Numerical Simulation of Mass Transfer between a Single Drop and an Ambient Flow

W. H. Piarah, A. Paschedag, and M. Kraume
Dept. of Chemical Engineering, Technical University Berlin, D-10623 Berlin, Germany

The calculation of mass transfer is one of the key problems in the design of liquid-liquid extractors. Mass transfer between drops and the continuous phase has been studied quite extensively, and a number of models has been developed. Kronig and Brink (1950) derived a theoretical solution to calculate the mass transfer from circulating droplets based on the flow patterns of the creeping flow regime ($Re < 1$). However, experimental results often exceed that predicted by Kronig and Brink (Skelland and Wellek, 1964; Brounshtein et al., 1970). Other theoretical investigations were carried out numerically by Brounshtein et al. (1970) and Brauer (1979). Their results for the case of predominant resistance to mass transfer inside the droplet are close to that of Kronig and Brink. To improve the agreement between the theoretical formulation and experimental result, some authors (Handlos and Baron, 1957; Steiner, 1986; Temos et al., 1996; Henschke and Pfennig, 1999) applied extended mass-transfer models possessing a special type of turbulent circulation in the droplet. However, of course, these models contradict the fact that due to all observations internal circulation in the droplet is laminar. Furthermore, most theoretical investigations on this subject (Kronig and Brink, 1950; Handlos and Baron, 1957; Brauer, 1979) were carried out for the limiting case of a transfer controlling resistance solely inside the droplet (internal problem). Actually, in liquid-liquid extraction the resistance to mass transfer in the continuous phase cannot be neglected (conjugate problem).

To overcome the above mentioned problems, a model formulation and solution method will be presented in this article, which can be used to calculate the concentration fields inside and outside of the droplet numerically without the assumption of turbulence inside the droplet.

Theoretical Formulation and Method

We consider the case of a spherical droplet moving in an infinite volume of another liquid with a soluble substance in both phases. The basic assumptions are:

- The droplet's shape and volume remain constant.
- Both fluids are Newtonian and incompressible.
- During mass transfer, the physical properties of the droplet and of the continuous phase are constant and interfacial mass transfer occurs only for the solute.
- The flow fields inside and outside of the droplet are steady and axisymmetric.
- There are no surface-active contaminants.

In this case the equation of motion for both phases may be written as follows:

$$-\rho_i \vec{w}_i \nabla \vec{w}_i - \nabla p_i + \mu_i \nabla^2 \vec{w}_i + \rho_i \vec{g} = 0 \qquad (1)$$

and the continuity equation reads

$$\nabla \vec{w}_i = 0 \qquad (2)$$

where the subscript i is equal 1 for the droplet and 2 for the continuous phase.

The boundary conditions related to this problem in spherical coordinates (θ - polar angle coordinate; r - radial coordinate) are:

At the droplet interface

$$\mu_1\left(\frac{\partial w_\theta}{\partial r} - \frac{w_\theta}{r}\right)_1 = \mu_2\left(\frac{\partial w_\theta}{\partial r} - \frac{w_\theta}{r}\right)_2 \qquad (3)$$

Correspondence concerning this article should be addressed to M. Kraume.

$$\left(p+\mu_1\frac{\partial w_r}{\partial r}\right)_1=\left(p+\mu_2\frac{\partial w_r}{\partial r}\right)_2+\frac{2\gamma}{R} \tag{4}$$

At the outer boundary of the domain (100 droplet diameters away from the particle)

$$\vec{w}_2=\vec{w}_\infty \tag{5}$$

On the symmetry axis

$$\frac{\partial \vec{w}_i}{\partial \theta}=0 \tag{6}$$

The governing transport equation for the unsteady mass transfer of solute A and the related initial and boundary conditions read as follows

$$\frac{\partial c_{Ai}}{\partial t}+\nabla(\vec{w}_i c_{Ai})=D_i\nabla^2 c_{Ai} \tag{7}$$

Initial Conditions:

inside the droplet $\quad c_{Ai}=c_{A10}$ (8)

in the continuous phase $\quad c_{A2}=c_{A2\infty}$ (9)

Boundary Conditions:
at the droplet interface

$$D_1\frac{\partial c_{A1}}{\partial r}=D_2\frac{\partial c_{A2}}{\partial r} \tag{10}$$

$$c_{A1\text{in}}=mc_{A2\text{in}} \tag{11}$$

at the outer boundary of the domain (100 droplet diameters away from the particle)

$$c_{A2}=c_{A2\infty} \tag{12}$$

on the symmetry axis

$$\frac{\partial c_{Ai}}{\partial \theta}=0 \tag{13}$$

For the numerical solution of the differential equations above, the commercial CFD code STAR-CD has been employed. Since the problem is axisymmetric, in spherical coordinates (r, θ, φ) all the parameters such as velocity, pressure, and concentration are independent of the coordinate φ. Therefore, the numerical computation is performed in a semicircular (r, θ) domain. Grid sensitivity analysis proved that an accurate solution is obtained using 30,000 cells, with a more dense grid near the interface between the droplet and the continuous phase. The change of grid size in radial direction is presented in Figure 1.

