A NOVEL GENERALIZED MULTIFLUID MODELLING APPROACH FOR THE SIMULATION OF MULTIPHASE FLOWS: APPLICATION TO INTENSIFIED LIQUID-LIQUID EXTRACTION

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Abstract

Intensified liquid-liquid extraction is a separation process found in several applications in different industries including nuclear power generation, metallurgy and biotechnology. The multiphase hydrodynamic behaviour typical of intensified liquid-liquid extraction processes often exhibits a broad range of interfacial scales, ranging from small droplets embedded in a continuous phase to large interfaces observed in the segregated flow regions. This precludes the application of standard off-the-shelf multiphase modelling approaches, which rely on the assumption of either dispersed or segregated flow regimes in the entire computational domain, for the simulation of such flows. A novel GEeneralized Multifluid Modelling Approach (GEMMA) has been developed and validated to allow for the simulation of such multiscale multiphase flows. In this work this approach is applied to the simulation of the hydrodynamic behaviour for acetone absorption in a rotating disc column. It is shown that the modelling approach can accurately predict key hydrodynamic parameters such as organic phase hold-up and mean Sauter diameter. In addition, the GEMMA approach allows for an adequate representation of both the finely dispersed flow observed in the active part of the column and of the large segregated interface present in the top separator. It is concluded that the proposed approach represents a promising tool for the simulation of complex multiscale multiphase flows such as those observed in intensified liquid-liquid extraction processes.

1 Introduction

Liquid-liquid extraction is a separation process found in several applications in different industries including nuclear power generation, metallurgy and biotechnology (Hanson (2015)). Regardless of the specific application, high extraction efficiencies and low residence times are generally desirable properties. These can be attained in intensified extraction, which is enabled by devices such as extraction columns and annular centrifugal contactors (ACCs) (Hanson (2015); Vedantam and Joshi (2006)). On the other hand, there is a general lack of understanding of the local hydrodynamics observed during the operation of these devices. In this context, computational fluid dynamics (CFD) has the potential to provide valuable insights into the hydrodynamic behaviour of intensified liquid-liquid extraction processes. However, the complexity of the flow features observed in these processes poses significant challenges to standard CFD approaches (Vedantam et al (2012); De Santis et al (2021a)).

With respect to the modelling of multiphase flows, standard approaches can be generally divided in two main categories: interfacial-scale averaging and interfacial-scale resolving approaches (Prosperetti and Tryggvason (2007); Marschall (2011)). The former, which leads to so-called multifluid models, is particularly appropriate for simulating flows characterised by a fine dispersion of droplet or bubbles (representing the dispersed phase) embedded in a continuous phase. The interface is not resolved explicitly and therefore suitable closures are needed for the interfacial exchange of mass, momentum and energy. The interfacial-scale resolving approach includes both Lagrangian interface-tracking models and interface-capturing (e.g. volume of fluid (VoF), level set) models. In this case, the numerical grid has to be refined enough to allow for an adequate resolution of the interface morphology, which hinders the applicability of this approach to finely dispersed flows. However, both regimes are present at the same time in most “real-life” multiphase problems, and specifically in intensified liquid-liquid extraction processes; as such, standard off-the-shelf multiphase modelling approaches are ill-suited for the simulation of such flows. Therefore, there is a need for a generalised multiscale modelling approach capable of dealing with the coexistence of a broad range of interfacial scales within the same computational domain.

In response to this, we have recently developed a GEneralized Multifluid Modelling Approach (GEMMA) which allows for the simulation of multiscale multiphase flows by introducing interface resolution capabilities on top of a standard multifluid framework; it has been shown (De Santis et al (2021b)) that the proposed approach results in an accuracy comparable to interface-resolving methods in

the segregate/large-interface regime, whilst a standard multifluid behaviour is recovered in dispersed/small-interface flows. Further, application to the simulation of a plunging water jet demonstrated the capability of the approach to handle the presence of a broad range of interfacial scales within the computational domain. Subsequently, it has been demonstrated that the GEMMA approach coupled with a reduced population balance model can provide an accurate representation of the hydrodynamic features of the multiphase flow observed in ACCs (De Santis et al (2021a)).

