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Drops moving in flow with chemical reaction

P. Neittaanmäki¹ and V. Rivkind²

Abstract. We propose a free boundary model described by coupled Navier–Stokes and chemical reaction equations with discontinuous coefficients to simulate the chemical reactions in viscous drops moving in a viscous incompressible fluid. Approximation of the solution by a special finite element method (FEM) with a method of mapping is discussed. Several numerical results are presented.

Keywords: Navier–Stokes, chemical reactions, free boundary problems, finite element method.

Subject classification: 35F30, 35Q10, 35R05, 35R35, 65M60, 65N30, 76D05.

1. Introduction

This paper is devoted to the problems of chemical reactions in viscous incompressible drops moving in a viscous incompressible fluid. These types of problems arise from many practical applications. The mathematical models governing the phenomena are free boundary problems for the coupled Navier–Stokes and chemical reaction equations with discontinuous coefficients and with two mediums (inside of the drop and outside of the drop which is an infinite domain). It is assumed that the process is isothermic and that the density of the substances, the diffusion coefficients and the viscosities are constant.

In the case of the first order chemical reaction we present a model under the assumption that the normal component of the velocity on the boundary of drops is proportional to the flow of chemical component across this boundary. We also consider

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the case of a second order chemical reactions inside and outside of drops.

For both of these models it is possible to prove the existence and uniqueness of the solution and the convergence of an approximate solution to the exact one with some assumptions about the data of the problem. The methods for proving these results are similar to those used for coupled Navier–Stokes and Stefan equations in [4].

Several numerical tests have been performed for these models. Based on these tests we can draw conclusions on the feasibility to model the flow of drops in chemical reactors by approximate models like diffusion boundary layer and Kronig–Brink model inside of drops and boundary layer model outside of drops.

The case of movement of drops was considered in [1], [7]. The theorems of existence, uniqueness and smoothness of solution with certain assumptions on initial data were discussed in [1], [7]. For coupled Navier–Stokes and Stefan equations (in bounded domain, one phase problem) we refer to [2], [8], and for two phase problems including drops to [4].

2. Mathematical model of the first order reaction

We consider the fluid with the density ρ_1 and the viscosity ν_1 which occupies a closed domain Ω_1 (inside of drop). The volume V of Ω_1 is fixed and has an unknown free boundary Γ . Another fluid (stream) with the density ρ_2 and the viscosity ν_2 occupies the domain $\Omega_2 = \mathbf{R}^3 \setminus \Omega_1$ (outside of drop), see Fig. 1.

The mathematical model for the first order chemical reaction of viscous incompressible drops con-

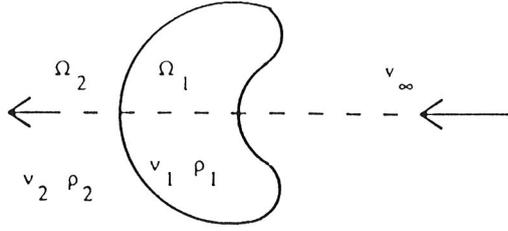


Figure 1.

sists of the determination of the unknown (free) boundary Γ , the velocity vector \vec{v} , the concentrations Θ_1, Θ_2 as well as the pressure p , which satisfy in Ω_i ($i=1,2$) the following system of equations

$$\left\{ \begin{array}{l} -\frac{1}{\text{Re}_i} \Delta \vec{v} + (\vec{v} \cdot \nabla) \vec{v} + \frac{1}{\rho_i} \nabla p = 0, \\ \nabla \cdot \vec{v} = 0, \quad \text{in } \Omega_1, \Omega_2, \\ \vec{v}|_{|x| \rightarrow \infty} = \vec{v}_\infty, \quad \vec{v} \cdot \vec{n}|_\Gamma = \left[\lambda \frac{\partial \Theta}{\partial n} \right] \Big|_\Gamma, \\ [\vec{\tau} \cdot S(\vec{v}) \cdot \vec{n}]|_\Gamma = 0, \\ \text{Re}_i = \frac{|\vec{v}_\infty| a}{\nu_i}, \quad |\vec{v}_\infty| = \text{const}, \quad i = 1, 2, \\ \text{Re}_1 = \mu \text{Re}_2, \quad \mu = \text{const}, \quad \text{Re}_2 = \text{Re}, \\ \int_{\Omega_1} dx = V, \\ -\frac{1}{\text{Pr}_i} \Delta \Theta + \vec{v} \nabla \Theta + k_i^2 \Theta = f_i, \quad \text{in } \Omega_1, \Omega_2, \\ \Theta|_\Gamma = \Theta_0, \quad \Theta|_{|x| \rightarrow \infty} \rightarrow 0, \\ \text{Pr}_1 = \mu_1 \text{Pr}_2, \quad \mu_1 = \text{const}, \quad \text{Pr}_2 = \text{Pr}, \\ \int_{\Omega_i} f_i dx = 0, \quad i = 1, 2. \end{array} \right. \quad (1)$$