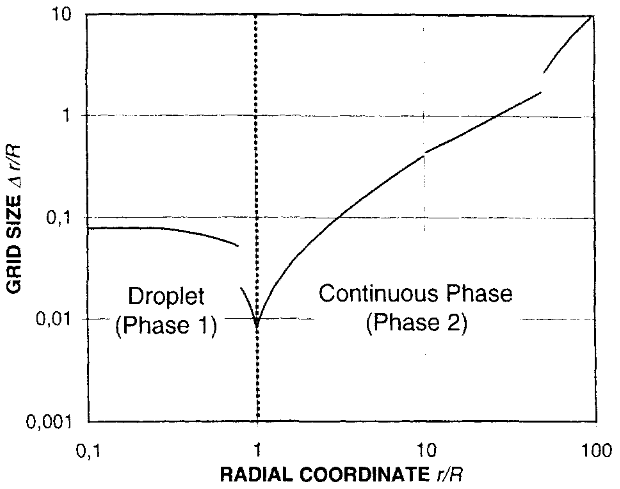

Figure 1. Change of grid size in radial direction.

Results and Discussion

Henschke and Pfennig (1999) have carried out experiments to investigate the transfer of acetone between a single droplet of n-butyl acetate and water as a continuous phase. In this case, the resistance to mass transfer in the continuous phase is of comparable order of magnitude to that in the dispersed phase which requires consideration as a conjugate problem. To validate our numerical calculation, the experiments of Henschke and Pfennig were simulated and both results have been compared. Figure 2 shows that the numerical results of the conjugate problem agree well with the experimental data of Henschke and Pfennig. The deviation of the results is especially small at a large Fourier number when the experimental accuracy should be better. Furthermore, it can be seen that the average droplet concentration calculated by Brounshtein et al. (1970) decreases slower than our numerical results due to smaller Reynolds numbers. The numerical results of Brounshtein et al. are based on the flow pattern of the creeping flow regime ($Re<1$).

In addition, the same case ($Re=2\rho_2 w_\infty R/\mu_2=292$, $\mu^*=\mu_1/\mu_2=0.73$ and $Pe=2w_\infty R/D_1=1.34\times10^5$) was described assuming a pure internal problem and the results were compared with those of Handlos and Baron (1957) and Kronig and Brink (1950). Handlos and Baron's model is based on the

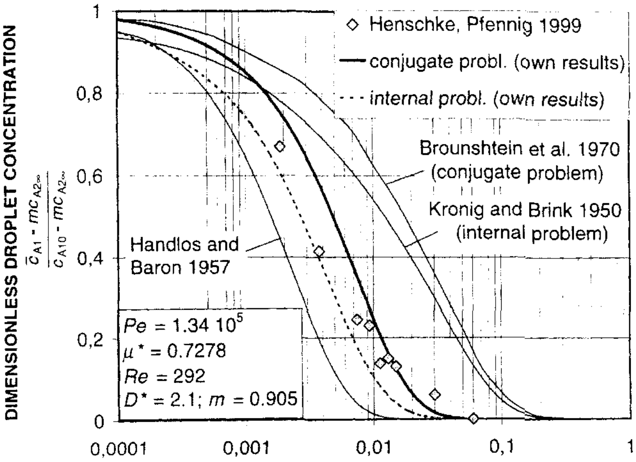

Figure 2. Average concentration in the droplet as function of Fourier number.

assumption that the circulation in the droplet is fully developed ($Re \approx 1{,}000$); thus, it can be considered as an upper limit of mass transfer. As expected, the average concentration in the droplet of our simulation decreases slower than that of Handlos and Baron because of a smaller Reynolds number (see Figure 2). Compared to the experimental data, our calculation of the internal problem leads to a faster decrease of the average concentration due to the absence of the continuous phase mass-transfer resistance. On the other hand, there is a significant difference between the numerical results of Kronig and Brink and our calculation. It can be seen that the average droplet concentrations calculated by Kronig and Brink decrease slower than our calculation due to a smaller Reynolds number. As mentioned above, Kronig and Brink derived a theoretical solution based on the flow pattern of the creeping flow regime ($Re < 1$).

