A NUMERICAL METHOD FOR MASS-TRANSFER IN TWO-FLUID FLOW WITH INTERFACES

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A new numerical algorithm for calculating advection and diffusion of a species within and between fluids with interfaces is described. The method, based on a Volume of Fluid (VOF) technique, employs a special treatment of computational cells which contain the interface to prevent errors in the predicted mass transfer due to smearing of the concentration profile. The algorithm is then used to predict mass transfer from a rising drop to the surrounding liquid for a particular experiment reported in the literature. A typical flow and concentration distribution calculated in the drop is shown, and the fractional change in the predicted solute concentration in the drop is found to be consistent with the experimental data.

INTRODUCTION

Mass transfer across deforming fluid interfaces such as drops, bubbles, or free surfaces is of generic interest and is important in a great many applications including most separation processes in which gases and liquids are involved, reactions with rising bubbles or falling drops in metal refining. In general, diffusion of material within each phase occurs as well as transfer across the interface. The diffusion coefficients in each phase usually differ, and the material concentration often exhibits a step change at the interface.

Since the interface is deforming, any numerical calculation of mass transfer across it must be performed simultaneously with a simulation of the fluid flow. However, there appears to be no published method which demonstrates accurate calculation of mass transfer across arbitrarily complicated deforming interfaces. Recent numerical studies include Ponoth and McLaughlin[1], who predicted the dissolution of steady-state (i.e. non-deforming) rising bubbles using a finite difference method with an adaptive mesh, and Chen et al.[2] who considered evaporation at the surface of gas bubbles rising in a liquid where the flow was simulated using a VOF method but was not validated for the evaporation case.

The numerical algorithm presented here for calculating advection and diffusion of a species within and between fluids with interfaces is based on novel enhancements to the VOF technique described in Rudman[3]. The problem is discretised on a uniform staggered mesh. Special treatment of computational cells which contain the interface is required to prevent errors in the predicted mass transfer of up to two orders of magnitude. Briefly, the transport of the bulk species concentrations in each phase involves VOF-based advection fluxes at interface cells and Flux-Corrected Transport[4] with centred fourth order differencing for the higher order fluxes away from the interface. The numerical discretisation of the diffusive fluxes is based on an analysis of one-dimensional diffusion across an interface.

Predictions are first compared with results of an analytical solution for a test problem involving diffusion from a sphere with uniform initial solute concentration during simple translation. This is followed by a calculation of the flow and concentration distribution for a rising drop in the circulating drop regime (non-oscillating) when mass transfer is controlled by the dispersed (drop) phase. The specific case considered corresponds to an experiment by Temos, Pratt & Stevens[5] for which data are presented in their Figure 6.

