AN ACCURATE NUMERICAL METHOD FOR COMPUTATION OF TWO-PHASE FLOWS WITH SURFACTANTS

Sashikumaar Ganesan¹, Andreas Hahn², Kristin Held², and Lutz Tobiska²

¹Numerical Mathematics and Scientific Computing, SERC, Indian Institute of Science Bangalore 560012, India e-mail: sashi@serc.iisc.in

 ² Institut für Analysis und Numerik, Otto-von-Guericke-Universität Magdeburg Postfach 4120, D-39016 Magdeburg, Germany.
e-mail: {andreas.hahn, kristin.held, tobiska}@ovgu.de

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Abstract. Numerical computations of two-phase flows with surface active agents (surfactants) are highly demanded in several scientific and engineering applications. Apart from the other challenges associated with the computation of two-phase flows, the presence of surfactants increases the complexity. Surfactants alter the flow dynamics significantly by lowering the surface tension on the interface. Moreover, the concentration of surfactants along the interface is often not uniform and thus Marangoni forces are induced. Adsorption and desorption of surfactants between the interface and the bulk phase may take place in the case of soluble surfactants.

1 INTRODUCTION

The presence of surface active agents (surfactants) at fluid interfaces can influence the flow dynamics considerably. Surfactants are amphiphilic organic compounds, which can be adsorbed at liquid/gas or liquid/liquid interfaces. They typically alter the interface dynamics by a reduction of surface tension. Furthermore, a nonuniform distribution produces gradients in the surface tension leading to tangential forces along the interface (Marangoni effect).

The numerical techniques to handle such complex problems can be roughly classified into interface tracking and interface capturing methods. Interface tracking methods, e.g. [10], use a separate grid for the interface or a set of interconnected points to mark the interface. An implicit description of the interface by a level set, color or phase-field function is used in interface capturing methods, see e.g. [7, 8]. The interface has to be reconstructed but the solution of the problem in the bulk phases can be done independently on the underlying grid for the interface and topological changes are simple to handle. In the diffuse-interface or phase-field function rapidly transists from zero on one side of the domain to one on the other side. The partial differential equations in the bulk and on the interface are extended on a larger regular domain with additional terms approximating the physical boundary condition at the interface. For details we refer to [11].

In this paper, we present an accurate sharp interface numerical method based on a coupled arbitrary Lagrangian-Eulerian and Lagrangian approach. It can be considered as an interface tracking approach. The mesh representing the interface is just a restriction of the mesh in the bulk phases in each time step. This resolution of the interface allows an accurate and natural incorporation of surface forces, Marangoni forces and jumps in density and viscosity. Spurious velocities are successfully suppressed by using isoparametric finite elements, discontinuous (at least phase-wise) pressure approximations and the Laplace-Beltrami technique for representing the curvature. The discrete representation of the interface is directly used to discretize the surface evolution-equation for the surfactant concentration on the interface. In particular, we discuss ways to extend the 3d-axisymmetric code based on the techniques and algorithms developed in [5, 6] to the fully 3d case.

2 GOVERNING EQUATIONS AND DISCRETIZATION

Our mathematical model for two-phase flows with soluble surfactants consists of the timedependent incompressible Navier-Stokes equations in both phases,

$$\rho_k \left(\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right) - \nabla \cdot \mathbb{S}_k(\mathbf{u}, p) = \rho_k \mathbf{e}, \qquad \nabla \cdot \mathbf{u} = 0 \quad \text{in} \quad \Omega_k(t) \subset \mathbb{R}^3, \quad k = 1, 2,$$

an evolution-equation for the surfactant concentration in the outer phase

$$\frac{\partial C}{\partial t} + (\mathbf{u} \cdot \nabla)C = D_c \Delta C \quad \text{in} \quad \Omega_1(t) \subset \mathbb{R}^3$$

and an evolution-equation for the surfactant concentration on the interface between them

$$\frac{\partial c_{\Gamma}}{\partial t} + \mathbf{U} \cdot \underline{\nabla} c_{\Gamma} + c_{\Gamma} \underline{\nabla} \cdot \mathbf{u} = D_s \underline{\Delta} c_{\Gamma} + S(c_{\Gamma}, C) \quad \text{on} \quad \Gamma_F(t).$$