This work presents the application of the GEMMA approach to predict the hydrodynamic behaviour of the multiphase flow observed in a Rotating Disc Column (RDC); the considered conditions within the column mimic the experimental investigation of Garthe (2006). The experimental data is also used to validate the numerical results.

The paper is organised as follows: an overview of the formulation of the model is provided in Section 2 and details of the numerical set-up used for the CFD simulation are given in Section 3; the numerical results are discussed in Section 4 and conclusions and future work are presented in Section 5.

2 Mathematical formulation

The GEMMA approach is built on top of a multifluid framework provided by the native *reactingMuliphaseEulerFoam* solver in OpenFOAM v7.0 (The OpenFOAM Foundation (2019)). *reactingMultiphaseEulerFoam* is a multifluid solver for systems of n compressible phases including interphase momentum, energy and mass transfer capabilities. The solver can also be coupled with an inhomogeneous population balance model (Krepper (2008)).

The GEMMA formulation is described in detail in (De Santis et al (2021b)), and only a short overview is provided here for the sake of brevity. The approach relies on the introduction of an ad-hoc multifluid formulation suitable for large interfacial scales. A binary scalar field C_α, depending on the local flow conditions and mesh size, is used to select the most appropriate local formulation for the model. In the cells where $C_\alpha = 1$ the local mesh size allows for an adequate resolution of the interfacial scales, and the aforementioned multifluid formulation for large interfacial scales is employed; in the other cells $C_\alpha = 0$, and a standard multifluid formulation suitable for small/dispersed interfacial scales provided by *reactingMultiphaseEulerFoam* is recovered.

The switch logic is based on the definition of an interface resolution quality (IRQ) index, which quantifies the capability of the mesh size to resolve the local interface curvature (De Santis et al (2021b); Anez et al (2016)); the IRQ is defined as:

$$IRQ = \frac{2}{\Delta\kappa} \tag{1}$$

where Δ is the local mesh size and κ is the local interface curvature evaluated as:

$$\kappa = -\nabla \cdot \left(\frac{\nabla\alpha}{|\nabla\alpha|}\right) \tag{2}$$

C_α is then set equal to one in the cells that satisfy the condition:

$$IRQ \geq IRQ_{crit} \tag{3}$$

where IRQ_{crit} is a user-defined threshold that allows control on the minimum resolution of the interface needed to activate the large-interface formulation. An additional control based on the local value of the volume fraction is included in the switch logic to ensure that C_α is zero in the cells containing pure phases (De Santis et al (2021b)). Also, for the cases in which the local dispersed phase diameter d is known (for instance, from the solution of a population balance equation), an additional check is performed and the switch is activated only in the cells where the condition:

$$d > \Gamma\Delta \tag{4}$$

is met; in Equation (4), Γ is another user-defined parameter which allow control on the minimum resolution of the dispersed phase diameter required to activate the large-interface resolution.

The large-scale multifluid formulation employed in the cells where C_α is equal to one introduces three main features:

- The introduction of a compression term to guarantee a sharp representation of the interface in the large-interface cells,
- The inclusion of suitable closures for interfacial transfer model which are compatible with the local morphology of the interface; in particular, the drag model of Marschall (2011) is used to account for interfacial friction in cells where $C_\alpha = 1$,
- The introduction of a multifluid-compatible formulation for the surface tension force.

In addition, a reduced population balance formulation is used to evaluate the local Sauter mean diameter of the organic phase in the present simulation. Similarly to De Santis et al (2021a), the One-Primary-One-Secondary-Particle-Method (Drumm et al (2010)) is used for this purpose, with the break-up and coalescence rates evaluated using the closures of Martinez-Bazan et al (1999) and of Prince and Blanch (1990), respectively. It should be pointed out that the choice of a reduced population balance approach is linked to the need to reduce the computational cost of the simulation, but the GEMMA approach is fully compatible with more complex population balance formulations.

