 \overline{c}^2 \right) x_{i,m} \right) + \nabla \cdot \left(\left(\alpha_1 \overline{c}^1 + \alpha_2 \overline{c}^2 \right) x_{i,m} \overline{\boldsymbol{u}} \right) = \nabla \cdot \left(\alpha_1 \overline{c}^1 \, \overline{D}^1 \nabla \overline{x}_i^1 + \alpha_2 \overline{c}^2 \, \overline{D}^2 \nabla \overline{x}_i^2 \right) \tag{12}$$

Besides the mixture mole fraction

$$x_{i,m} = \frac{\alpha_1 \overline{c}^1 \overline{x}_i^1 + \alpha_2 \overline{c}^2 \overline{x}_i^2}{\alpha_1 \overline{c}^1 + \alpha_2 \overline{c}^2} \tag{13}$$

the phasic averaged mole fractions $\overline{x}_i^1$ and $\overline{x}_i^2$ are also unknown, thus having an unclosed single-field formulation. As described in the previous chapter, Eq(8) is used for closure. However, to conduct the closure, the equilibrium molar fractions $(x_i)_{\Sigma,1}$ and $(x_i)_{\Sigma,2}$ have to be linked to the phasic averaged mole fractions $\overline{x}_i^1$ and $\overline{x}_i^2$. This is done by following the work of Marschall et al. (2012) stating:

$$\begin{aligned} (x_i)_{\Sigma,1} &\approx \overline{x}_i^1 \\ (x_i)_{\Sigma,2} &\approx \overline{x}_i^2 \end{aligned} \tag{14}$$

This assumes that the local mole fraction at the interface equals the volume averaged phasic mole fraction. In consequence, the accuracy of the model strongly depends on a sufficient spatial resolution of the interface, especially when high mole fraction gradients are expected.

Using the relations in Eq(14), Eq(8) can be rewritten as

$$\overline{x}_i^2 = \frac{A_{ij} \overline{x}_i^1}{1 + (A_{ij} - 1) \overline{x}_i^1} \, . \tag{15}$$

Apparently, Eq(15) is a non-linear correlation. However, a linear correlation is needed to close the single-field formulation and keep it a single equation. The approach applied in this study to circumvent this problem is to discretise Eq(15) semi implicit:

$$\overline{x}_i^2 = \underbrace{\frac{A_{ij}}{1 + (A_{ij} - 1)\left(\overline{x}_i^1\right)^{old}}}_{K} \, \overline{x}_i^1 = K \, \overline{x}_i^1 \tag{16}$$

Using Eq(16) and Eq(13) finally leads to the closed single field formulation:

$$\begin{aligned} \partial_t \left(\left(\alpha_1 \overline{c}^1 + \alpha_2 \overline{c}^2 \right) x_{i,m} \right) + \nabla \cdot \left(\left(\alpha_1 \overline{c}^1 + \alpha_2 \overline{c}^2 \right) x_{i,m} \overline{\boldsymbol{u}} \right) \\ = \nabla \cdot \left(\alpha_1 \overline{c}^1 \, \overline{D}^1 \nabla \left(x_{i,m} \frac{\alpha_1 \overline{c}^1 + \alpha_2 \overline{c}^2}{\alpha_1 \overline{c}^1 + K \alpha_2 \overline{c}^2} \right) \right) + \nabla \cdot \left(\alpha_2 \overline{c}^2 \, \overline{D}^2 \nabla \left(x_{i,m} K \frac{\alpha_1 \overline{c}^1 + \alpha_2 \overline{c}^2}{\alpha_1 \overline{c}^1 + K \alpha_2 \overline{c}^2} \right) \right) \end{aligned} \tag{17}$$

5. Validation of the GCST model

To solve the derived single-field formulation numerically, Eq(17), in the exact form as stated above, is discretised by using the finite volume method and temporally by using the implicit Euler method. The fully discretised equation is then implemented into a transient solver environment.

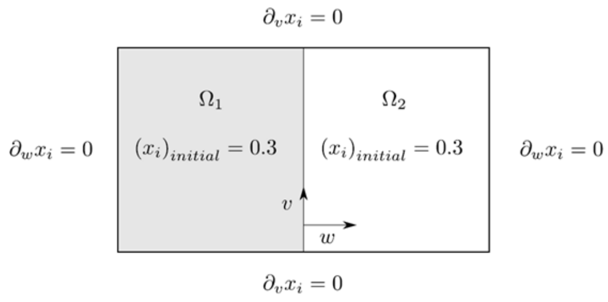

Figure 2: Set-up of test case to validate GCST model.

Since an analytical solution is needed to validate the model, the test cases are limited to simple set-ups. The considered set-up for validation is displayed in Figure 2. The computational domain comprises a two-phase region with both phases being stagnant. Throughout the domain, an initial mole fraction of $x_i = 0.3$ is assumed. On all boundaries $\partial_v x_i = 0$ and $\partial_w x_i = 0$ respectively is imposed which leads to a 1D type of problem. By setting the relative volatility to $A_{ij} = 4.2$ the initial set-up is not stable but interfacial species transfer will occur transiently changing the profile of the molar fraction.

To facilitate the analytical calculation, the molar fraction in Ω_1 must be constant. This can be achieved by setting the fluid properties to meet the following constraints:

$$\begin{aligned} &\overline{c}^1\,\overline{D}^1 \gg \overline{c}^2\,\overline{D}^2 \\ &\overline{c}^1 \gg \overline{c}^2 \end{aligned} \tag{18}$$

Table 1 lists the specified fluid properties coping with the constraints stated above. It should be noted that the selected parameters make no claim to have any physical meaning but are specifically chosen to fulfil Eq(18).