RESULTS

The prediction of diffusion with and without advection is first tested for the idealised case of a spherical drop, initially with uniform solute concentration, which is translated vertically with a prescribed uniform sinusoidal velocity field in a liquid in which the solute concentration can be regarded as zero (dispersed phase diffusion control). This problem has an analytical solution which is identical to that for diffusion from a stationary sphere[6]. It is essentially the Newman equation referred to in the solvent extraction literature[7]. Figure 1a shows that the fractional solute content in the sphere for diffusion without convection is almost identical to the analytical solution, and the corresponding prediction for combined advection and diffusion underpredicts the solute content for dimensionless times below 100 and overpredicts at times greater than 200. Figure 1b shows that the solute concentration changes proceed symmetrically within the sphere as required, except for some flattening on the left and right sides associated with the purely vertical velocity field.
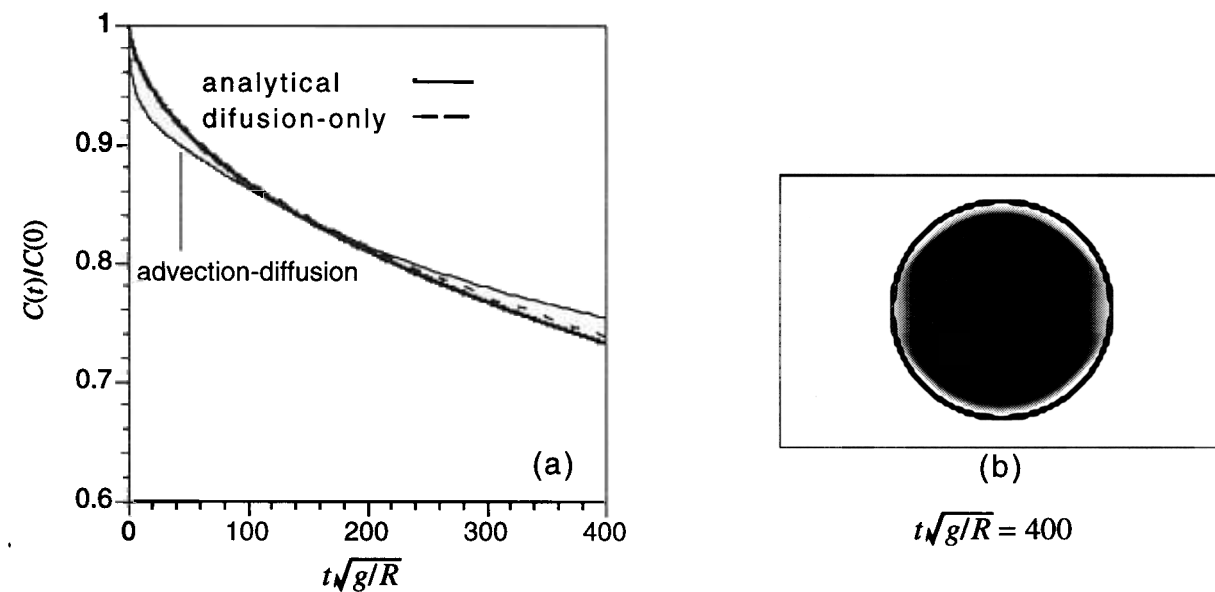

Figure 1: (a) Predicted solute content of a translating sphere of radius a vs the analytical solution for $Pe = R^{3/2} g^{1/2}/D_1 = 5 \times 10^4$, and (b) a snapshot of the evolving concentration distribution.

Figure 2 shows a typical solute concentration distribution and velocity vector field relative to the leading edge of a drop, predicted for an experiment of Temos, Pratt & Stevens[5] when mass transfer is controlled by the dispersed phase and the drop is non-oscillating. The region shown is only a small section of the computational domain around the drop. The Reynolds number equals 382, based on drop diameter and continuous phase properties. Figure 2 shows a toroidal recirculation within the drop as well as the region of highest solute concentration at the core of the recirculation cells. The central portion of the drop becomes depleted in solute as it is swept by the recirculation towards the drop surface where it can enter the continuous phase.

Because the initial solute concentration in the drop just after release into the liquid column is not known for the experiment of Temos, Pratt & Stevens[5], direct comparison of predicted and experimental drop concentration values is not possible. An attempt is made to compare the two by superimposing the plots shown in Figure 3. Because the initial concentration is unknown, the correct relative positioning of each plot is also unknown. The vertical shift imposed in Figure 3 is one which brings the predicted and experimental results into closest alignment. Although this cannot test the actual values predicted, it does provide a comparison of the predicted and experimental fractional concentration change. Figure 3 shows that the predicted fractional change is consistent with the concentration data for this experiment.
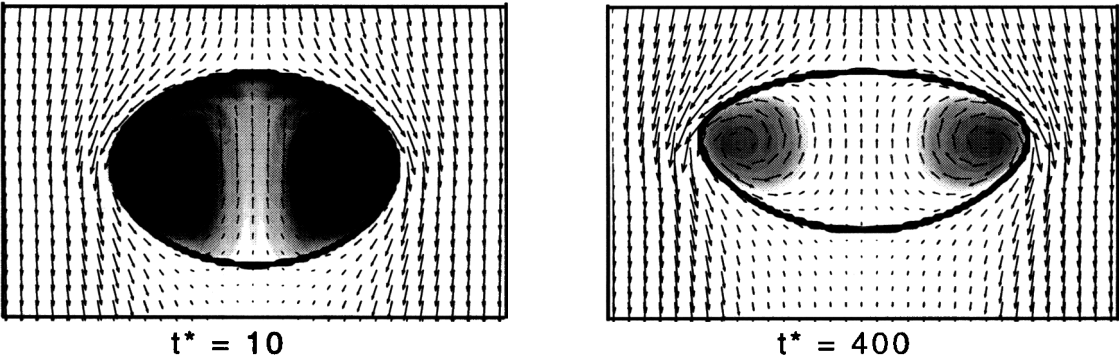