We assume that $\Gamma_F(t) = \partial \Omega_2(t) = \partial \Omega_2(t) \cap \partial \Omega_1(t)$, i.e. the inner phase $\Omega_2(t)$ has no contact to the remaining part of the boundary of the outer phase. We impose the kinematic, force balancing

and flux boundary conditions

$$\begin{aligned} [\mathbf{u}] &= 0, \quad \nu \cdot [\mathbb{S}(\mathbf{u}, p)] \cdot \nu + \sigma(c_{\Gamma}) \mathcal{K} = 0 & \text{on } \Gamma_F(t), \\ \mathbf{u} \cdot \nu &= w, \quad \tau_i \cdot [\mathbb{S}(\mathbf{u}, p)] \cdot \nu - \tau_i \cdot \nabla \sigma(c_{\Gamma}) = 0 & \text{on } \Gamma_F(t), \\ -(\nu \cdot D_c \nabla C) &= S(c_{\Gamma}, C) & \text{on } \Gamma_F(t), \end{aligned}$$

on the interface $\Gamma_F(t)$, and appropriate boundary conditions on the remaining part of the boundary. The coupling term $S(c_{\Gamma}, C)$ is often modeled by

$$S(c_{\Gamma}, C) = k_a C \left(1 - \frac{c_{\Gamma}}{c_{\Gamma, \infty}} \right) - k_d \frac{c_{\Gamma}}{c_{\Gamma, \infty}}.$$

In case of insoluble surfactants, we neglect the surfactant concentration equation in the bulk phase and set the source term $S(c_{\Gamma}, C)$ equal to zero. In the above equations the following notations have been used: $\mathbb{S}_k(\mathbf{u}, p) = \mu_k \mathbb{D}(\mathbf{u}) - p\mathbb{I}$, $\mathbf{e} = (0, 0, -g)$, \mathbf{u} - velocity, p - pressure, t - time, ρ_k - density, μ_k - dynamic viscosity, σ - surface tension, g - gravity, C - surfactant in outer phase, D_c - diffusion coefficient of C, c_{Γ} - surfactant on interface, D_s - diffusion coefficient of c_{Γ} , ν - normal and τ_i - tangential unit vectors of the interface, \mathcal{K} - sum of principal curvatures, $\mathbf{U} = \mathbf{u} - (\mathbf{u} \cdot \nu)\nu$ - tangential velocity at the surface, w - velocity of the interface, k_a - adsorption coefficient, k_d - desorption coefficient, $c_{\Gamma,\infty}$ - maximum surface packing surfactant concentration, and $\overline{\Sigma}$ - surface gradient.

The time-dependent Navier-Stokes equations, the bulk and the surface transport equations are solved simultaneously using the coupled ALE-Lagrangian method in 3d-axisymmetric configuration [5]. Since the interface is resolved in the ALE approach, surface forces, Marangoni forces and jumps in the material parameters are incorporated accurately into the model. Further, the Laplace-Beltrami technique for representing the curvature is used [3]. An advantage of using the ALE approach is that a separate lower dimensional interface mesh is not needed for approximating the surface evolution-equation. Instead, we use the discrete representation of the interface directly.

The correct choice of finite elements for the spatial discretization is crucial in the simulation of two-phase flows, since one may expect discontinuous pressures due to the Young-Laplace law. For the axisymmetric case, the inf-sup stable two-dimensional P_2^+/P_1^{disc} - element is succesfully used. It consists of a discontinuous, piecewise linear pressure space and a continuous, piecewise quadratic velocity space enriched with one cubic bubble function per cell. Unfortunately, the three-dimensional variant of the P_2^+/P_1^{disc} - element requires five bubble functions per cell to be inf-sup stable, which entails a vigorous increase of degrees of freedom. Table 1 and 2 show the increase of degrees of freedom with refinement level of a simplicial mesh in comparision to the standard Taylor-Hood element (P_2/P_1).