Here $\text{Re}_i > 0$, $i = 1, 2$ denote Reynolds numbers and $\text{Pr}_i > 0$, $i = 1, 2$ Prandtl numbers. \vec{n} is the outward unit normal vector to Γ , $S(\vec{v})$ is matrix with elements $S_{ij}(\vec{v}) = \nu \left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right)$, and ν_i , ρ_i , λ_i are positive constants, ν_i is a viscosity in Ω_i , ρ_i is a density in Ω_i and k_i^2 are velocities of chemical reactions.

Function z determining Γ satisfies the equation

$$\Delta \Big|_\Gamma z - \alpha(\Theta) z = \frac{1}{\sigma(\Theta)} \left[-\frac{1}{\rho} p + \vec{n} \cdot S(\vec{v}) \cdot \vec{n} \right] \Big|_\Gamma, \quad (3)$$

where $\Delta|_\Gamma$ is the Laplace-Beltrami operator, $[\vec{v}]|_\Gamma$ denotes a jump of the function \vec{v} on Γ . Further, σ is the surface tension coefficient and, usually, it is considered that $\sigma(\Theta)$ depends linearly on the concentration: $\sigma(\theta) = \sigma_0 - \sigma_1 \theta$. For the coefficient $\alpha(\Theta)$ we assume that $\alpha(\Theta) \sim g(\rho_2 - \rho_1)$, where g is the acceleration of the weight force.

We remark that without the term $(\vec{v} \cdot \nabla) \vec{v}$ and the term $\vec{v} \nabla \Theta$ in (2), and moreover, if $f_i = 0$, $\Theta_0 = 0$, then the liquid in Ω_1 occupies a ball of radius a .

The theorem of existence and uniqueness of the solution for (1)–(3) can be proved in a similar way as in [4] for coupled the Navier-Stokes and Stefan equations (see [5]).

3. Mathematical model of the chemical reaction of the second order

Let us consider a mathematical model for a chemical reaction of the second order. In this case the systems are also governed by hydrodynamical equations (1)–(2). However, the boundary condition for the normal component of the velocity is changed to $v \cdot n|_\Gamma = 0$ and the coupled equations,

$$\frac{\partial \theta_1}{\partial \tau} + \text{Pe}_1 \left(v_r \frac{\partial \theta_1}{\partial r} + \frac{v_\theta}{r} \frac{\partial \theta_1}{\partial \varphi} \right) = \Delta \theta_1 - k_2 \theta_1 \theta_2 \quad (4)$$

$$\frac{\partial \theta_2}{\partial \tau} + \text{Pe}_1 \left(v_r \frac{\partial \theta_2}{\partial r} + \frac{v_\theta}{r} \frac{\partial \theta_2}{\partial \varphi} \right) = n \Delta \theta_2 - \frac{k_2}{m} \theta_1 \theta_2 \quad (5)$$

which describe the concentrations of mediums inside Ω_1 and outside Ω_2 of the drop, have been added. Here τ is the Fourier criterion, $\tau = \frac{Dt}{a^2}$, D is a diffusion coefficient, t is a time, φ the dimensionless polar angle, $r = \frac{r_1}{a}$ the radius, and k_2 is the velocity of the chemical reaction.

