

APPLICATION OF THE DENSITY-FUNCTIONAL METHOD
FOR NUMERICAL SIMULATION OF FLOWS
OF MULTISPECIES MULTIPHASE MIXTURES

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Numerical examples of application of the density functional used to describe isothermal flows of two-phase two-species mixtures are given. The following flows are calculated in a two-dimensional formulation: impact of a drop on a liquid layer, breakdown of a drop in the velocity field of the Couette flow, formation of the wetting angle of a drop on a solid surface, and development of the Rayleigh–Taylor and Kelvin–Helmholtz instabilities at the gas–liquid interface.

Key words: *two-phase flow, density-functional method, drop deformation.*

Introduction. The density-functional method allows one to describe a multispecies multiphase mixture in a continuous manner, without density jumps and phase interfaces. This is achieved by introducing squared gradients of densities of the components into the expression for free energy of the mixture (or into the expression for entropy) [1–5]. The main advantage of this approach is the possibility of finding the distribution of phases in space as one of the results of solving a single continuous problem. In this case, there is no need to *a priori* prescribe the geometry of interphase surfaces, solve hydrodynamic equations for each phase separately, and use conditions on jumps. An important feature of the density-functional method is the allowance for the structure of the interphase region. This leads to effects observed for a liquid flow in a capillary with surface-active walls [6] and for the spectrum of capillary-gravitational waves [7]. In addition, the method allows one to describe the flow of a gas–condensate mixture in a capillary [8].

In the present work, we consider the use of the density-functional method in numerical simulations of multiphase flows. The governing equations of the theory are derived in Sec. 1. Section 2 deals with the algorithms of numerical implementation of these equations and examples of calculations of a number of typical isothermal two-phase flows: coalescence of a drop with a liquid layer, breakdown of a drop in the velocity field of a shear flow, drop spreading on a solid surface, and development of the Rayleigh–Taylor and Kelvin–Helmholtz instabilities at the gas–liquid interface. Finally, the results obtained and the further capabilities of the method are discussed.

1. Governing Equations of the Density-Functional Theory. In the present work, we consider isothermal flows only. The basic theoretical postulates for this case are briefly described below [3]. The theory of nonisothermal flows is described in [4].

Let an M -species mixture (gas or liquid) fill a domain D with a piecewise-smooth boundary ∂D corresponding to the contact with a motionless solid phase. We use the following notation: n_i is the molar density of the i th species; the subscripts i , j , and k are assumed to run through the values $1, \dots, M$ corresponding to the number of the species in the mixture, and the subscripts a , b , and c take the values 1, 2, and 3 corresponding to the Cartesian coordinates x^a . If not indicated otherwise, summation is performed over repeated subscripts. The following abbreviated notation is used for derivatives: $g_{,i} = \partial g / \partial n_i$ and $\partial_a g = \partial g / \partial x^a$.

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Static Conditions. The functional of free energy of the mixture is given by the expression

$$F = \int_D \omega dV + \int_{\partial D} f_* dA, \quad (1.1)$$

where $\omega = f + (1/2)\nu_{ij} \partial_a n_i \partial_a n_j + \rho\varphi$, $f = f(n_i)$ is the free energy of a unit volume of a homogeneous mixture, $f_* = f_*(n_i)$ is the free energy of a unit surface, $\nu_{ij} = \nu_{ij}(n_k)$ are the coefficients of a positive symmetric matrix, dV and dA are the volume and surface elements, $\varphi = \varphi(x^a)$ is the gravitational potential, $\rho = m_i n_i$ is the mass density of the mixture, and m_i is the molar mass of the i th component.

The equilibrium states of the mixture are critical points of functional (1.1) for a fixed number of particles of components in the mixture

$$N_i = \int_D n_i dV. \quad (1.2)$$

Calculating the variation of functional (1.1), we obtain the expression

$$\delta F = \int_D \Phi_i \delta n_i dV + \int_{\partial D} \Phi_{i*} \delta n_i dA. \quad (1.3)$$

Here

$$\Phi_i = f_{,i} + m_i \varphi + (1/2)\nu_{jk,i} \partial_a n_j \partial_a n_k - \nu_{ij,k} \partial_a n_j \partial_a n_k - \nu_{ij} \Delta n_j;$$

$$\Phi_{i*} = f_{*,i} - \nu_{ij} l_a \partial_a n_j,$$

where l_a is the inner normal to the surface ∂D and $\Delta = \partial_a \partial_a$.