Table 1: Degrees of freedom in 2d				Table 2: Degrees of freedom in 3d		
Level	P_2/P_1	P_2^+/P_1^{disc}	-	Level	P_{2}/P_{1}	P_2^+/P_1^{disc}
1	347	666	-	1	4 673	16055
2	1 3 3 9	2 6 2 6		2	32 821	122 365
3	5 267	10434		3	245 254	955 415
4	20 899	41 602		4	1 894 568	7 550 827

Therefore, we suggest an extended Taylor-Hood element, which uses a continuous, piecewise linear pressure space within each fluid phase but allows pressure jumps across the interface. The

extended Taylor-Hood element is inf-sup stable and shows excellent mass conservation similar to the P_2^+/P_1^{disc} - element. In order to reduce the number of needed remeshing steps we applied an elastic mesh update as in [5, 6].

3 NUMERICAL EXPERIMENTS

We performed several numerical tests to validate the numerical scheme and to demonstrate the influence of surfactants on the flow dynamics. Here, we concentrate on fully 3d computations and refer to [5, 6] for the axisymmetric case with insoluble and soluble surfactants.

3.1 Clean oscillating droplet

The first test example is used to validate the Navier-Stokes solver by computing a one-phase flow with a capillary surface for different Reynolds numbers. At time t = 0 the droplet is in rest $\mathbf{u}(x,0) = \mathbf{0}$ and has an ellipsoidal shape with the equatorial radii $r_1 = 1$, $r_2 = 1$, and $r_3 = 1.2$, where r_i corresponds to the x_i -axis, i = 1, 2, 3. Close to the tip at $x = (0, 0, r_3)$ the surface tension force is larger and the droplet starts to compensate the imbalance of forces. Due to the viscosity of the fluid for $t \to \infty$ the spherical equilibrium shape of the fixed volume droplet is reached with radius $R_{\infty} = 1.06266$. As proposed in [2], we compute the damping factor δ_n by

$$\delta_n := \sqrt[n]{\frac{r_{max}(t_n) - R_{\infty}}{r_{max}(t_0) - R_{\infty}}},$$

where $r_{max}(t_n)$ denotes the maximal elongation of the droplet in x_3 -direction at time t_n for the *n*-th period. In the following diagrams, we used the relative tip position $r_{rel} = r_{max} - R_{\infty}$ instead of the real tip position $(0, 0, x_3)$. The discretization in space leads to an algebraic system with 19247 degree of freedom; the time step size has been set to 0.01.

In Fig. 1 the influence of the Reynolds number $\text{Re} = \rho U_{\infty} L/\mu$ on the damping property is shown for the Weber number $\text{We} = \rho U_{\infty}^2 L/\sigma = 13.5$. For small Reynolds numbers we observe



Figure 1: Relative tip position for We = 13.5 and different Reynolds numbers over time (left). Damping factor for the second period and Re = 1000 (right).

a fully damped transition of the droplet shape into the equilibrium. As expected, for increasing Reynolds numbers the periodic behaviour becomes more pronounced and the damping factor is approaching one. Fig. 2 gives an impression on the velocity field at time t = 0.15 and t = 3.28,



Figure 2: Velocity field of the clean oscillating droplet for Re = 1000 and We = 13.5 at time t = 0.15 (left) and t = 3.28 (right).

respectively. We tested the long time behaviour of different time-discretization schemes and found no visible differences as shown in Fig. 3. Nevertheless, the conservation of volume is almost one order of magnitude better for the fractional-step- θ -scheme due to its higher computational effort. In Table 3 the relative error

$$\Delta V(t) := \left(\left| \left| \Omega_h(0) \right| - \left| \Omega_h(t) \right| \right| \right) / \left| \Omega_h(0) \right|$$

in the conservation of the discrete volume $|\Omega_h(t)|$ for the different time discretizations at the final time t = T = 100 are given. We see that the error in all cases is below 0.2% which

time discretization	rel. error $\Delta V(T)$
backward Euler	1.5811-3
Crank-Nicolson	1.5801-3
frac-step- Θ	5.4226-4

Table 3: Relative error in conservation of volume.

indicates a high accuracy of the scheme.