The boundary conditions for (4)–(5) depend on the limiting resistance of the dispersed phase or

fluid flow. In the case of dispersed phase the concentrations of the medium inside of the drop satisfy the boundary conditions

$$\Theta_1|_{\Gamma} = 1, \quad \frac{\partial \Theta_2}{\partial n}|_{\Gamma} = 1, \quad (6)$$

and initial conditions

$$\Theta_1|_{\tau=0} = 0, \quad \Theta_2|_{\tau=0} = 1. \quad (7)$$

If the process of the outside flow has a limiting resistance we have the exterior boundary value problem. Boundary and initial conditions used in this case read as follows

$$\Theta_1|_{\Gamma} = 1, \quad \frac{\partial \Theta_2}{\partial n}|_{\Gamma} = 0, \quad (8)$$

$$\Theta_1|_{|x| \rightarrow \infty} = 0, \quad \Theta_2|_{|x| \rightarrow \infty} \rightarrow 1, \quad (9)$$

$$\Theta_1|_{\tau=0} = 0, \quad \Theta_2|_{\tau=0} = 1. \quad (10)$$

4. The finite element method for the equations with chemical reactions

We propose the following scheme for finding the numerical solution to the problem (1)–(3):

Step 1. If we know the $(n-1)$ -th approximation of $v_h^{(n-1)}$, $p_h^{(n-1)}$, $\Theta_h^{(n-1)}$, $\Gamma_h^{(n-1)}$, we solve by FEM with discretization parameter h the Navier–Stokes problem (1) with fixed boundary conditions on the fixed surface Γ_h . As a result we get $v_h^{(n)}$, $p_h^{(n)}$. This main step has an iterative structure based on the following procedure: i) apply the domain decomposition method for problems with discontinuous parameters on the Γ , and ii) solve the nonlinear Navier–Stokes equation iteratively, using the solution of the linearised Navier–Stokes equation at the start of each step.

Step 2. Once we have $v_h^{(n)}$, we can obtain $\Theta_h^{(n)}$ from the boundary value problem (2) with fixed boundary. Since the chemical reaction equations also have discontinuous parameters, we use a domain decomposition method and follow an iterative procedure analogous to the previous step to solve (2).

Step 3. Here we determine a new approximation of the free surface Γ_h from the boundary conditions (3) which relates the mean curvature of surface and the jump of normal projection of stress tensor. This problem will be solved using the n -th iteration of the stress tensor by the solution of the problem for the special parabolic equation:

$$\frac{\partial \bar{z}}{\partial t} = \Delta|_{\Gamma} z - \alpha(\Theta)z - \frac{1}{\sigma(\Theta)} \left[-\frac{1}{\rho} p + nS(\bar{v}^{(n)})n \right]. \quad (11)$$

We solve the above first and second steps by mapping of flowfield onto a standard domain and applying the finite element method in curvilinear coordinates, connected with this mapping. After the determination of new shape of free surface we generate a new mapping and a new grid for finite element approximation on the next main iteration. This can be done by solving an equation similar to (11), and substituting on the stress place parameter, which governs the process of collapsing the grid to the center of the drop (for inside grid) or the value of steps to infinity (for outside grid). Typical grids used are given in Fig. 2.

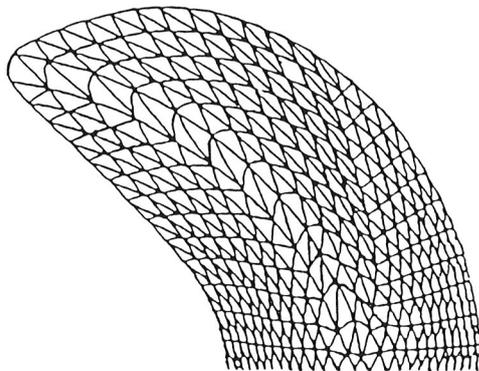


Figure 2.

We shall outline FEM in general curvilinear coordinates. Let $\{y^i\}_{i=1}^3$ denote the Euclidian coordinates and $y^i = y^i(x^1, x^2, x^3)$ a smooth function of curvilinear coordinates $\{x^i\}$. Then the metrics

g_{ij} are defined by the formula:

$$g_{ij} = \sum_k \frac{\partial y^k}{\partial x^i} \frac{\partial y^k}{\partial x^j} \equiv \frac{\partial y^k}{\partial x^i} \frac{\partial y^k}{\partial x^j} \quad (12)$$