Using expression (1.3), we can write the variation equation for the equilibrium states with the Lagrangian multipliers Λ_i

$$\delta F - \Lambda_i \delta N_i = 0,$$

which yields the system of elliptic equations

$$\Phi_i - \Lambda_i = 0 \quad (1.4)$$

and the boundary conditions

$$\Phi_{i*} = 0. \quad (1.5)$$

The Lagrangian multipliers Λ_i should be determined by solving problem (1.4), (1.5) with respect to unknown density fields of the component n_i from additional integral conditions (1.2).

Dynamic Conditions. As the governing hydrodynamic equations for isothermal flows, we use the conventional equations for densities and momentum [9]

$$\partial_t n_i + \partial_a I_{ia} = 0; \quad (1.6)$$

$$\rho(\partial_t v_a + v_b \partial_b v_a) = \partial_b p_{ab} - \rho \partial_a \varphi, \quad (1.7)$$

where I_{ia} is the flux vector of the i th component, $v_a = \rho^{-1} m_i I_{ia}$ is the mean-mass velocity, and p_{ab} is the stress tensor in the mixture.

We introduce auxiliary quantities: the vector of the diffuse flux of the i th component $Q_{ia} = (I_{ia} - n_i v_a)$, $\tau_{ab} = p_{ab} - \sigma_{ab}$, and

$$\sigma_{ab} = (\omega - \Phi_i n_i) \delta_{ab} - \nu_{ij} \partial_a n_i \partial_b n_j. \quad (1.8)$$

The hydrodynamic description involves usual boundary conditions on dynamic variables: the no-slip condition

$$v_a \Big|_{\partial D} = 0 \quad (1.9)$$

and the impermeability condition for the diffuse fluxes

$$l_a Q_{ia} \Big|_{\partial D} = 0. \quad (1.10)$$

In addition, we assume that the boundary conditions (1.5) are satisfied in both static and dynamic cases.

We determine the functional of the total energy of the system, including the free energy (1.1) and the kinetic energy of the mixture:

$$E = F + \frac{1}{2} \int_D \rho v_a v_a dV. \quad (1.11)$$

Direct calculation of the derivative of functional (1.1) in time with allowance for expression (1.3) and Eqs. (1.5)–(1.10) yields the relation

$$\frac{dE}{dt} = \int_D \Sigma dV, \quad (1.12)$$

where

$$\Sigma = -\tau_{ab} \partial_a v_b + Q_{ia} \partial_a \Phi_i. \quad (1.13)$$

The hydrodynamic model should be dissipative, i.e., the following inequality, which is an analog of the condition of nonnegative entropy production, should be satisfied:

$$\frac{dE}{dt} \leq 0. \quad (1.14)$$

Owing to relations (1.12) and (1.13), inequality (1.14) is satisfied if the following inequality holds:

$$\Sigma \leq 0. \quad (1.15)$$

Note, relations (1.12) and (1.13) allow us to interpret τ_{ab} as the tensor of viscous stresses, since it is this component of the total stress tensor p_{ab} that contributes to dissipation. Hence, σ_{ab} should be interpreted as the tensor of static stresses in the mixture, because it is independent of velocity and is determined only by the distribution of densities of the components.

To close the hydrodynamic problem (1.6)–(1.10), (1.5), we need material relations, i.e., expressions for the quantities τ_{ab} and Q_{ia} . These expressions should be compatible with inequality (1.15). Of greatest interest is the model that is the minimum generalization of the model of a viscous multispecies mixture [9, 10]. Thus, for the viscous stress tensor, we use the linear-viscous Navier–Stokes model

$$\tau_{ab} = (\mu_v - (2/3)\mu_s)\partial_c v_c \delta_{ab} + \mu_s(\partial_a v_b + \partial_b v_a), \quad (1.16)$$

where μ_v and μ_s are the positive coefficients of volume and shear viscosity, respectively.

For diffuse fluxes, we use relations that generalize the Fick law

$$Q_{ia} = -D_{ij} \partial_a \Phi_j, \quad (1.17)$$

where D_{ij} is a symmetric nonnegative matrix satisfying the additional condition

$$D_{ij} m_j = 0. \quad (1.18)$$

Let us discuss the physical meaning of the tensor of static stresses σ_{ab} . For a homogeneous mixture, this tensor reduces to the usual stress tensor in an ideal liquid $\sigma_{ab} = -p \delta_{ab}$, where $p = (n_i f_{,i} - f)$ is the hydrostatic pressure.