3.2 Bulk concentration diffusion test

In this test case we verify the diffusion of the bulk concentration and the mass transfer from the bulk phase to the interface. A similar test has been performed for the front-tracking method in [10] and in the 3d-axisymmetric case in [6]. We consider an initially clean sphere with radius $r_0 = 1$ suspended in a large cylinder. We assume an initial bulk concentration $C_0 = 1$ and that the mass transfer is solely due to the molecular diffusion. Moreover, a simplified version of the source term $S(c_{\Gamma}, C) = k_a C$ is considered, so that the mass transfer takes place only from the bulk phase to the interface. For a short period of time or in an infinite domain, an analytical approximation of the bulk concentration C is given by

$$C = C_0 \left(1 - \frac{k_a \sqrt{\pi D_c t} / D_c}{1 + \sqrt{\pi D_c t} \left(1 + k_a r_0 / D_c \right) / r_0} \frac{r_0}{r} \operatorname{erfc}\left(\frac{r - r_0}{2\sqrt{D_c t}}\right) \right)$$

where $\operatorname{erfc}(x)$ is the complementary error function. The droplet is placed at the center of the cylindrical tube that extends 5 and 8 droplet radii in the radial (*r*-) and axial (*z*-) directions,



Figure 3: Long time behaviour of the clean oscillating droplet for Re = 1000 and We = 13.5 with different time discretizations; backward Euler (be), Crank-Nicolson (cr) and fractional-step- Θ -scheme (fs).



Figure 4: Bulk concentration diffusion test. Isosurfaces at t = 5, 8, 10.

respectively. In our computation, an unstructured tetrahedral grid has been used with 21209 degrees of freedom. Furthermore, we used $k_a = 1$, $D_c = 0.1$ and the Crank-Nicolson method with a time step of 0.1 for the time discretization. For illustrating the transport of the bulk concentration to the interface, contour plots of the bulk surfactant concentration in the (r, z)-cutting plane at different times are shown in Fig. 4. As qualitatively expected, the contours of C are circles around the droplet with centre equal to the droplet centre. For a quantitative comparison with the analytical approximation above, the bulk concentration profiles are evaluated in radial direction through the center of the droplet and shown in Fig. 5 (left) at different times. The concentration in the bulk phase at the interface which appears in the surface evolution equation as a source term is validated by comparing it with the analytical approximation above over a period of time in Fig. 5 (right). The computational results are in good agreement with the analytical approximation. Nevertheless, the steep fronts of the concentration near the droplet occuring for small t cause an error which is accumulated over time, see Fig. 5 (right).



Figure 5: Bulk concentration along radial direction at times t = 0.1, t = 1, t = 4, and t = 10 (left) and at the droplet over time (right). Comparison of numerical and analytical approximation.

3.3 Surface evolution-equation

The next test example has been used to validate the discretization of the surface evolutionequation. For this we consider the unit sphere $\Gamma = S^2$ and solve the diffusion problem

$$\frac{\partial c_{\Gamma}}{\partial t} = \underline{\Delta} c_{\Gamma} \quad \text{on } \Gamma, \qquad c_{\Gamma}(x,0) = x_1 x_2.$$

We consider Γ as a fixed time-independent surface. There is an analytical solution given by

$$c_{\Gamma}(x,t) = x_1 x_2 \exp(-6t).$$

On a sequence of meshes we used three different discretizations in space, the affine P_1 finite element, the affine P_2 finite element, and the isoparametric P_2 finite element. For the timediscretization the backward Euler scheme has been used with a time step of 0.001 and end time T = 2. In order to compare the analytical solution $c_{\Gamma} : \Gamma \to \mathbb{R}$ living on the smooth surface Γ with the numerical solution $c_{\Gamma,h} : \Gamma_h \to \mathbb{R}$ living on the approximated surface Γ_h we lift it on the discrete surface by the projection in radial direction, i.e.