After the verification of the simulation, some further investigations were carried out dealing with the influence of the Reynolds number on mass transfer for the conjugate problem which has not been done so far. Other parameters that influence mass transfer such as diffusivity ratio $D^* = D_1/D_2$, distribution coefficient $m = c_{A1in}/c_{A2in}$ and Peclet number Pe are considered constant. To illustrate the situation in which the resistance to mass transfer in the continuous phase is of comparable order of magnitude to that in the dispersed phase, the value of diffusivity ratio D^* and equilibrium constant m were chosen to be 1. In this article only the conditions $Pe = 100$ and Reynolds numbers from 0,1 (creeping flow) to 100 are considered.

The influence of the Reynolds number on the time-dependent average concentration inside the droplet is shown in Figure 3. For the conditions investigated, the contribution of the convection to the mass transfer compared to that of the diffusion is relatively small ($Pe = 100$). Therefore, a Reynolds number variation from 0.1 to 1 does not show any significant influence on mass transfer. When the Reynolds number is increased from 1 to 10, the change of the time-dependent concentration becomes distinct. In general, mass transfer is improved with increasing Reynolds number. This is underlined by the Sherwood number $Sh = 2\beta R/D_1$ presented in Figure 4 for the same parameters as considered in Figure 3. In Figures 3 and 4 the internal problem is shown for comparison (depicted by a dashed line). As mentioned above, mass transfer for the conjugate problem is slower than that for the internal problem. For the cases investigated, it was found that the average concentration in the droplet of the conjugate problem decreases approximately three times slower than that of the internal problem which corresponds to an approximately 65% smaller Sherwood number.

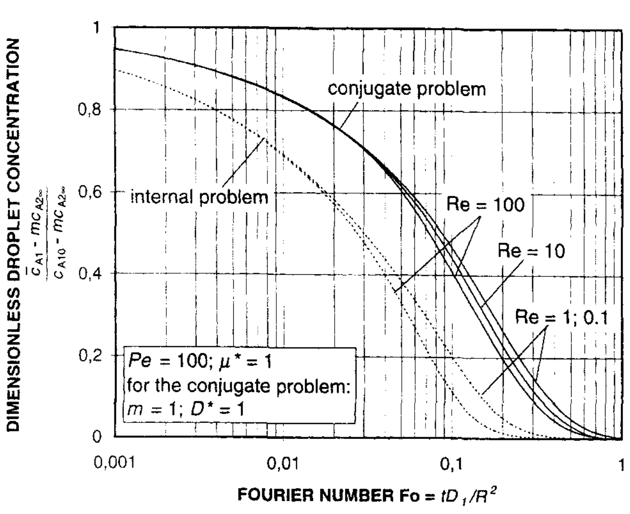

Figure 3. Influence of the Reynolds number on the average concentration in the droplet.

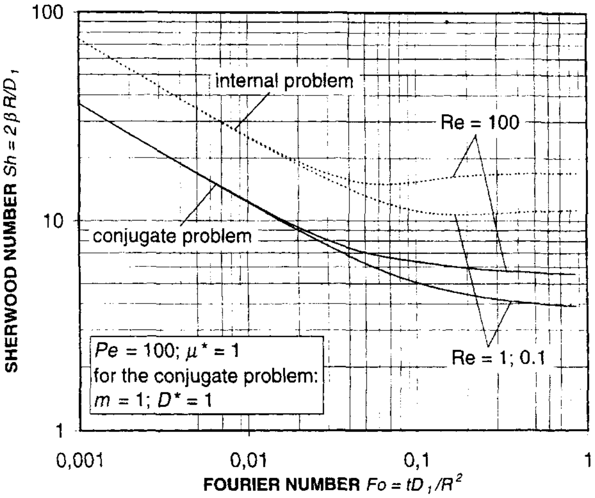

Figure 4. Sherwood number as function of Fourier number.

Notation

c = concentration (kg/m^3)
$\bar{c}$ = average concentration in the droplet (kg/m^3)
$D^* = D_1/D_2$ = diffusivity ratio
g = gravitational acceleration (m/s^2)
$m = c_{A1in}/c_{A2in}$ = distribution coefficient
p = pressure (N/m^2)
R = radius of the droplet (m)
t = time (s)
θ = polar angle coordinate (°)
w = velocity (m/s)
$\beta = (-R/3t)\ln((\bar{c}_{A1} - mc_{A2\infty})/(c_{A10} - mc_{A2\infty}))$ = mass-transfer coefficient (m/s)
$\mu^* = \mu_1/\mu_2$ = viscosity ratio
γ = surface tension (N/m)
ρ = density (kg/m^3)

Subscripts

A = solute A
in = interface
0 = initial condition
∞ = large distance from the droplet

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