3 Numerical set-up

The computational domain mimics the experimental set-up of Garthe (2006), for which a schematic representation is provided in Figure 1. The RDC has a total height of 4400 mm and an active height of 2950 mm, corresponding to 59 compartments; the compartments are delimited by static baffles (not depicted in Figure 1) attached to the static external wall of the active section of the column with a 50 mm spacing. The

shaft driving the rotating discs has a rotating speed of 200 RPM.

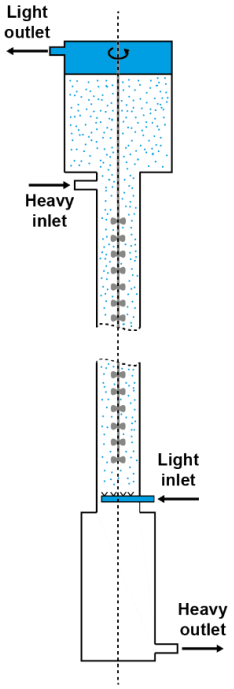

Figure 1: Schematic of the RDC.

In the experiments the column is used for acetone absorption in a water/toluene/acetone system. Since the objective of the numerical simulation is to evaluate the hydrodynamic behaviour of the system, and the acetone concentration was kept relatively small in the experiments, mass transfer is not accounted for in the simulation. The physical properties for the aqueous and the organic phases are taken from Garthe (2006).

The computational domain makes use of the axial symmetry of the rig (with the exclusion of the inlet and outlet pipes), and consists of a 5-degree wedge with rotational periodicity imposed on the front and back boundaries of the domain. This numerical set-up has the advantage of reducing the computational cost with respect to the simulation of the full geometry of the column; at the same time, the computational domain could be readily modified for the use of a larger angle for the wedge if deemed necessary in future work, without the need to modify the numerical set-up.

The numerical grid consists of unstructured hexahedral cells with a characteristic size of 2 and 0.8 mm in the bulk and at the walls, respectively, for a total cell count approximately equal to 120,000.

Dirichlet conditions enforcing flow rates consistent with the experimental set-up (i.e. volumetric flow rates of 40 and 48 l/h for the aqueous and the organic phase, respectively) are used for the velocity at the heavy phase inlet and outlet, and at the light phase inlet, whilst a Neumann condition is used for the light phase outlet; a no-slip condition is used for the velocity at the walls and moving wall conditions are used to account for the rotation of the shaft and of the rotating discs. For the operating pressure, Neumann conditions are used on all the boundaries but the light outlet, where a Dirichlet condition on the total pressure is enforced. For the reduced population balance method, a Dirichlet condition corresponding to a mean Sauter diameter of 3 mm is imposed at the light phase inlet for the number density, whilst a zero-gradient condition is used on the other boundaries.

The simulation has been performed using the open-source code OpenFOAM v7.0; the convective terms in all the transport equations have been discretised using a second-order total variation diminishing (TVD) scheme, whilst the time-derivative terms have been discretised using a first-order Eulerian scheme; the MULtidimensional limiter for Explicit Solution (MULES) is used to guarantee the boundedness of the volume fraction solution (Berberovic et al (2009)).

The time-step size is selected to guarantee a maximum Courant number value less than or equal to 0.5. The simulation has been run for 200 s of physical time to reach a statistically steady state plus an additional 400 seconds to collect statistics.

Both phases are considered to be turbulent and a large eddy simulation (LES) has been used to account for turbulence effects, with a dynamic Smagorisnky model (Smagorinsky (1963); Verma and Mahesh (2012)) for the subgrid stresses and a wall-function approach (Launder and Spalding (1974)) used to relax near-wall mesh requirements.

