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Rigorous modelling of multicomponent mass transfer in reactive absorption

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In the last years, many works on the modelling of multicomponent reactive separation using the Maxwell-Stefan relations have been published. However, most of them use a bootstrap equation and need a film thickness. Thus, we have developed a general model of multicomponent mass transfer in reactive absorption for ionic system using continuum mechanics to describe the local phenomena in each phase. This description is based on the works of Truesdell [1] and Slattery [2]. It does not require any mass transfer coefficient or film thickness.

Model description

Using the transport theorem, the balances of each conservative quantity could be written as an equation of conservation applied to each point of the continua. In order to describe the evolution of the n_c various components we require to solve the conservation of mass for species $i \in \{1, \dots, n_c - 1\}$.

$$\frac{\partial}{\partial t}(\rho_i) + \vec{\nabla} \cdot (\rho_i \vec{v}_i) = R_i M_i, \quad i \in \{1, \dots, n_c - 1\} \quad (1)$$

The reaction term R_i is described as proposed by Bilardello et al. [3]

$$R_i = \sum_{j=1}^{n_r} \nu_{ij}^c \Gamma_j + \sum_{k=1}^{n_{re}} \nu_{ik}^c \xi_k \quad (2)$$

It takes into account the n_{rc} kinetically controlled chemical reactions and the n_{re} instantaneously balanced chemical reactions, as dissociation equilibrium in aqueous phase. The enhancements of these equilibrium are obtained implicitly thanks to chemical equilibrium equations.

$$\sum_{i=1}^n \nu_{ik} \mu_i = 0, \quad k \in \{1, \dots, n_{re}\} \quad (3)$$

As we use only $n_c - 1$ equations of continuity for species, we have to solve the equation for overall conservation of mass.

$$\frac{\partial}{\partial t}(\rho) + \vec{\nabla} \cdot (\rho \vec{v}) = 0 \quad (4)$$

This equation allows to prevent the total mass balance from numerical approximations.

Then, in order to describe the movement of the various constituents, we need to solve equations of momentum conservation. As we do not know any fluid behaviour that can express the stress tensor for species, we use an approximate equation based on the Maxwell-Stefan approach [2] [4]

$$-RT \sum_{j=1}^n \frac{c_i c_j}{c_i D_{ij}} \frac{(\vec{v}_i - \vec{v}_j)}{c_i} = c_i \vec{\nabla} \mu_i + c_i s_i \vec{\nabla} T - \frac{\rho_i}{\rho} \vec{\nabla} P + \frac{z_i \rho_i}{M_i} F^0 \vec{\nabla} \varphi, \quad i \in \{1, \dots, n_c - 1\} \quad (5)$$

with φ approximated by

$$\vec{\nabla} \varphi = \frac{RT \sum_{j=1}^{n-1} z_j D_{jn}^0 \vec{\nabla} c_j}{F^0 \sum_{j=1}^{n-1} z_j^2 D_{jn}^0 c_j} \quad (6)$$

It is an approximation of the momentum conservation for species, thus we choose to use only n_c-1 Maxwell-Stefan equations. In order to describe the behaviour of the whole phase, we use the conservation of the overall momentum with the stress tensor achieved from the Newtonian model

$$\frac{\partial}{\partial t}(\rho_i \vec{v}) + \vec{\nabla} \cdot (\rho_i \vec{v} \otimes \vec{v}) - \vec{\nabla} \cdot \left[(-P + \lambda \vec{\nabla} \cdot \vec{v}) \mathbb{I} + 2\eta \overline{\overline{D}} \right] - \rho_i \vec{g} = 0 \quad (7)$$

Finally to describe the evolution of temperature, the conservation of energy is required. Thus, neglecting the viscous and the diffusive dissipations and the effect of mass transfer on heat transfer (Dufour effect), the energy balance is

$$\frac{\partial}{\partial t} \left(\rho_i \left(\tilde{u} + \frac{1}{2} \vec{v}^2 \right) \right) + \vec{\nabla} \cdot \left(\rho_i \left(\tilde{u} + \frac{1}{2} \vec{v}^2 \right) \vec{v} \right) - \vec{\nabla} \cdot (\kappa \vec{\nabla} T) - \rho_i Q = 0 \quad (8)$$

To close the system, some summation equations are needed: summation of mass concentration and summation of mass flow.

Resolution and application

The steady state resolution for an isothermal study is performed in a two-dimensional geometry. The overall two-dimensional system of differential and algebraic equations is solved using a finite differences method of discretisation. This method generates a non-linear algebraic system solved by a Newton-Raphson's method taking into account the sparse structure of the Jacobian matrix. We obtain the profiles of concentration, temperature, pressure and velocity at each point of the geometry. The absorption of carbon dioxide in soda is investigated for different loading pH. An example of result is presented on figure 1. It shows the evolution of CO₂ concentration in a counter current gas-liquid system at pH = 10. These results obtained without any transfer coefficient are in agreement with reactive absorption theory. Thus, the concentration is greater at the interface and decrease progressively toward the bulk. A pH study shows the influence of the hydroxide ions on the mass transfer of CO₂ in the soda solution and on the concentration profiles. These results are not presented in this paper but are available.

Nomenclature

c molar concentration, mol.m ⁻³	\tilde{u} mass internal energy, J.kg ⁻¹
D Maxwell-Stefan diffusivity, m ² .s ⁻¹	\vec{v} velocity, m.s ⁻¹
D° Diffusivity at infinite dilution, m ² .s ⁻¹	z_i ionic charge
$\overline{\overline{D}}$ deformation tensor, s ⁻¹	κ thermal conductivity, W.m ⁻¹ .K ⁻¹
F° Faraday constant, C.mol ⁻¹	λ bulk viscosity, Pa.s
g gravity, m.s ⁻²	η dynamic viscosity, Pa.s
M_i molar mass, kg.mol ⁻¹	φ electrostatic potential, V
P pressure, Pa	μ mass chemical potential, J.kg ⁻¹
Q energy source, J.s ⁻¹ .kg ⁻¹	ν stoichiometric coefficient of chemical reaction
R_i rate of chemical reaction, kg.m ⁻³ .s ⁻¹	ρ_i mass concentration of i , kg.m ⁻³
R gas constant, 8,314 J.mol ⁻¹ .K ⁻¹	ρ_t overall mass concentration, kg.m ⁻³
s entropy, J.kg ⁻¹ .K ⁻¹	ξ enhancement of chemical equilibrium, kg.m ⁻³ .s ⁻¹
T temperature, K	

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Figure 1 : CO₂ concentration evolution in a counter current gas-liquid system at pH = 10.