The tensor σ_{ab} can differ from the stress tensor of an ideal liquid in the region where the density gradients reach considerable magnitudes. To find the role of these gradients in the stress tensor, it is convenient to consider the equilibrium two-phase state with the dependence on only one coordinate x^1 . In this case, $n_i = n_i(x^1)$ and $v_a = 0$. As $x^1 \rightarrow \pm\infty$, the density of components and the stresses converge to the densities and stresses in phases *A* and *B*: $x^1 \rightarrow -\infty$, $n_i \rightarrow n_{iA}$, and $\sigma_{ab} \rightarrow -p_A \delta_{ab}$; $x^1 \rightarrow +\infty$, $n_i \rightarrow n_{iB}$, and $\sigma_{ab} \rightarrow -p_B \delta_{ab}$.

As is known [3], the conditions of thermodynamic equilibrium of the state considered yield the condition of mechanical equilibrium: $\partial_1 \sigma_{11} = 0$. From here and from the above-described considerations, it follows that $\sigma_{11} = -p_A = -p_B$ and $\sigma_{22} = \sigma_{33} = \sigma_{11} + \nu_{ij} \partial_1 n_i \partial_1 n_j = -p_A + \nu_{ij} \partial_1 n_i \partial_1 n_j$.

Thus, the density gradients contribute to the transverse components of the stress tensor. They provide emergence of tension in the interphase region. The integral of this distributed tension over the longitudinal coordinate should be identified with the coefficient of interphase surface tension [3]

$$\gamma = \int_{-\infty}^{+\infty} \nu_{ij} \partial_1 n_i \partial_1 n_j dx^1. \quad (1.19)$$

It should be noted that the expression for the tensor of static stresses σ_{ab} is derived from the expression for free energy (1.1) in a standard manner. The expression for the stress tensor in more complicated models of continuous media was derived in [11, 12]. The stress tensor for the model of a single-species liquid with a squared density gradient in free energy was considered in [13].

2. Examples of Numerical Simulations of Two-Phase Flows. The study of capabilities of the density-functional method for numerical simulation of multiphase flows is at the early stage. Some encouraging results were obtained for a number of typical problems; part of them are described below for the case of plane isothermal flows of viscous two-phase two-species media.

The governing system of equations is composed of the equations of conservation of the species (1.6) and the equation of momentum (1.7). We use expression (1.8) for the tensor of static stresses σ_{ab} , (1.16) for the tensor of viscous stresses τ_{ab} , and (1.17) for the diffusion fluxes Q_{ia} .

In using the density-functional method for two-phase media, such as liquid–liquid or gas–liquid systems, one should specify a particular form of the function of free energy and the values of the coefficients of viscosity, diffusion, and surface tension on the contact between the mixture and the solid phase, as well as the coefficients ν_{ij} .

If the deviations of densities from certain fixed equilibrium values are small, the free energy of one phase (phase A) can be presented as the quadratic polynomial

$$f_A(n_i) = f_{A0} + f_{Ai}(n_i - n_{iA}) + (1/2)f_{Aij}(n_i - n_{iA})(n_j - n_{jA}). \quad (2.1)$$

Here, n_{iA} is the undisturbed value of molar density for phase A .

The coefficients f_{A0} and f_{Ai} do not enter the hydrodynamic equations; they are used to calculate the initial undisturbed pressure and chemical potentials. These coefficients can be set to zero. The coefficients important for hydrodynamic simulation are only f_{Aij} , which are chosen in accordance with data on the volume modulus of elasticity for phase A :

$$E_A = f_{Aij} n_{iA} n_{jA}. \quad (2.2)$$

In the case of a two-phase flow (phases A and B), the free energy f is determined via the expressions for $f_A(n_i)$, $f_B(n_i)$ as follows:

$$f = f_A f_B / (f_A + f_B). \quad (2.3)$$

It is assumed that the shear and volume viscosities of each phase are known. Viscosity for an arbitrary density of components is calculated by empirical formulas, which allow one to find the viscosity of the mixture from the known viscosity of the components (see [14, formula (VIII-54)]):

$$\mu_s = \left(c_A \mu_{sA}^{1/3} + c_B \mu_{sB}^{1/3} \right)^3, \quad \mu_v = \left(c_A \mu_{vA}^{1/3} + c_B \mu_{vB}^{1/3} \right)^3. \quad (2.4)$$

