



## Numerical simulation of multi-component mass transfer in rigid or circulating drops: Multi-component effects even in the presence of weak coupling

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### ABSTRACT

Numerical simulation results of mass transfer to and from drops with applications to liquid–liquid extraction processes are considered. Multiple solute components (specifically 2 solute components) are assumed to be present in the drop. The system is described using the theory of multi-component mass transfer, in which a flux of one component can be coupled to a concentration gradient in the other. The nominal strength of this coupling is determined by the off-diagonal elements of a diffusivity matrix. Naively it might be thought that, if the off-diagonal matrix elements are small compared to the diagonal ones, then the influence of coupling between components is essentially negligible. It is shown however that this is not always the case. Particular focus is given to the case where one solute component has an imposed concentration difference between the drop interior and the drop surface, whilst the other solute has no such difference imposed. Mass transfer is still observed for the latter component, which is a clear indication of coupling due to multi-component diffusion effects. The mass fraction of the component with no imposed concentration difference evolves first by deviating from its initial value, but later returns back to this initial value. It is possible to place a bound on the extent of this deviation in terms of the elements of the diffusivity matrix and any concentration difference imposed on the other component. Circulation flow, if present within the drop, is found only to have a weak influence on the maximum extent of the aforementioned deviation. It has however a role in speeding up the rates of deviation and subsequent return of component mass fraction compared to a non-circulating rigid drop case. Circulation also determines the order in which individual pointwise locations in the drop experience this deviation and subsequent return: only points near the drop surface experience a rapid evolution in the absence of circulation, whereas points either near the surface or near the axis evolve rapidly in the presence of circulation.

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### 1. Introduction

Mass transfer by liquid–liquid extraction is important in many chemical engineering operations [1–5]. During liquid–liquid extraction, one or more solutes transfers to or from dispersed drops of one solvent phase to a surrounding continuous phase of an immiscible solvent. The mass transfer process is driven by diffusion from one phase to the other immiscible phase. One complicating feature however is that when more than one solute is present, it is possible to have multi-component mass transfer in which a concentration gradient of one component drives a flux of another [6–17]. In order to analyse this complicating feature it is sufficient to con-

sider a ternary system, with two solutes and one solvent present (and this is what will be studied here), although it should be emphasised that the multi-component mass transfer theory applies more generally to a system with an arbitrary number of components.

Coupling between concentration gradients and fluxes of different components can have serious consequences for the liquid–liquid extraction process. The nominal strength of the coupling is measured by off-diagonal elements of a diffusivity matrix [16]. When these off-diagonal elements are large, strong coupling between solute components is expected, and multi-component effects certainly cannot be ignored. The converse however is not true. Even in the case where off-diagonal elements are small (compared to diagonal ones), it might not be possible to neglect multi-component effects altogether. Suppose, for example in the case where two solutes are present, that solute 1 is the one we are trying to transfer and solute 2 is a contaminant present in both dispersed drop and surrounding immiscible continuous phases. If the transfer of solute 1 also drives transfer of solute 2, then the

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concentration of solute 2 in either solvent phase will change over time, even if the concentration (or more generally chemical potential [16,18]) of solute 2 is initially equal in both phases, meaning that there is no mass transfer driving force for solute 2 on its own. It is possible that the level of the contaminant solute 2 might be acceptable initially (for some desired target product purity) but evolves over time to become unacceptable. Even very small changes in the level of solute 2 might need to be determined via the theory of multi-component mass transfer if the initial concentration level is near to some critical purity. It therefore needs to be determined under what circumstances these multi-component effects are important, and under what circumstances a simpler single component theory might be acceptable. This may depend not only on the size of the off-diagonal diffusivity matrix elements, but also on the initial and boundary conditions applied to the solute concentrations: such a case will be studied here.

Apart from multi-component effects, another complicating feature of mass transfer in liquid–liquid extraction is circulation [19–27]. It has been described [24–26] how circulation drives material around an internal stagnation point located typically quite close to the equatorial plane and at a radial coordinate about 0.7 of the radius of the drop. Circulation speeds up mass transfer compared to a rigid (non-circulating) drop by advecting material from the drop surface to the drop interior. In particular, transferred material is carried by the circulation from near the drop surface to along the drop axis.

However if the streamline pattern within the drop is laminar [26], which is often the case [24,25,28,29], the system still relies on diffusive mass transfer to take material from the surface-and-axis towards the internal stagnation point. This problem has been analysed in detail [26], but it was claimed that the systems studied (although formally multi-component ones) actually behaved quite similar to single component ones. It is the wish to observe ‘true’ multi-component effects including the presence of circulation which has prompted the present work.

This paper is laid out as follows. In the next section (i.e. Section 2), the theory of multi-component mass transfer is introduced. After that a special case of multi-component mass transfer is considered (Section 3), whereby one of the components has no imposed concentration difference: this leads to some simplifications in the formulation, and also some insightful physical interpretations of the resulting multi-component coupling terms. Section 4 briefly describes the numerical methodology used to solve the multi-component mass transfer equations. Numerical results are analysed in Section 5, whilst Section 6 offers conclusions.