The inverse of matrix g_{ij} will be called g^{ij} , $g^{ij}g_{jk} = \delta_k^i$. The strain tensor e in curvilinear coordinates is defined by

$$e^{ik} = 1/2(g^{kr}u_{;r}^i + g^{ir}u_{;r}^k), \quad (13)$$

$$u_{;r} = \frac{\partial u}{\partial r}.$$

The variational formulation of our Navier–Stokes boundary value problem (1) in general curvilinear coordinates is given by

$$\begin{aligned} & \int_{\Omega} e_h^{ik}(\vec{v})g_{ij}g_{kl}e_h^{j1}(\vec{\eta}_h)\sqrt{\det(g_3)}dx^1dx^2dx^3 \\ & = - \int_{\Omega} (v_h^k v_h^i)_{;k} g_{ij} \eta_h^j \sqrt{\det(g_3)} dx^1 dx^2 dx^3 \\ & + \int_{\Omega} p_h / \rho_h \eta_{h;k}^k \sqrt{\det(g_3)} dx^1 dx^2 dx^3 \\ & + \int_{\partial\Omega} e^{ik}(v_h)g_{ij}v_h^j g_{kl}n^l \sqrt{g_2} ds^1 ds^2 \\ & - \int_{\partial\Omega} g^{ik} p_h / \rho_h g_{ij} v_h^j g_{kl} n^l \sqrt{\det(g_2)} ds^1 ds^2, \end{aligned} \quad (14)$$

where $\det(g_3)$ is the Jacobian of metrics g_{ij} on volume and $\det(g_2)$ is Jacobian of metrics on the boundary $\partial\Omega$.

The equation $\operatorname{div} \vec{v} = 0$ is satisfied in the mean sense:

$$\int_{\omega} p_h \eta_{h;k}^k \sqrt{\det(g_3)} dx^1 dx^2 dx^3 = 0 \quad (15)$$

on some submanifolds ω of domain Ω .

In the axially symmetric case which we are interested, $\partial\Omega$ is a coordinate surface $x^2 = \text{const}$ and x^3 is polar angle. Therefore some elements in tensor convolutions became equal to zero.

The variational form of chemical equations in curvilinear coordinates can be written in a similar manner. As usual we find the functions $\vec{v}_h^i \in P_2$;

$p_h \in P_1$ that satisfy (14) for arbitrary smooth test function η_h^k belong to the same class as v_h .

Like in [3], [4], [6], [8] it is possible to prove that our approximate solution \vec{v}_h, Θ_h converge to the exact one and the rate of convergence is of order $O(\bar{h})$ in the space $H^1(\Omega_1) \cap H^1(\Omega_2)$ for velocities and of order $O(\bar{h}^2)$ for concentrations (Θ_1, Θ_2 in the case of chemical reactions) where \bar{h} is an average step of a FE-grid. For details see [5].

In a similar way we can proceed in numerical solving the model of chemical reaction of the second order.

5. Numerical examples

In the numerical tests, we have applied the method of mapping with special finite element grid (see Fig. 2). The FE-approximation used the finite elements P_2 for approximations \vec{v}_h and Θ_h and P_1 for the pressure p_h .

Fig. 3 illustrates the evolution of a concentration of medium in the case of the first order nonstationary chemical reaction with parameters: $\text{Re} = 50$, $\text{We} = 3$, $\mu = 2$, $\text{Pe}_1 = \text{Pe}_2 = 500$. In this case the equations (2) are nonstationary:

$$\frac{\partial \Theta}{\partial \tau} + \text{Pe}_i(\vec{v} \cdot \nabla \Theta) + k^2 \Theta = \Delta \Theta, \quad (16)$$

$$\Theta|_{|x| \rightarrow \infty} \rightarrow \infty. \quad (17)$$

All other conditions are the same as in stationary model (1)–(3).

From Fig. 3 it is clear that changing of the shape of drop is more slowly than the evolution of the diffusion process. It is possible to use these results for the simplification of practical calculations.