Here

$$c_A = z_B / (z_A + z_B); \quad c_B = z_A / (z_A + z_B);$$

$$z_A = \left(\sum_{i=1}^2 (n_i - n_{iA})^2 \right)^{1/2}; \quad z_B = \left(\sum_{i=1}^2 (n_i - n_{iB})^2 \right)^{1/2}.$$

To calculate the matrix D_{ij} , we note that Eq. (1.17) for $\nu_{ij} = 0$ yields the expression for the concentration flux of components

$$q_{ia} = n^{-1} Q_{ia} = -n^{-1} D_{ij} \left(\frac{\partial \mathfrak{x}_j}{\partial c} \right)_n \partial_a c - n^{-1} D_{ij} \left(\frac{\partial \mathfrak{x}_j}{\partial n} \right)_c \partial_a n,$$

where $n = \sum_{i=1}^2 n_i$ is the total density, $c = n_1/n$ is the concentration of component No. 1, and $\varkappa_i = f_{,i}$ is the chemical potential.

Thus, the tabulated diffusion coefficient d of component No. 1 in the mixture is related to the matrix D_{ij} as

$$d = n^{-1} D_{1j} \left(\frac{\partial \varkappa_j}{\partial c} \right)_n. \quad (2.5)$$

The diffusion coefficient d is calculated from the known values in phases d_A and d_B : $d = c_A d_A + c_B d_B$. If the free energy is prescribed [see (2.1)–(2.3)], then, based on the coefficient d , Eq. (2.5) with allowance for (1.18) uniquely determines the matrix D_{ij} . The surface tension on the mixture–solid contact is assumed to be a linear function of the densities of the components:

$$f_* = \xi_{1i} n_i + \xi_0. \quad (2.6)$$

The parameters ξ_0 and ξ_{1i} are calculated from the known values of surface tension on the contact with the solid body for phases A and B :

$$\theta_A = \xi_{1i} n_{iA} + \xi_0, \quad \theta_B = \xi_{1i} n_{iB} + \xi_0. \quad (2.7)$$

The system of linear equations (2.7) always has a solution, but this solution is not unique. The arbitrariness in the choice of dependence (2.6) affects the distribution of components near the solid wall but does not affect the wetting angle.

The matrix of coefficients ν_{ij} is assumed to be constant and proportional to a unit matrix. The unknown coefficient is found from the formula for surface tension between the phases A and B [3] similar to relation (1.19):

$$\gamma = \int_0^{+\infty} \nu_{ij} \partial_r n_i \partial_r n_j dr. \quad (2.8)$$

The integral in formula (2.8) is calculated for the static solution of the problem of a drop of phase A placed into phase B or, vice versa, of a drop of phase B placed into phase A .

Thus, relations (2.1)–(2.8) allow us to fix the model parameters. For all problems considered, we used $\gamma = 0.1$ (hereinafter, all dimensional quantities are given in the SI system).

System (1.6)–(1.10), (1.5), (1.16), (1.17) was solved numerically with the use of an explicit conservative difference scheme based on the concept of the method of coarse particles [15]. In this case, the “Lagrangian” stage completely coincides with [15], and the “Eulerian” stage is constructed under the assumption of the absence of pressure in conservation equations (the terms with the pressure gradient are determined via the derivatives of the free energy f : $p = n_i f_{,i} - f$).

To study the capabilities of the density-functional method, the following model problems were numerically analyzed:

- 1) interaction of a liquid drop (phase A) surrounded by another liquid (phase B) with a plane layer of the liquid (phase A);
- 2) fragmentation of a liquid drop (phase B) by a liquid flow (phase A);
- 3) interaction of a liquid drop (phase A) surrounded by another liquid (phase B) with a flat solid wall;
- 4) development of the Rayleigh–Taylor and Kelvin–Helmholtz instabilities at the interface between two media.

The gray scale in computational cells in Figs. 1–4 shows the distribution of concentration of one of the components present, which actually corresponds to the spatial distribution of the corresponding phase.