$$c^{l}(x,t) = c\left(\frac{x}{\|x\|},t\right), \quad x \in \Gamma_{h}.$$

Then, the error $e = c^l - c_{\Gamma,h}$ in the usual $L^2(H^1)$ - and $L^2(L^2)$ -norms are computed by means of a suitable quadrature rule. The results are summarized in Tables 4-6. Different from the

h	L2(H1)	order	L2(L2)	order
0.5032	4.245		1.0010	
0.2562	2.069	1.002	0.2690	1.894
0.1287	1.024	1.014	0.0068	1.984
0.0644	0.511	1.036	0.0017	1.983

Table 4: Approximation with affine P_1 finite elements.

case of a flat surface the use of affine P_2 elements instead of affine P_1 elements does not lead to improved error estimates. However, isoparametric P_2 elements show the optimal order of

h	L2(H1)	order	L2(L2)	order
0.5032	2.1357		0.2024	
0.2562	1.1043	0.9516	0.0496	2.029
0.1287	0.5571	0.9871	0.0123	2.009
0.0644	0.2792	0.9966	0.0031	2.005

Table 5: Approximation with affine P_2 finite elements.

h	L2(H1)	order	L2(L2)	order
0.5032	3.328-1		2.067-2	
0.2562	8.571-2	1.9575	2.390-3	3.1126
0.1287	2.169-2	1.9820	2.929-4	3.0287
0.0644	5.453-3	1.9921	3.668-5	2.9975

Table 6: Approximation with isoparametric P_2 finite elements.

convergence also in the non-flat case of a sphere.

A further test example describes the distribution of surfactants on a static unit sphere with convection. The tangential velocity field is given by

$$\mathbf{u}(x,t) = 4\pi \left(x_2, -x_1, 0 \right)$$

and corresponds to a rotation of the sphere around the x_3 -axis with two rotations per unit time. Note that $\nabla \cdot \mathbf{u} = 0$. We solve the diffusion-convection problem

$$\frac{\partial c_{\Gamma}}{\partial t} + \mathbf{u} \cdot \underline{\nabla} c_{\Gamma} = \frac{1}{\text{Pe}} \underline{\Delta} c_{\Gamma} \quad \text{on } \Gamma, \qquad c_{\Gamma}(x,0) = x_1 x_2 + 0.5$$

where $Pe = U_{\infty}L/D_s$ denotes the Péclet number. The surface Γ and the surface equation are discretized in space by isoparametric P_2 elements on grids with mesh sizes $h_1 = 0.3249$, $h_2 = 0.1646$, and $h_3 = 0.0826$. The backward Euler scheme has been used for the time discretization. We consider a case in which convection dominates diffusion and set Pe = 10000. In Fig. 6 we present the surfactant concentration on the surface at time t = 0 (left) and after two rotations at time t = 1 (right) for the finest mesh level $h = h_3$. Due to the low diffusion the two snapshots are almost identical as expected. We compute the relative error in the mass conservation

$$\Delta m(t) = |m(0) - m(t)| / m(0), \qquad m(t) = \int_{S^2} c(x, t) \, d\gamma, \qquad m(0) = 2\pi$$

and obtained after two rotations on the finest mesh level $\Delta m(1) \approx 10^{-8}$. Even on the coarser mesh $h = h_1$ we still have $\Delta m(1) < 10^{-5}$ which demonstrates the high accuracy of the scheme.

