

## NUMERICAL STUDY OF ETHYLENE BUBBLE RISING IN BENZENE FLOW

*Elena Khlebnikova<sup>1\*</sup>, Anatoliy Vorobev<sup>2</sup>, Elena Ivashkina<sup>1</sup>, Tatyana Lubimova<sup>3</sup>*

<sup>1</sup> Tomsk Polytechnic University, Tomsk, Russia

<sup>2</sup> Southampton University, Southampton, UK

<sup>3</sup> Perm State University, Perm, Russia

Received October 5, 2017; Accepted December 23, 2017

---

### Abstract

The bubble rising in viscous liquids is enough common in many industrial processes and an important fundamental problem in fluid dynamics. In the current work, we deal with the numerical modeling of a single gaseous bubble that rises in liquid. In particular, we are interested in benzene alkylation that is conducted by the chemical interaction of ethylene and benzene in the presence of the Lewis catalyst. Thus, the rate of absorption is the main focus of this work.

**Keywords:** : bubble rising; ethylbenzene; numerical approach; modeling; multiphase flow.

---

## 1. Introduction

The reactants mixing modeling is needed in various processes of chemical engineering [1-4]. The understanding of the system flow dynamics is a great importance in engineering applications. Rising bubbles in liquid have been enough studied theoretically [5-6] and experimentally [7-8]. In such systems slow and limited miscibility of gaseous species in liquids frequently limits reaction rates, so reactants are supplied in excessive quantities in order to obtain the desired amounts of the products. The mixing rate would strongly depend on the contact area of the phases, and, typically, to increase the contact between the phases, the gaseous phase is fed into a liquid-filled reactor at in the form of small bubbles.

In particular we are interested in alkylation of benzene with ethylene for ethylbenzene production. This technology is the large-capacity commercial process in terms of benzene consumption and processing: 75% of produced in the world benzene is used in production of ethylbenzene and isopropylbenzene. Market of ethylbenzene is closely linked to 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 gas/liquid mixture using the phase field approach was examined.

## 2. Experimental

The interface separating gas/liquid mixture was represented as a transitional boundary of finite thickness. The concentration field  $C$ , defined as the mass fraction of one liquid in the mixture, is used to trace the evolution of the interfacial boundary. The specific free energy function is defined as a concentration function and gradient of concentration [9]

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

In this expression, the second term, proposed by Cahn and Hilliard, takes into account the surface tension effects. The capillary constant  $\epsilon$  is assumed sufficiently small so to be able to neglect this term everywhere except at the places of large concentration gradients, where the interface is located. The double-well potential is frequently used for the classical part of the

free energy function,  $f_0$ , especially, when the primary interest is tracing the evolution of the immiscible interface.

The free energy function (1) can be used to re-derive the Navier-Stokes equations for the mixture. The so-obtained full Cahn-Hilliard-Navier-Stokes equations are however very complex for numerical treatment because of quasi-compressibility that forces to use the full continuity equation even for description of two incompressible liquids. The Boussinesq approximation [10] of the full equations was used to define the evolution of the gas bubble rising in benzene.

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

$$(1+\rho)\frac{\partial \vec{u}}{\partial t}+(\vec{u}\cdot\nabla)\vec{u}=-\nabla\pi+\frac{1}{Re}\nabla^2\vec{u}-Ca\nabla^2\mu+2GrC, \quad (2)$$

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

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

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

Here, the common notations are used for the variables. These equations are applied to the whole multiphase system, including the interface. One sees that the Navier-Stokes equation contains an addition force that defines morphology of the interface. The diffusion is defined the generalised Ficks law, i.e. through the gradient of the chemical potential  $\mu$ . The diffusion term includes in addition to the usual concentration diffusion, the barodiffusion terms.

$\vec{\gamma}$  is the unit vector directed upward.

The equations are written in non-dimensional form and include the following non-dimensional parameters;

$$\text{the Grashof number: } Gr = \varphi \frac{gL^*}{\mu^*}, \quad (6)$$

$$\text{the Reynolds number: } Re = \frac{\rho^*\mu^{*1/2}L^*}{\eta^*}, \quad (7)$$

$$\text{the Peclet number: } Pe = \frac{\rho^*L^*}{\alpha\mu^{*1/2}}, \quad (8)$$

$$\text{the capillary number: } Ca = \frac{\epsilon}{\mu^*L^*}, \quad (9)$$

where  $L^*$  - typical size;  $\rho^*$  - the density scale;  $\mu^*$  - the unit of the chemical potential;  $\eta^*$  - the viscosity scale;  $\alpha$  - the mobility constant; and  $\varphi = (\rho_2 - \rho_1)/\rho_1$  is the density contrast, with  $\rho_2$  and  $\rho_1$  being the densities of the pure components of the binary mixture.

The rise of an isolated single bubble was examined. The computational domain is represented by a vertical cylinder with circular cross section. 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. It is assumed that the size of the bubble is sufficiently less than 1 (the radius of the computational domain), so the influence of the boundary conditions imposed on the cylinder's walls can be neglected. 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 axisymmetric symmetry is assumed, and owing to the symmetry only a half of cylinder was considered.