Problem 1. At the initial time, the upper half-plane contains a quiescent layer of the liquid phase B , and the lower half-plane contains a layer of the liquid phase A . A drop of the liquid phase A moves from the upper region to the lower region at an angle of 30° to the horizontal line (see Fig. 1a). The initial velocity of the drop is 10.0. The field of mass forces is directed vertically downward. Conditions (1.13) and the absence of diffusion fluxes of components (1.14) are set on the upper and lower boundaries. Periodic conditions are imposed on the left and right boundaries. The initial parameters of the problem are as follows: the computational domain is 60×50 , the cells of the difference grid are squares with a side of 0.001 m, $m_1 = 18$, and $m_2 = 200$; $\mu_{sA} = 10^{-3}$, $\mu_{sB} = 10^{-2}$, $\mu_v = 10\mu_s$; the mass densities of the phases are $\rho_A = 1000$ and $\rho_B = 800$; $E_A = E_B = 10^9$, and $d_A = d_B = 10^{-9}$.

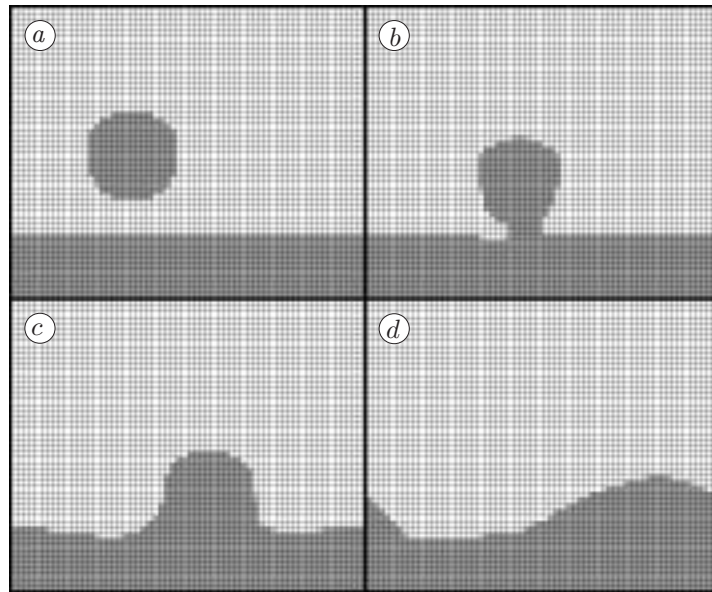


Fig. 1

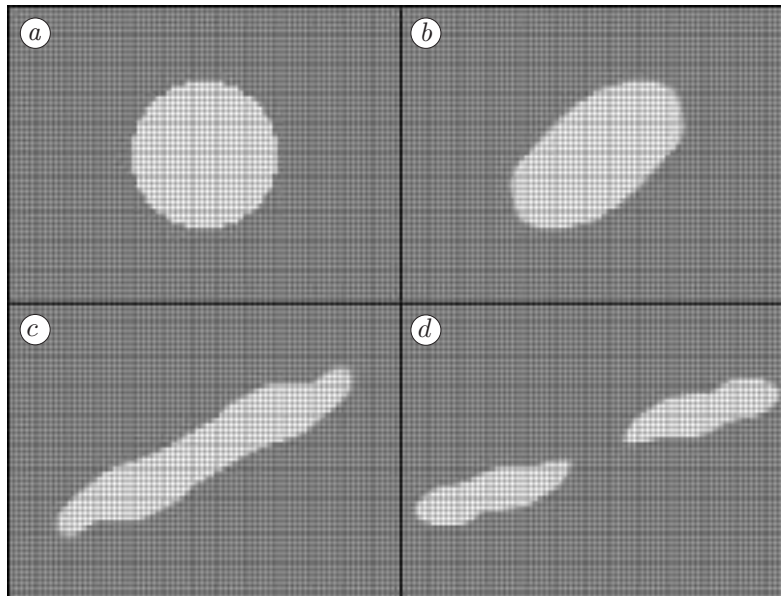


Fig. 2

Figure 1b–d shows the dynamics of the process at consecutive times. The interaction results in the formation of a structure with a thin stem of phase *A* (Fig. 1b) with subsequent “outflow” of the liquid from the drop into the layer (Fig. 1c) subjected to disturbances in the form of gravitational-capillary waves (Fig. 1d).