## 2. Multi-component theory of mass transfer

The equation that governs multi-component mass transfer can be written [16,26] as:

$$\frac{\partial \mathbf{w}}{\partial t} = -Pe \mathbf{u} \cdot \nabla \mathbf{w} + \mathbf{\Delta} \nabla^2 \mathbf{w}. \quad (1)$$

The various terms in this equation can be defined as follows.

The term  $\mathbf{w}$  is a vector of solute mass fractions, so that with two solutes present  $w_1$  denotes solute 1 and  $w_2$  denotes solute 2. The terms  $\nabla$  and  $\nabla^2$  are the gradient and Laplacian operators (made dimensionless with respect to the drop radius  $R$ ). Meanwhile  $t$  denotes time. This has been made dimensionless on a diffusive time scale,  $R^2/(D)$ , where  $R$  is the drop radius, and  $(D)$  is a typical diffusivity scale, comprised of the average of the infinite dilution diffusivities of all the various components in the system.

The term  $Pe$  is the Peclet number, which is defined as  $RU_{drop}/(D)$ , where  $U_{drop}$  is the drop translation speed relative to the continuous phase. Physically  $Pe$  represents the ratio between the diffusive time scale and the streamline circulation time scale. A typical value of

$Pe$  is of the order of tens of thousands [26], i.e. diffusion is slow and circulation is rapid, but high  $Pe$  simulations are numerically stiff and hence expensive to solve. Moreover, for much of a drop’s evolution, any high  $Pe$  simulation results asymptote towards a ‘master curve’ [26]. Thus useful intuition can be gained with less numerical expense for  $Pe = 1000$  or even  $Pe = 100$ .

The field  $\mathbf{u}$  is the velocity field within and around the drop. It is made dimensionless on the scale  $U_{drop}$ . For simplicity (and by contrast with a number of other studies [30–33]) we will assume that the velocity field  $\mathbf{u}$  is steady over time, and that stresses on the drop surface associated with the velocity field are insufficient to deform it out of spherical. Subject to these assumptions, the field  $\mathbf{u}$  can be determined by (numerical) solution of the Navier–Stokes equations (see e.g. [28,29]) given the drop Reynolds number  $Re$ , and also the ratios between internal and external viscosity, and between internal and external density. It has also been shown that truncated Galerkin expansions [24–26,34,35] give acceptable approximations to the velocity fields. In what follows, we shall employ the same truncated Galerkin velocity field as in [26], which corresponds to  $Re = 30$  with equal internal and external viscosities, and equal internal and external densities. A sketch of the resulting streamline pattern is shown in Fig. 1. Notice that the field  $\mathbf{u}$  is axisymmetric, and hence the mass fraction field  $\mathbf{w}$  is likewise.

The term  $\mathbf{\Delta}$  is the multi-component diffusivity matrix [16] (made dimensionless on the scale  $(D)$ ). In the case where there are two solutes, it is a  $2 \times 2$  matrix, with elements  $\Delta_{11}$ ,  $\Delta_{12}$ ,  $\Delta_{21}$ ,  $\Delta_{22}$ . The off-diagonal elements  $\Delta_{12}$  and  $\Delta_{21}$  describe the multi-component couplings between the concentration gradient of one solute and the mass flux of another. The off-diagonal elements can be either positive or negative, depending on the direction in which a gradient of one solute drives a flux of the other. Moreover, if the off-diagonal elements vanish, the two solutes behave as independent single component systems.

The elements of the matrix  $\mathbf{\Delta}$  can be predicted via the so called Maxwell–Stefan theory [10,16,36–39]. These matrix elements are found to be functions of mass fraction. However it has been shown that during the course of a given liquid–liquid extraction operation, the variation of  $\mathbf{\Delta}$  is quite weak [40]. Thus  $\mathbf{\Delta}$  can be assumed to be a constant during the course of any given extraction operation. This has already been anticipated in Eq. (1) as the diffusivity  $\mathbf{\Delta}$  has been taken outside the Laplacian operator. More details regarding  $\mathbf{\Delta}$  will be given presently.

Eq. (1) needs to be solved with appropriate sets of initial conditions and boundary conditions. One has a choice of solving either [27] a coupled/‘conjugate’ problem (both inside and outside the drop, with mass fluxes and chemical potentials matched on the drop surface), or an external problem (i.e. outside the drop only) or an internal problem (i.e. inside the drop only). Technically it is the (more complicated) coupled problem which should be solved, whereas the external and internal problems are merely approximations. The external problem tends to be most relevant for extraction from a gas bubble [15,41] rather than the liquid drop case (on the grounds that mass transport tends to be extremely rapid in the gas phase, implying the transport in the external liquid phase is the rate-limiting step). It has been claimed however [26] that the internal problem may be a reasonable approximation to the full coupled liquid–liquid problem, especially in the limit of large  $Pe$ . The reason is as follows. Liquid inside the drop circulates around many times as mass transfer proceeds, whereas (in the frame of reference of the drop) liquid outside the drop flows past the drop surface once only. Fluid elements outside the drop therefore have less time to exchange mass amongst themselves (and so exchange mass over a smaller distance) than those inside the drop. If concentration differences tend to be realised over smaller distances outside the drop than inside it, this means the concentration differences must also be smaller outside to keep the fluxes matched. Hence, in the inter-

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