The governing equations are supplemented with the boundary conditions below.

$$\text{At the lower end: } z = 0 : u_r = u_z = 0, \partial C / \partial z = 0, \partial \mu / \partial z = 0 \quad (10)$$

$$\text{At the upper end: } z = H : u_r = u_z = 0, \partial C / \partial z = 0, \partial \mu / \partial z = 0 \quad (11)$$

$$\text{At the centreline: } r = 0 : u_r = u_z = 0, \partial C / \partial r = 0, \partial \mu / \partial r = 0 \quad (12)$$

$$\text{At the tube's wall: } r = 1 : u_r = 0, \partial u_z / \partial r = 0, \partial C / \partial r = 0, \partial \mu / \partial r = 0 \quad (13)$$

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  $90^\circ$ . 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 centerline are written to reflect the axial symmetry.

The typical shapes of the rising bubbles for four different Reynolds numbers and for two different capillary numbers are shown in Fig. 1. The actual time moments and the actual sizes of the bubbles are shown as well.

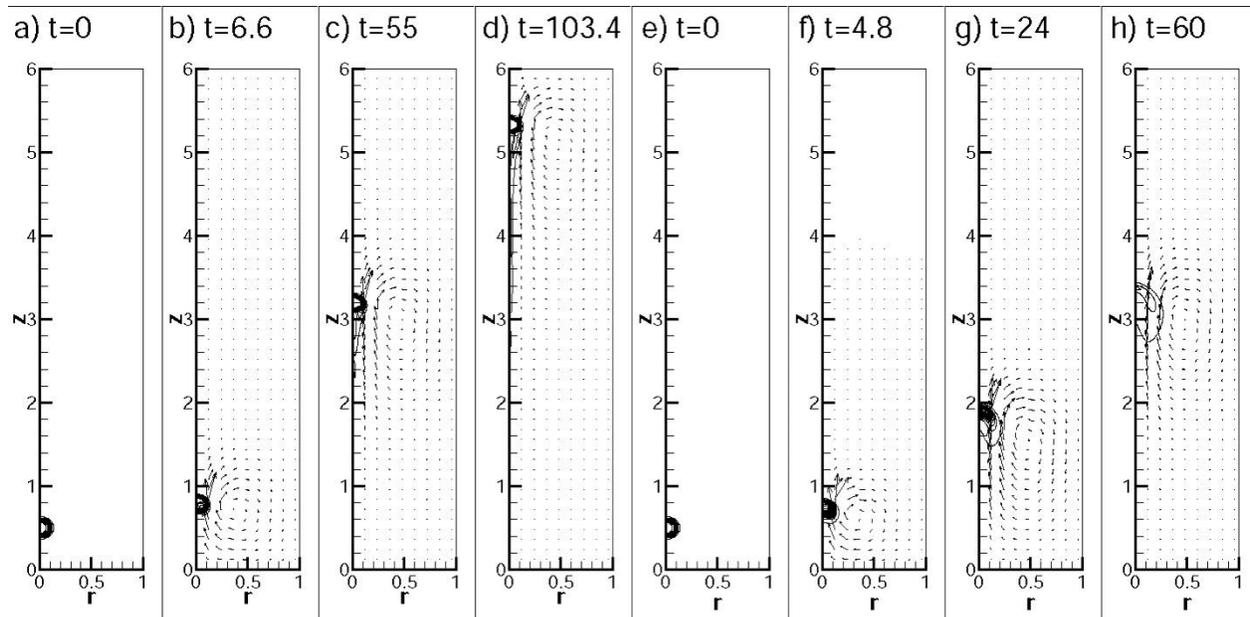


Figure 1. Bubbles at the middle of their rise for  $H = 6$ ,  $r_0 = 0.2$ ,  $A = -0.5$ ,  $Pe = 105$ ,  $Gr = 0.1$ , (a-d)  $Ca = 8 \cdot 10^{-4}$ ; (e-h)  $Ca = 2 \cdot 10^{-4}$ ; (a,e)  $Re = 10$ , (b,f)  $Re = 25$ , (c,g)  $Re = 50$ , and (d,h)  $Re = 100$

Obviously, the interface becomes thinner if the capillary number is lower. Deformation of the bubble is stronger at lower capillary numbers, which is explained by smaller surface tension coefficient at such numbers. In this case, at sufficiently higher Reynolds numbers, the bubble can even break. The moment of the break up is depicted in Fig. 1d. At  $Ca = 2 \cdot 10^{-4}$ , the bubble breaks up at  $Re > 50$ . At  $Ca = 8 \cdot 10^{-4}$ , the bubble breaks up at  $Re > 100$ .