Problem 2. At the initial time, a drop of the liquid (Phase *B*) is located in a nonuniform flow of another liquid (phase *A*). The latter is the Couette flow formed by the upper and lower boundaries moving with identical velocities $v_0 = 10$ (in their planes) in different directions (the upper boundary moves from left to right, and the lower boundary moves in the opposite direction), with conditions (1.13) and (1.14) imposed on the boundaries (Fig. 2a). There is no mass force. Periodic conditions are imposed on the left and right boundaries. The initial parameters

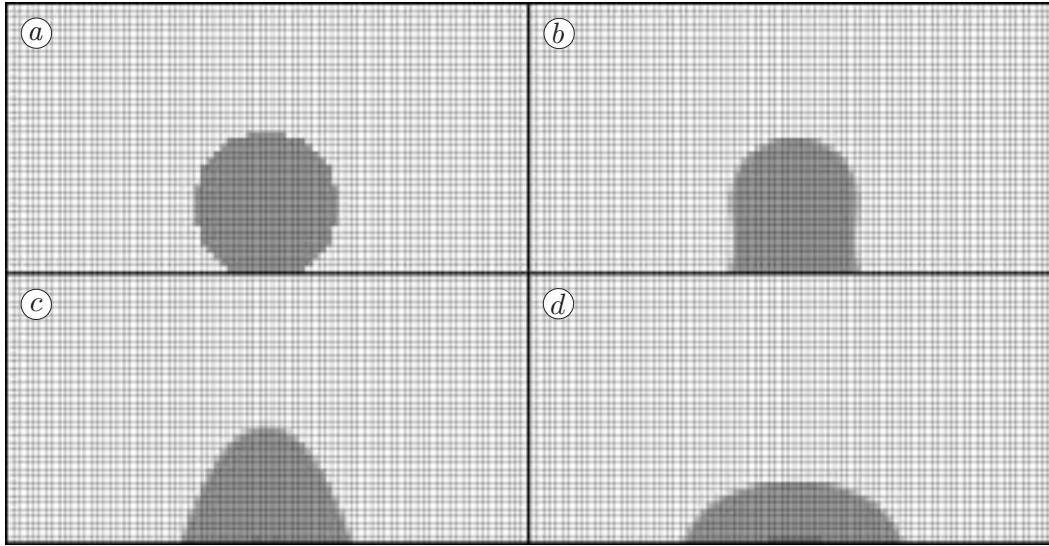


Fig. 3

of the problem are as follows: the computational domain is 80×60 , $\mu_s = 10^{-2}$ for two phases, all the remaining parameters are similar to those in problem 1.

Because of viscous forces, the drop starts to deform (Fig. 2b) and extends downstream. Figure 2c shows a typical thinning of the drop in the central part. Later on, this part is broken, and two drops are formed (Fig. 2d).

Problem 3. At the initial time, a quiescent spherical drop of the liquid phase A surrounded by the liquid phase B touches the lower boundary of the computational domain (Fig. 3a) simulating the solid surface. Conditions (1.9) and (1.10) are set for velocity and diffusion fluxes on the lower boundary, and condition (1.5) is specified for the molar densities of the phases. The function of surface tension at the mixture–solid interface is constructed by formulas (2.6) and (2.7) with $\theta_B - \theta_A = 0.06$. The conditions of free boundaries are imposed on all the remaining boundaries of the computational domain. There is no mass force. The initial parameters of the problem are as follows: the computational domain is 80×40 , the size of the square cells of the difference grid is 10^{-4} , $\mu_s = 10^{-3}$ for phases A and B , all the remaining parameters are similar to those of Problem 1.

Figure 3b–d shows the dynamics of stabilization of the drop shape on the solid surface with formation of a sharp wetting angle.

Problem 4. At the initial time (Fig. 4a), the upper half-plane contains the gas phase A moving from right to left with a velocity v_0 , and the lower half-plane contains the liquid phase B moving in the opposite direction with the same velocity. The mass force is directed perpendicular to the interface toward the lighter gas phase. This is the Rayleigh–Taylor instability case [16]. In the vicinity of the interface, initial disturbances of velocities are set in the form

$$v_x = \text{sign}(y)A \sin(kx) e^{-k|y|}, \quad v_y = A \cos(kx) e^{-k|y|}.$$

Here k is the wavenumber, A is the amplitude of disturbances, x is the streamwise coordinate, and y is the transverse coordinate counted from the interface. The boundary conditions $v_x = v_0$ and $v_y = 0$ are imposed on the left boundary for $y < 0$, the conditions $v_x = -v_0$ and $v_y = 0$ are set on the right boundary for $y \geq 0$, and the free-boundary conditions are set on all remaining boundaries of the computational domain. The initial parameters of the problem are as follows: the computational domain is 200×50 , the size of the square cells of the difference grid is $4 \cdot 10^{-4}$, $m_1 = 2$, $m_2 = 200$, $\mu_s = 10^{-5}$ for phase A and $\mu_s = 10^{-3}$ for phase B , $\mu_v = 10\mu_s$, $\rho_A = 2$, $\rho_B = 800$, $E_A = 10^7$, $E_B = 10^9$, $d_A = 10^{-6}$, $d_B = 10^{-9}$, $v_0 = 5$, $A = 1$, and $k = 314$.