3.4 Contaminated oscillating droplet

As the final test example we consider again an oscillating droplet but now contaminated with surfactants. The Navier-Stokes equations coupled with the evolution-equation for the surfactant



Figure 6: Surfactant concentration on the surface at time t = 0 (left) and after two rotations at time t = 1 (right) for a convection dominated test case (Pe = 10000).

concentration on the interface has to be solved. We use the linear Henry's equation of state leading to the boundary condition in non-dimensionless form

$$\left(\frac{2}{\operatorname{Re}}\mathbb{D}(\mathbf{u}) - p\mathbb{I}\right) \cdot \nu = \frac{1}{\operatorname{We}} \left(1 + \operatorname{E}(0.5 - c_{\Gamma})\right) \mathcal{K}\nu - \frac{\operatorname{E}}{\operatorname{We}} \underline{\nabla}c_{\Gamma} \qquad \text{on } \Gamma_{F}(t)$$

where E denotes the surfactant elasticity [6, 10]. The initial configuration is as in the first example, i.e. at t = 0 the droplet is in rest, has an ellipsoidal shape with the equatorial radii $r_1 = r_2 = 1$ and $r_3 = 1.2$. Additionally a uniform surfactant concentration of $c_{\Gamma} = 0.5$ at t = 0is assumed. Note that our setting guarantees that a uniform surfactant concentration of $c_{\Gamma} = 0.5$ corresponds to the case of a clean surface with the same Weber and Reynolds number. In Fig.7



Figure 7: Relative tip position for the clean and contaminated oscillating droplet for We = 13.5, Re = 1000, Pe = 1, and E = 10. Higher damping rate for the contaminated droplet due to additional Marangoni forces induced by the non-uniform surfactant concentration.

the relative tip position is shown for the clean and contaminated droplet. The moving surface

leads to a non-uniform surfactant concentration and Marangoni forces arise which counteract the change of surfactant concentration. This results, as we can see in Fig. 7, into higher damping rates and lower frequences compared to the clean droplet. In Fig. 8 (left) the damping factor



Figure 8: Damping factor versus surface elasticity for Pe = 1 (left) and versus Péclet number for E = 10 (left). Computations for Re = 1000 and We = 13.5.

for different surface elasticities and Re = 1000, Pe = 1, and We = 13.5 is presented. Note that the damping factor for the contaminated droplet is always smaller compared with the damping factor $\delta_2 = 0.9441$ for the clean droplet. This agrees with the higher damping rates of the contaminated droplet in Fig. 7. Nevertheless, for increasing surface elasticity we observe less damping which on the first glance is surprising. However, the surface elasticity influences both the normal and tangential forces and their interplay determines the behaviour of the droplet. If the diffusion becomes larger (smaller Pe-numbers) we expect an almost uniform surfactant concentration such that $\underline{\nabla}c_{\Gamma} \approx 0$ and the influence of Marangoni forces is less pronounced. This effect can be clearly seen in Fig. 8 (right). Smaller Pe-numbers cause less damping.



Figure 9: Surfactant concentration on the surface of the oscillating droplet over a fixed scale at times t = 0 (left), t = 2.3 (middle), and t = 4.5 (right). The concentration is nearly uniform and lower on larger surfaces (left and right).

However, Fig. 8 (right) shows that the interplay of the normal and Marangoni forces is rather complex. Instead of decreasing monotonically - as one could expect - the damping factor even develops a minimum. Finally, we show in Fig. 9 the surfactant concentration on the surface of the oscillating droplet with a fixed scale at different times. The initial state Fig. 9 (left) has a uniform surfactant concentration and we observe that the surfactant concentration increases by shrinking the surface area (middle) and decreases by streching it (right). However, a closer look with a different scale in Fig. 10 shows that the surfactant concentration is indeed non-uniform such that the above mentioned Marangoni forces really appear.



Figure 10: Surfactant concentration on the surface of the oscillating droplet on an adapted scale at times t = 2.3 (left) and t = 4.5 (right). Note that the minimum of concentration on the smaller surface (left) is larger than the maximum on the larger surface (right).

4 CONCLUSIONS

An accurate finite-element method has been proposed for flows with surfactants. It is based on a coupled arbitrary Lagrangian-Eulerian and Lagrangian approach in which the interface is resolved by the mesh. The numerical scheme has been validated by a set of simple 3d test examples completing those of [5, 6] for the 3d-axisymmetric case.

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