4 Results and discussion

Figure 2 shows the time-averaged organic phase volume fraction in the central compartments (i.e. compartments #29 and #30) of the RDC.

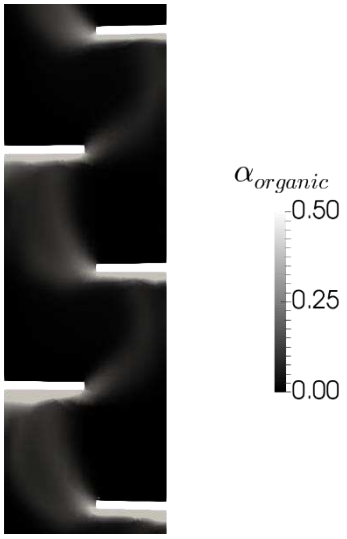

Figure 2: Time-averaged organic phase volume fraction in the central compartments.

Consistently with what has been observed in other CFD investigations of RDCs (Drumm et al, (2010); Attarakih et al (2015)), it can be seen that the organic phase tends to accumulate beneath the discs and the

stator baffles, forming pockets in those regions. However, this accumulation is due to a large number of droplets, and therefore there is not a defined large-scale interface between these organic phase pockets and the surrounding aqueous phase. At the same time, it is observed that the organic phase also establishes a S-shaped path (although with a low volume fraction value compared to the pockets under the baffles) between the discs and the static baffles.

The time averaged velocity magnitude contours for the two phases in the central compartments, together with the corresponding velocity vectors, are depicted in Figure 3. The upward S-shaped motion of the organic phase highlighted in Figure 2 is evident from the velocity vectors; the heavy phase velocity field, on the other hand, is dominated by the presence of two large counter-rotating vortices in each compartment, and no evident downward motion is visible.

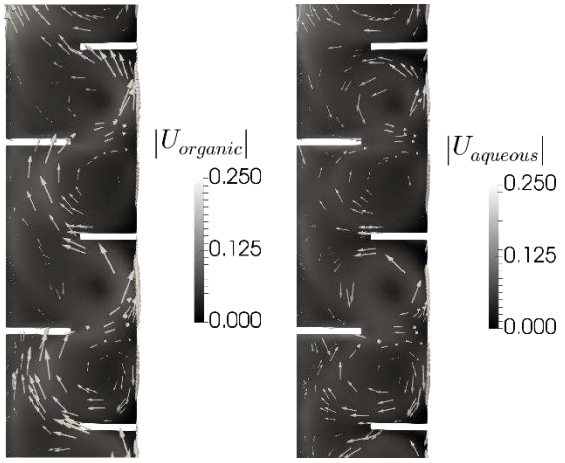

Figure 3: Time-averaged velocity magnitude for the organic (left) and the aqueous (right) phases with superimposed corresponding velocity vectors in the central compartments.

The benefit of using a generalized approach such as GEMMA in the simulation of this case is highlighted in Figure 4, showing the resolution of the large interface found in the top separator of the column. It can be seen that the approach is capable of detecting the large interfacial scales associated with this feature, which results in the activation of the C_α switch at the interface. This, in turn, allows for a "sharp" representation of the interface, without any significant numerical diffusion that is typical of the standard "dispersed" multifluid formulation when resolving large segregated interfaces.

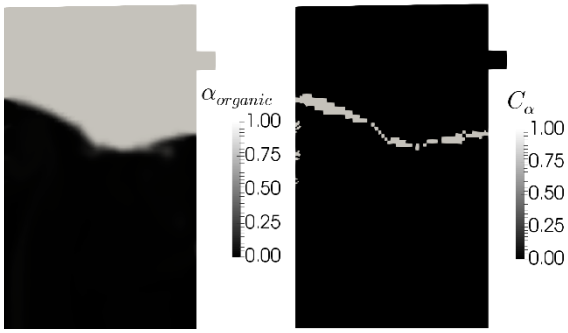

Figure 4: Instantaneous organic phase volume fraction (left) and $\boldsymbol{C_\alpha}$ switch (right) contours at the top separator of the column.