Figure 2: Predicted solute concentration distribution and velocity vector field relative to the leading edge of the drop (initial radius a) for an experiment[5] at two dimensionless times $t^* = t\sqrt{g/a}$ (dispersed phase control with $Re = 382$ based on drop diameter and continuous phase properties)
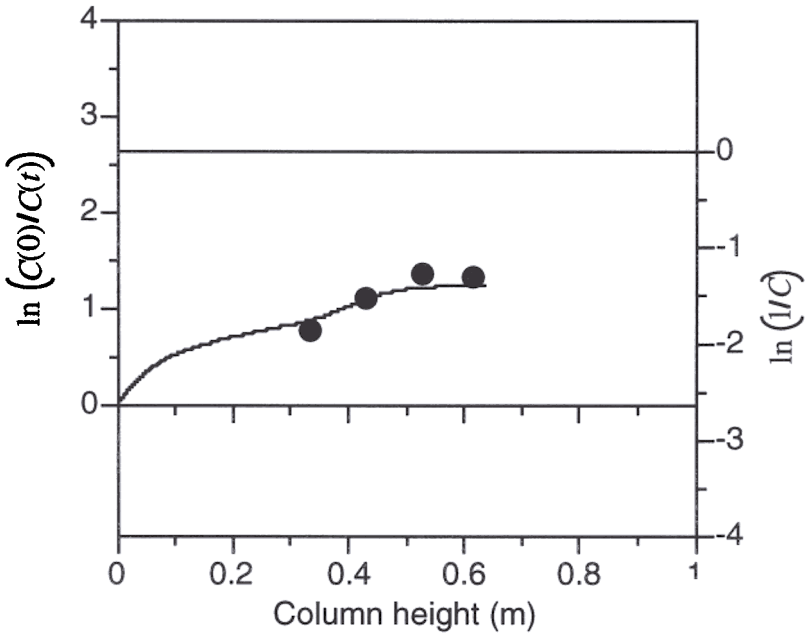

Figure 3: Superimposed plots based on predicted fractional solute content and experimental values[5] ● of solute concentration in the drop against height reached by the rising drop. The vertical axis in each case involves the inverse of the concentration.

CONCLUSION

There is a large body of literature concerning mass transfer between a rising drop and the surrounding liquid[7-8]. Typically, the drop deforms and an internal circulation is set up within it as it rises. In the absence of a full solution of the coupled flow and mass transfer, mathematical models have necessarily been approximate in their representation of the operating mechanisms. The Volume-of-Fluid numerical method used here has the potential to provide accurate predictions of the detailed flow and concentration distribution in and around the deforming drop, and does not require simplifying assumptions about the drop shape or the form of the internal circulation. The method employs a special treatment of computational cells which contain the interface to prevent advective smearing of the concentration profile there, and approximates the diffusive fluxes based on an analysis of one-dimensional diffusion across an interface.

In preliminary calculations, the method accurately predicts diffusion from a drop undergoing a uniform sinusoidal translation. Calculation of an experiment by Temos, Pratt & Stevens[5] for dispersed phase control of mass transfer predicted fractional changes in overall drop concentration which were consistent with the experimental data. The predicted concentration distribution and relative velocity field were presented, highlighting the recirculation pattern within the drop. However, many more tests against available data are required to fully evaluate the numerical model.

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