NUMERICAL MODELLING OF ABSORPTION OF GASEOUS INCLUSIONS RISING IN LIQUID

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In the current work we deal with the numerical modelling of a single gaseous bubble that rises in liquid taking into account absorption of the gaseous inclusion by liquid. In particular, we are interested in the process of benzene alkylation that is conducted by the chemical interaction of ethylene and benzene in the presence of the Lewis catalyst. The gaseous inclusions of ethylene rise in a chemical reactor that is filled with the liquid mixture of benzene and catalyst. For the chemical reaction to occur ethylene needs to be first absorbed by the mixture, so the slow and limited miscibility of ethylene limits the reaction rates, and in order to obtain the desired amounts of the products the reactants are normally supplied in excessive quantities. The alkylation of benzene, used for manufacturing of ethylbenzene and isopropylbenzene, is carried out in the world on a massive scale with the overall consumption of up to 75% of benzene that is produced in the world. Ethylbenzene is further used as an intermediate for the production of styrene.

In the present work, the gas/liquid mixture is represented as a heterogeneous binary system with the mass transfer through phase boundaries. The evolution of the mixture is carried out on the basis of the phase-field approach. Within this approach, the interface is represented as a transitional layer of a finite thickness, and the concentration field C, that is defined as the mass fraction of one liquid in the mixture, is used to trace the position and shape of the interface. The specific free energy function is defined as a function of concentration and gradient of concentration [1]

$$f(C, \nabla C) = f_0(C) + \frac{\epsilon}{2}(\nabla C)^2 \qquad (1)$$

Here, the second term, proposed by Cahn and Hilliard, is introduced to take into account the surface tension effects. The capillary constant ϵ is assumed to be sufficiently small so this term is negligible everywhere except for the places of large concentration gradients, i.e. at interfaces. The double-well potential is frequently used for the classical part of the free energy function, f_0.

The free energy function (1) can be used to re-derive the Navier-Stokes equations for the mixture. The resultant full Cahn-Hilliard-Navier-Stokes equations are however very complex for numerical treatment because of the quasi-compressibility effect that forces to use the full continuity equation even for description of two incompressible liquids. In the current study the Boussinesq approximation of the full equations was used in order to determine the evolution of the gas inclusions rising in liquid.

The governing equations reflect the conservation of momentum, species, and mass [1]:

$$\frac{\partial \vec{u}}{\partial t} + (\vec{u} \cdot \nabla)\vec{u} = -\nabla \Pi + \frac{1}{Re}\nabla^2 \vec{u} - C\nabla\mu + 2GrC\vec{\gamma}, \qquad (2)$$

$$\frac{\partial C}{\partial t} + (\vec{u} \cdot \nabla)C = \frac{1}{Pe}\nabla^2\mu, \qquad (3)$$

$$\nabla \cdot \vec{u} = 0, \qquad (4)$$

$$\mu = Gr(\vec{r} \cdot \vec{\gamma}) + 2AC + 4C^3 - Ca\nabla^2 C. \qquad (5)$$

Here, the common notations are used for the variables; $\vec{\gamma}$ is the unit vector directed upward. These equations are applied to the whole multiphase system, including the interface. One can notice that the Navier-Stokes equation contains an additional (Korteweg) force that defines the interface morphology. The diffusion is defined by the generalized Fick's law, i.e. through the gradient of the chemical potential μ. As a result, in addition to the usual concentration diffusion the diffusion term includes the effect of barodiffusion.

The above equations are written in non-dimensional form. The equations include the following non-dimensional parameters: the Grashof number, $Gr = \varphi \frac{gL*}{\mu*}$, the Reynolds number, $Re = \frac{\rho*\mu*^{1/2}L*}{\eta*}$, the Peclet number, $Pe = \frac{\rho*L*}{\alpha\mu*^{1/2}}$, the capillary number, $Ca = \frac{\epsilon}{\mu*L*}$. Here $L*$ is the typical size, ρ_1 is the density scale, $\mu*$ is the unit of the chemical potential, η_1 is the viscosity scale, α is the mobility constant, and $\varphi = (\rho_2 - \rho_1)/\rho_1$ is the density contrast, with ρ_2 and ρ_1 being the densities of the pure components of the binary mixture.

We model the rise of an isolated single bubble numerically using the finite difference approach. The computational domain is represented by a vertical cylinder with a circular cross section. The axisymmetric symmetry is assumed. The cylinder's radius is taken as the length scale. The radial and axial coordinates are denoted by r and z, respectively. The cylinder is closed at the bottom and top ends. We assume that the size of the bubble is sufficiently less than the radius of the computational domain, so the influence of the walls on the shape and evolution of the bubbles are negligible. For the same reason, the initial position of the bubble is chosen at z = 0.5 (sufficiently far from the lower end), and the height of the cylinder, H = 6, is also chosen so large to observe a sufficiently long rise of the bubble before it reaches the upper end.