Finally, the compartment-averaged light phase hold-up and Sauter mean diameter obtained from the simulation are compared with the experimental measurements of Garthe (2006) in Figure 5 and Figure **6**, respectively. The simulated organic phase hold-up in the active section of the column is roughly constant, with an average value of 0.067 which is in a good agreement with the experimental observations. For the Sauter mean diameter, the numerical results tend to overpredict its value slightly along the column compared to the experimental measurements. Experimental error ranges were not reported, but the average variance with respect to the experiments data points appears to be relatively small. In this respect, it should be pointed out that no efforts were mode to optimize the break-up and coalescence kernels used in the reduced population balance to improve the agreement with the experimental data.

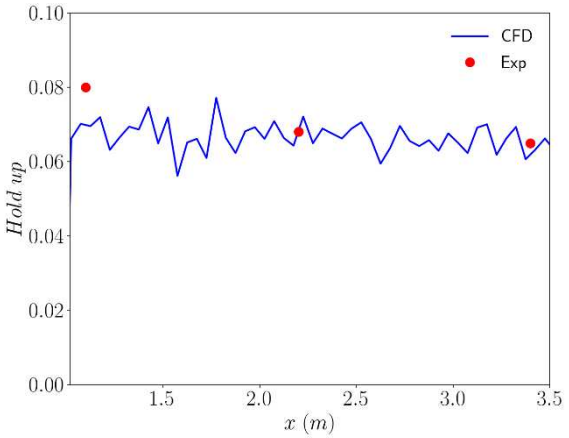

Figure 5: Compartment-averaged organic phase hold-up along the active section of the column axis: numerical results and experimental measurements.

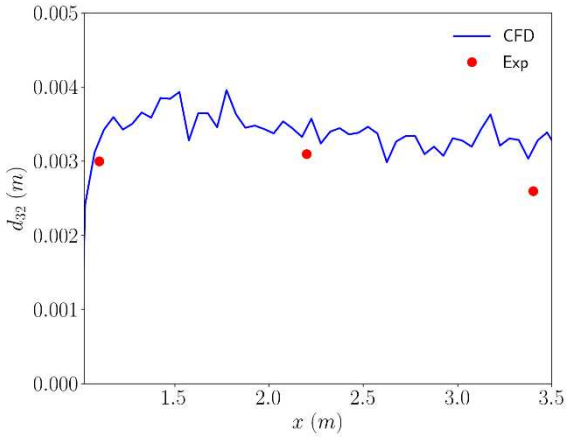

Figure 6: Compartment-averaged organic phase Sauter mean diameter along the active section of the column axis: numerical results and experimental measurements.

Both hold-up and Sauter mean diameter are key hydrodynamic parameters for the prediction of the mass transfer performance of the device; therefore, the accurate results obtained with GEMMA are seen as an encouraging first step for the prediction of mass transfer within the column.

5 Conclusions

A novel GEneralized Multifluid Modelling Approach (GEMMA) suitable for the prediction of multiscale multiphase flow has been applied to the simulation of the hydrodynamic behaviour of a rotating disc column used for acetone absorption. A comparison with experimental measurements has shown that the proposed approach is capable of providing an accurate representation of the complex multiscale flow found within the device; this is regarded as key for the prediction of the mass transfer performance of the column. Further, it has been shown that the GEMMA approach can adapt to the local interfacial scales, with the large segregated interface observed in the top separator of the column being correctly identified and resolved using the large-interface formulation of the model. It is concluded that the proposed approach represents a promising tool for the simulation of complex multiscale multiphase flows such as those encountered in liquid-liquid extraction; future activities will include further model assessment and the inclusion of local predictive capabilities for concentration-driven mass transfer within the GEMMA framework.

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