Table 1: Fluid properties

	Ω_1	Ω_2
c in $\frac{\mathrm{mol}}{\mathrm{m}^3}$	100	1
D in $\frac{\mathrm{m}^2}{\mathrm{s}}$	1e-4	1e-6

For region Ω_2, which is assumed to be a semi-infinite region, the profile of the molar fraction eventually can be calculated analytically as a function of space and time (Welty et. al., 2009):

$$x_i = \operatorname{erf}\left(\frac{w}{2\sqrt{\overline{D}^2 t}}\right)\left((x_i)_{initial} - \frac{A_{ij}(x_i)_{initial}}{1 + (A_{ij} - 1)(x_i)_{initial}}\right) + (x_i)_{initial} \tag{19}$$

In addition, the analytical solution of the profile of the molar flux $j_i^\star$ is given by (Polifke and Kopitz, 2009)

$$j_i^\star = -\frac{\sqrt{\overline{D}^2}\left((x_i)_{initial} - \frac{A_{ij}(x_i)_{initial}}{1 + (A_{ij} - 1)(x_i)_{initial}}\right)\overline{c}^2}{\sqrt{\pi t}} \exp\left(-\left(\frac{w}{2\sqrt{\overline{D}^2 t}}\right)^2\right). \tag{20}$$

After spatially discretising the domain once with 101 and once with 1001 evenly spaced cells, the set-up described is simulated using the GCST model. The evaluation of the test case after $t = 2.5\,s$ is depicted in Figure 3. The obtained profiles of the molar fraction as well as the profile of the molar flux in the case of 101 cells show sufficient, in the case of 1001 cells show good agreement with the analytical solution. Against the background of the single-field formulation used by the GCST model, special notice should be given to the good rendition of the interfacial molar fraction jump.

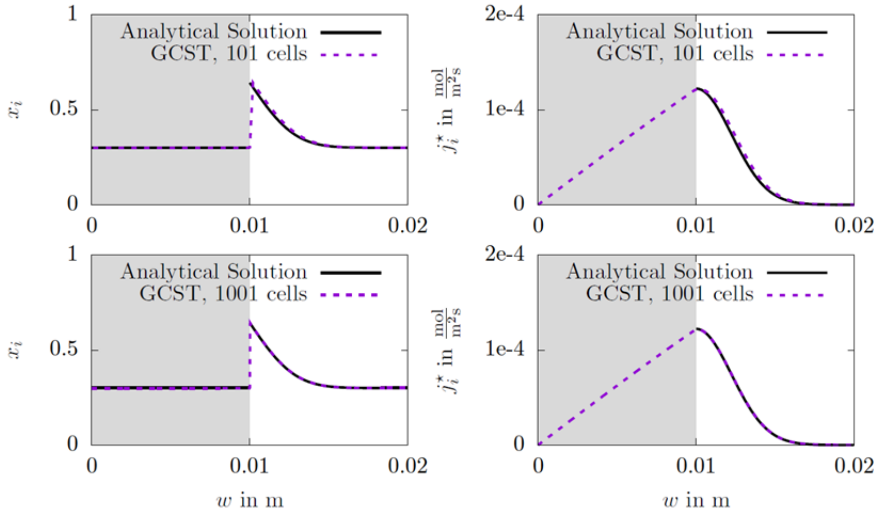

Figure 3: Profiles of the molar fraction (left) and molar flux (right) at $t = 2.5\,s$ using 101 cells (top) and 1001 cells (bottom)

6. Conclusions and Outlook

This study presents the novel GCST model, which is a generalisation of the CST model, enabling the simulation of simple rectification processes. This is done by changing the transport variable from molar concentration to molar fraction. In addition, the closure assumption is generalised, which is now valid for any binary mixture of any composition. By consequently applying conditional volume averaging to the underlying transport equation and subsequent transmission condition, a single-field formulation is derived that is readily available for a consistent discretisation using the finite volume method. The model is validated by comparing the simulation result of a planar diffusion case to its analytical solution. Sufficient resolution provided the result perfectly matches the solution. Even though the test case is a 1D set-up, the model readily works in 3D.

Since the GCST model is developed inside the algebraic VoF framework, the method is applicable to any solver using this framework, either on structured or unstructured grids. Combining the model presented with, for example, a 3D simulation of the hydrodynamics in a structured packing will allow its separation performance to be predicted, whereby only a small set of physically justifiable fluid and mixture properties have to be defined. This will not only help to optimise packing geometries but also to gain a deeper insight into the process itself. Conducting the simulation of the hydrodynamics and species transfer in a structured packing is ongoing work and results as well as a detailed description of the setup are to be published.

Symbols		
A_{ij}	Relative volatility	
c	Volumetric molar concentration	$\frac{\mathrm{mol}}{\mathrm{m}^3}$
D	Diffusion coefficient	$\frac{\mathrm{m}^2}{\mathrm{s}}$
H	Henry coefficient	
$\boldsymbol{n}$	Normal vector	
$\boldsymbol{u}$	Velocity vector	$\frac{\mathrm{m}}{\mathrm{s}}$
x	Molar fraction	
α	Volume fraction	
$\overline{\phi}$	Volume averaged quantity	
$\overline{\phi}^{1,2}$	Phasic averaged quantity of phase 1, 2	

Subscripts	
1	Phase 1
2	Phase 2
i	Component i of binary mixture
j	Component j of binary mixture
Σ	Parameter evaluated on interface

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