The governing equations (2)-(5) are supplemented with the following boundary conditions:

At the lower end: $z = 0 : u_r = u_z = 0, \partial C/\partial z = 0, \partial \mu/\partial z = 0$

$$(10)$$

At the upper end: $z = H : u_r = u_z = 0, \partial C/\partial z = 0, \partial \mu/\partial z = 0$

(11)

At the centreline: $r = 0 : u_r = u_z = 0, \partial C/\partial r = 0, \partial \mu/\partial r = 0$

(12)

At the tube's wall: $r = 1 : u_r = 0, \partial u_z / \partial r = 0 \; \partial C/\partial r = 0, \partial \mu/\partial r = 0$

(13)

Thus, all walls are assumed to be impermeable. The no-slip boundary conditions are used for the velocity field. For the chemical potential, we impose the absence of the diffusive flux through the walls. The conditions for the concentration should reflect the wetting conditions. We however consider the simplest case when the molecules of the mixture components interact with the wall equally, so the contact angle is 90^0. In fact, we are interested in the evolution of the bubble far from the boundary conditions, so the wetting properties are not important for the current study. The boundary conditions at the centreline are written to reflect the axial symmetry.
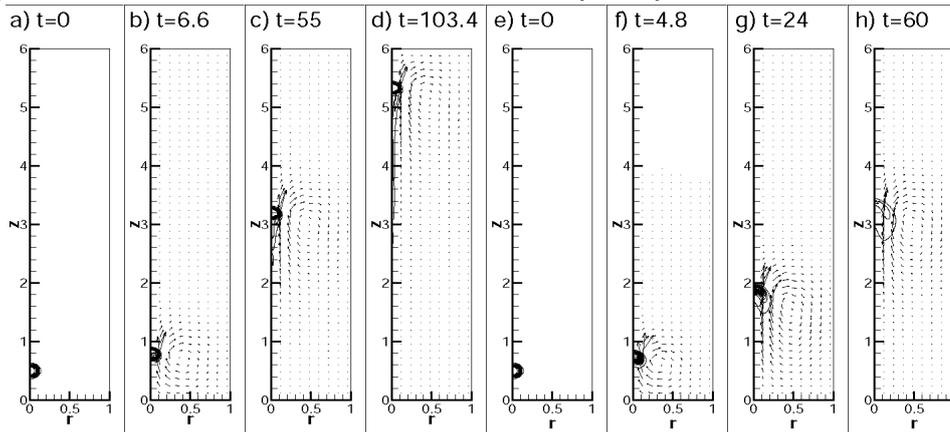
Fig. 1. The snapshots showing the stages of the rising bubbles at Pe=10^7 (a-d) and Pe=10^5 (e-h). The lines represent the concentration fields, and the vectors depict the velocity fields. The results are obtained for H=6, r_0=0.1, A=-0.5, Gr=0.1, Re=100, Ca=10^{-4}. The actual time moments are depicted in the figure.
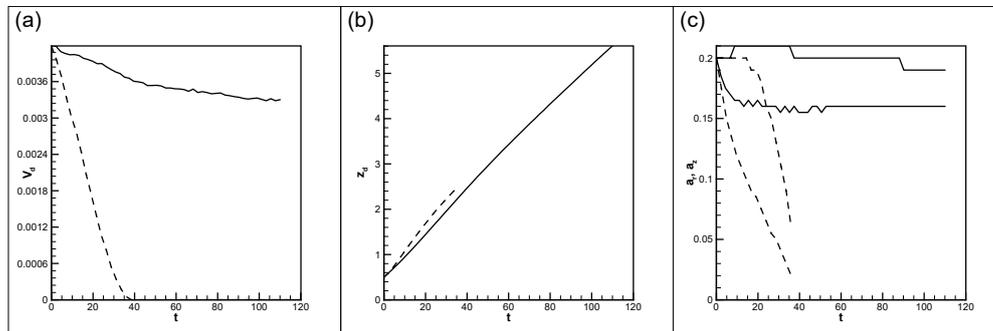
Fig 2. (a) The droplet's volume vs. time; (b) the vertical position vs. time; and (c) the size of the droplet in the radial and vertical directions vs. time. The curves are plotted for two Peclet number (Pe=10^7 – solid lines and Pe=10^5 – dashed lines) and for the other parameters as in Figure 1.

Figure 1 depicts the typical shapes of the rising bubbles and Figure 2 shows the time evolution of the bubble's volume, position and shape. The results are given for two different values of the Peclet number, for the cases with weaker and stronger interfacial diffusion. In the first case, Pe=10^7, one sees that the bubble's shape is deformed over the rise, but its volume just slightly decreases over the rise. In the second case, for Pe=10^5, the bubble becomes fully absorbed by the ambient liquid before the bubble reaches the middle of the computational domain. The dissolution of the bubble is accompanied by the convective motion near its surface, with the intensity of the motion determined by the Reynolds number, so at higher Reynolds numbers the bubbles rise faster, but they decrease in size also much faster.

Currently, we have investigated the evolution of the bubble in the case of weaker interfacial diffusion. We have compared the obtained results with the data available for immiscible systems, and found that all classical formulae earlier obtained on the bases of other theoretical and experimental approaches can be successfully reproduced with the help of the phase-field approach. In future work, we aim to focus on the effect of absorption, the rate of absorption of bubbles of different sizes, and the influence of the absorption on the speed of bubble rise and on the evolution of the bubble shape.

References

1. Vorobev A., Boussinesq approximation of the Cahn-Hilliard-Navier-Stokes equations // Phys. Rev. E, 82(5), 2010, 056312, 10pp.