Fig. 4 presents the evolution of concentrations of both mediums in time for the case of chemical reaction of the second order inside of a drop. The main attention of the computations was focused on the relationship of the criteria of chemical reaction (the Sherwood number, average concentrations, degree of extraction of substance, and others) to the speed of motion of the drop (Re_i), the Peclet criterion, and the numbers $\mu = \nu_1/\nu_2$. The

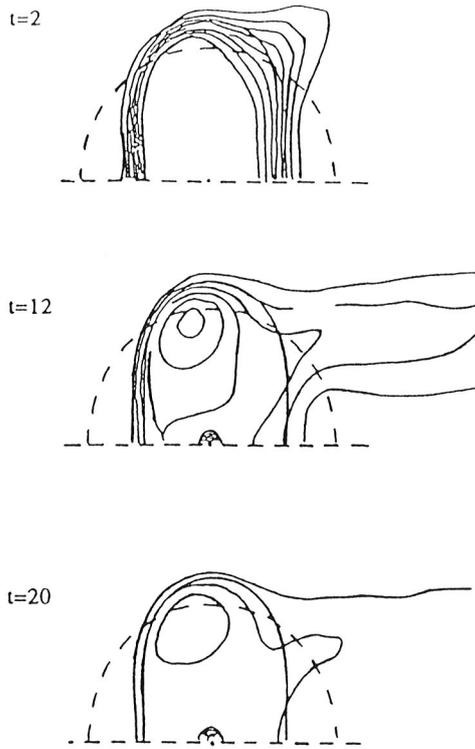


Figure 3.

evolution of the average concentrations on time t ($\tau = Dt/a^2$) can be seen in Fig. 4 for $m = l = 1$, $We = 3$, $\mu = 1, 2$ and $k = 100$ serve as an example of such dependences. We note the monotone character of the dependence of the quantities on $Re_1 = Re_2$, Pe , and μ . Just as in the case of the mass transfer without chemical reaction, the effect of the parameters Re_1 , Re_2 and μ can be accounted with satisfactory accuracy by introducing $Pe_{eff} = Pe \cdot |v_{max}|$. In graphical illustration of numerical tests of Fig. 4 following parameters are chosen: all graphs correspond the case of $\mu = \frac{v_1 \rho_1}{v_2 \rho_2} = 1.2$; $Re_1 = Re_2 = Re$; $k = 100$; $l = m = 1$; to $We = 0.2$; curve 1 is for $Re = 0$, $Pe = 160$; curve 2 is for $Re = 10$, $Pe = 160$ ($Pe_{eff} = 184$); curve 3 is for $Re = 0$, $Pe = 320$;

curve 4 is for $Re = 50$, $Pe = 160$ ($Pe = 272$); curve 5 is for $Re = 10$, $Pe = 400$; curve 6 is for $Re = 50$, $Pe = 400$. Curve 3 of Fig. 4 illustrates the concentration Θ_1 and Θ_2 for values $Re_1 = Re_2 = 50$, $\mu = 1, 2$ and $Pe = 160$, while curve 4 are quantities for $Re_1 = Re_2 = 0$, $Pe = 320$ and $\mu = 1, 2$, and has a similar Pe_{eff} as that of curve 3. Thus, in Fig. 4 the curve 3 and 4 practically coincide. Such regularities are also observed for other values of the parameters Re_i , μ , m , l , and k , and also for the degree of extraction of substance, the Sherwood number, and others.

All this indicates the possibility of extending the results and conclusions, based on the Hadamard-Rybczynski hydrodynamics of the drop, to a wide range of numbers Re_i and μ with replacement of the number Pe used by the numbers Pe_{eff} . We remark that the same conclusions apply in the case of chemical reactions of first order.

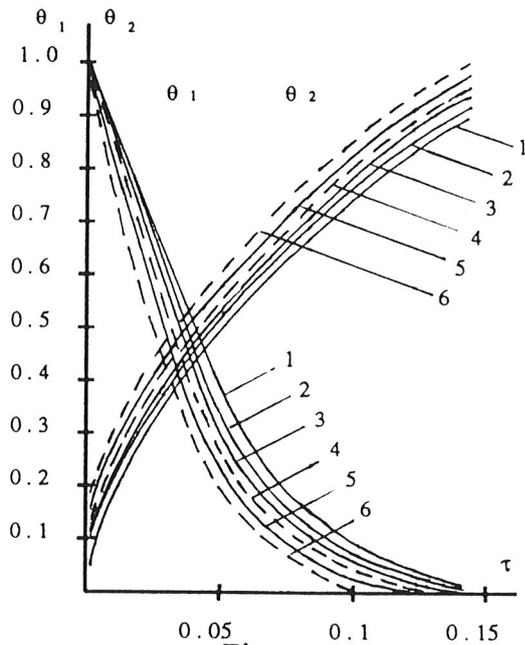


Figure 4.

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