It follows from the analysis of the dispersion equation for the chosen parameters [16] that the Rayleigh–Taylor and Kelvin–Helmholtz instabilities are developed near the interface, which first leads to an insignificant bending of the interface (Fig. 4b). Then, this process transforms to the nonlinear stage with strong deformation of the interface (Fig. 4c) and to formation of individual gas bubbles (Fig. 4d).

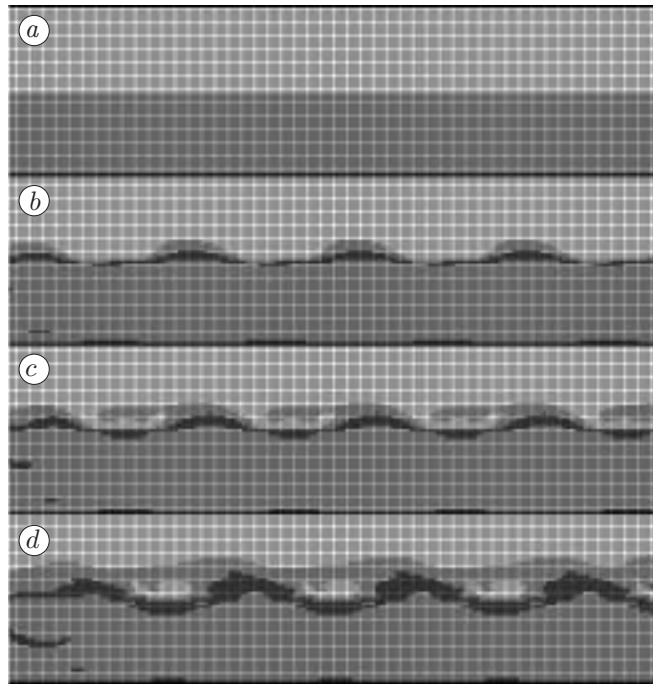


Fig. 4

Conclusions. The numerical solutions obtained correspond to advanced theoretical concepts and experimental data on dynamics of drops, wetting phenomena, and wave processes at the interface of liquid phases [17–20]. Therefore, we can state that the density-functional method can rather effectively simulate the dynamics of two-phase mixtures in the isothermal case. The numerical scheme of through count proposed is economical and can be easily extended to mixtures with a large number of components or a large number of phases, and also to spatial cases.

Concerning the accuracy of the method, we should distinguish between the accuracy of computations in the single-phase region and the accuracy of computations in the interface region. In the single-phase region, the contribution of higher derivatives to hydrodynamic equations is negligibly small, and the model actually reduces to the usual model of a multispecies viscous liquid or gas. Correspondingly, the accuracy of computations is little different from the accuracy of commercial software systems for single-phase viscous mixtures. Concerning the interface region, the accuracy here depends significantly on the cell size of the computational grid. In numerical simulations, the size of the phase-transition region is normally 3–5 cells. Thus, the accuracy of prediction of spatial evolution of the interface is proportional to the characteristic cell size. Currently, the numerical program developed allows computations with the number of cells of approximately 10^5 on a personal computer of average performance. In our opinion, this provides sufficient accuracy for describing the dynamics of small drops or multiphase multispecies mixtures in capillaries and porous materials, i.e., in problems of microhydrodynamics with typical dimensions of $1\ \mu\text{m}$ to $1\ \text{mm}$.

As for hydrodynamic problems such as multiphase flows in tubes or other engineering devices, quite a large number of grid cells are needed to reach adequate accuracy. This problem can be solved by improving the numerical scheme or owing to an increase in computer performance.

For nonisothermal problems, the theory has been well developed [4], but numerical implementation of the corresponding system of equations requires additional research.

Visualization of results was performed with the help of the software system VR-Geo (Joint-Stock Company “Servis-Nafta”).

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