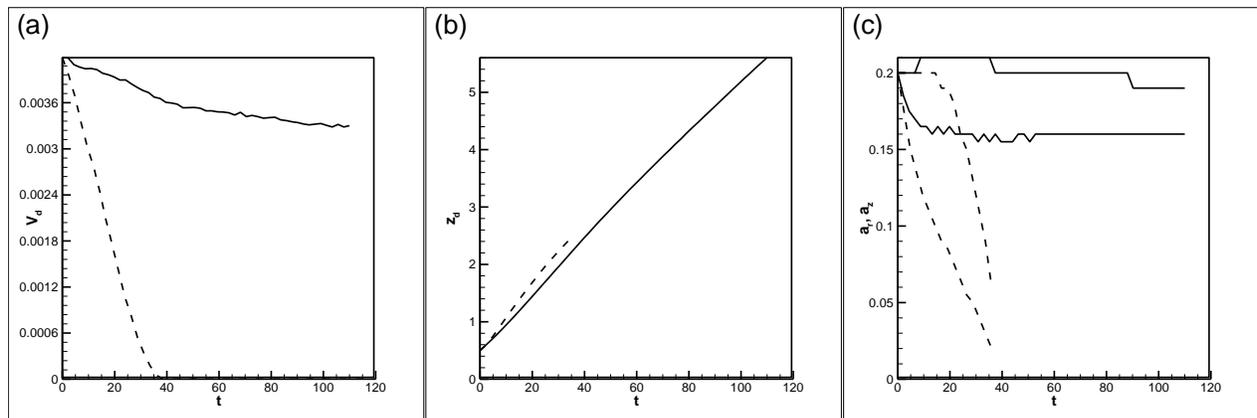


Figure 2. (a) The droplet's volume vs. time; (b) the vertical position vs. time; and (c) the size of the droplet in the radial and vertical directions vs. time. The curves are plotted for two Peclet number ( $Pe=10^7$  – solid lines and  $Pe=10^5$  – dashed lines) and for the other parameters as in Figure 1

The speed of the bubble rising is high at high Reynolds number. All bubbles in Fig. 1 started to rise from the same point and are shown at the similar positions, but at the different time moments, as the rise time is smaller if the Reynolds number is increased. The dissolution of

the bubble is accompanied by the convective motion near the bubble. The intensity of this motion is also increased at higher Reynolds numbers.

The results in Fig. 1 are shown for  $Pe = 10^5$ . At such Peclet numbers the diffusion through the interface is small, but it is different from zero. The actual sizes of the bubbles are indicated in Fig. 1. One sees that the diffusion is strongly intensified by convective flows, so that at higher Reynolds numbers the bubbles rise faster, but they decrease in size also much faster.

Fig. 2 shows the time evolution of the bubble's volume, position and shape.

An unexpected result is an impact of the Peclet number on the speed of rise. For the cases with higher Peclet numbers (weaker diffusion) the time of the initial acceleration is much lower and the attached speed of rise is also lower. The agreement between the Stokes prediction and the numerical results are getting better at higher Peclet numbers, which is expectable.

### 3. Conclusions

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.

### Acknowledgements

The study was financially supported by RFBR as part a research project № 16-51-10079 "Mixing dynamics in diffusion-limited reactions

### References

- [1] Meyers RA. Handbook Of Petrochemicals Production Processes. McGraw – Hill Professional. 2005.
- [2] Lee S. Encyclopedia of Chemical Processing. CRC Press. 2005. P. 3291.
- [3] Khlebnikova E, Bekker A, Ivashkina E, Dolganova I, Yurev E. Procedia Chemistry. 2015, 15: 42-48.
- [4] Belinskaya NS, Frantsina EV, Ivanchina ED, Popova NV, Zyryanova IV, Averyanova EV. IOP Conference Series: Earth and Environmental Science. 2016; 43: 1-6.
- [5] Davies RM, and Taylor FI. The Mechanics of Large Bubbles Rising through Extended Liquids and through Liquids in Tubes. Proc. R. Soc. Lond. 1950; A 200(1016):375-390.
- [6] Moore DW. The rise of a gas bubble in a viscous liquid, J. Fluid Mech. 1959; 6(1):113-130.
- [7] Ryskin G and Leal LG. Numerical solution of free-boundary problems in fluid mechanics. Part 2. Buoyancy-driven motion of a gas bubble through a quiescent liquid. J. Fluid Mech. 1984; 148:19-35.
- [8] Bhaga D and Weber ME. Bubbles in viscous liquids: shapes, wakes and velocities. J. Fluid Mech. 1981; 105:61-85.
- [9] Vorobev A. Boussinesq approximation of the Cahn-Hilliard-Navier-Stokes equations. J. Physical Review E 82, 2010; 056312.
- [10] Lowengrub J, Truskinovsky L. Quasi-incompressible Cahn-Hilliard fluids and topological transitions. R. Soc. Lond. Proc. Ser. A Math. Phys. Eng. Sci. 1998; 454(1978): 2617–2654.

*To whom correspondence should be addressed: Dr. Elena Khlebnikova, Chemical Engineering Department, Tomsk Polytechnic University, 30, Lenin Avenue, Tomsk, 634050, Russia; tel.: (+7-3822) 563443; fax: (+7-3822) 563-435; e-mail: [elena.khle@gmail.com](mailto:elena.khle@gmail.com)*