

# SOLIDIFICATION OF EMULSION MOVING UNDER EFFECT OF THERMOCAPILLARY FORCES AND MICROACCELERATION

© A.G. PETROVA, V.V. PUKHNACHOV, E.N. ZHURAVLEVA

Russia

**1. Statement of a problem.** The system of governing equations for the motion of emulsion, offered by V. Pukhnachov and O. Voinov in 1995 ([1]), involves the following unknown functions: temperature of mixture  $T$ , volume concentration of the disperse phase  $c$ , average velocities of drops in carrying phase, pressure of carrying phase. It consists of mass conservation laws for disperse and carrying phases, momentum equation, energy equation and the equation of relative phase velocity. The described system is very complicate due to its nonlinearity and high order. However, in the case of one-dimensional motion with plane, cylindrical or spherical waves it can be radically simplified. One-dimensional with plane waves motion of emulsion under the effect of thermocapillary forces and microacceleration with the average volume velocity of mixture equal to zero is described by the following mathematical model with two unknown functions: temperature  $T$  and concentration of disperse phase  $C$

$$\begin{aligned} & \frac{\partial}{\partial t} C + \frac{\partial}{\partial x} \left( C(1-C) \left( L \frac{\partial T}{\partial x} + Kg \right) \right) = 0, \\ & \rho_d \lambda_d C \left[ \frac{\partial T}{\partial t} + (1-C) \left( L \frac{\partial T}{\partial x} + Kg \right) \frac{\partial T}{\partial x} \right] + \rho_m \lambda_m (1-C) \times \\ & \times \left[ \frac{\partial T}{\partial t} - C \left( L \frac{\partial T}{\partial x} + Kg \right) \frac{\partial T}{\partial x} \right] = k_m \frac{\partial}{\partial x} \left( (1-MC) \frac{\partial T}{\partial x} \right), \end{aligned} \quad (1.1)$$

We use indexes  $d$  and  $m$  to denote the parameters of the disperse and the matrix respectively Let us introduce the following notations:

$$\begin{aligned} M &= 3(k_m - k_d)/(2k_m + k_d), \\ K &= 2R^2(\rho_d - \rho_m)(\mu_d + \mu_m)/(3\mu_m(2\mu_m + 3\mu_d)), \\ L &= 2Rk_m\sigma\mu_m/((2\mu_m + 3\mu_d)(2k_m + k_d)), \end{aligned}$$

---

This work was supported by grants of INTAS, N94-529 and Russian Foundation of Basic Research N99-01-00529.

where  $R$  is the radius of the drops,  $\sigma$  is the surface tension coefficient. Here  $\rho_m^l, \rho_m^s, \rho_d^l, \rho_d^s, \lambda_m^l, \lambda_m^s, \lambda_d^l, \lambda_d^s, k_m^l, k_m^s$  — are known positive constants. The constant  $g$  is positive, if the acceleration of force of weight is directed to the side of greater temperature and is negative otherwise.

The velocities and the pressure are restored from the additional equations after the temperature and concentration are found.

We consider the process of solidification under the assumption that the solid matrix is motionless. A jump of density at solidification is neglected. In the liquid matrix with solidified drops the effect of thermocapillary forces is absent ( $L = 0$ ), in solidified matrix both the thermocapillary forces and the floating force are absent ( $L = Kg = 0$ ). The conditions on the line of discontinuity have the form:

$$[C]D = [C(Kg + LT_x)](1 - C), \quad [T] = 0,$$

$$[U]D = [(\rho_d \lambda_d - \rho_m \lambda_m)C(1 - C)(Kg + LT_x)T] - [k_m(1 - MC)T_x], \quad (1.2)$$

Here the symbol  $[.]$  denotes the difference of values of the function in front and behind the jump,  $D$  is the velocity jump,  $U$  stands for the enthalpy.

ADDITIONAL ASSUMPTIONS. The supposition of smallness of the disperse substance concentration gives a reason for linearization as the main method of research of the problem in the work under study. Since the process of the thermal conduction in the first approximation is determined by the parameters of the matrix, we assume that:

- the solidification front  $x = s_m(t)$  for the matrix is the Stefan boundary for the temperature problem;
- the isotherm  $T = T^d$ , where  $T^d$  is the temperature of the disperse substance solidification can be the line of discontinuity only for the concentration of the disperse substance; let  $x = s_d(t)$  be the equation of this isotherm;
- the temperature gradient is greater than zero.

Let the temperature  $T^m$  of the matrix solidification be less than the temperature  $T^d$  of the disperse substance solidification. We denote as  $C^s(x, t)$  the solid disperse substance concentration in the solid matrix (the domain  $x < s_m(t)$ ); as  $C^{sl}(x, t)$  - the solid disperse concentration in the liquid matrix (the domain  $s_m < x < s_d$ ); as  $C^l$  - the liquid disperse concentration in the liquid matrix (the domain  $x > s_d(t)$ ). So the linearized problem of determining the functions  $C^{sl}(x, t), C^l(x, t), C^s(x, t)$ , will be the following:

$$\frac{\partial}{\partial t} C^l + \frac{\partial}{\partial x} \left( C^l \left( L \frac{\partial T^l}{\partial x} + Kg \right) \right) = 0, \quad x > s_d(t);$$

$$\frac{\partial}{\partial t} C^{sl} + \frac{\partial}{\partial x} \left( C^{sl} Kg \right) = 0, \quad s_m(t) < x < s_d(t);$$

$$\frac{\partial}{\partial t} C^s = 0, \quad x < s_m(t);$$

$$\frac{ds_d}{dt} \left( C^l - C^{sl} \right) = C^l \left( L \frac{\partial T^l}{\partial x} + Kg \right) - C^{sl} Kg, \quad x = s_d(t);$$

$$\frac{ds_d}{dt} \left( C^{sl} - C^s \right) = C^{sl} K g, \quad x = s_m(t). \quad (1.3)$$

The temperature  $T^l(x, t)$  and the free boundary  $s_m(t)$  are preliminary found from the classic two-phase Stefan problem:

$$\begin{aligned} \rho_m^l \lambda_m^l \frac{\partial T^l}{\partial t} &= k_m^l \frac{\partial^2 T^l}{\partial x^2}, & x > s_m(t); \\ \rho_m^s \lambda_m^s \frac{\partial T^s}{\partial t} &= k_m^s \frac{\partial^2 T^s}{\partial x^2}, & x < s_m(t); \\ T^s = T^l = T^m, & \quad \frac{ds}{dt} \gamma = k_m^s \frac{\partial T^s}{\partial x} - k_m^l \frac{\partial T^l}{\partial x}, & x = s_m(t), \end{aligned} \quad (1.4)$$

with some boundary and unitial conditions, for example,

$$\begin{aligned} T^s(0, t) &= f^s(t) < T^m, \quad T_x^l(\infty, t) = 0, \quad s(0) = s_0; \\ T^s(x, 0) &= \varphi^s(x) < T^m, \quad x \in (0, s(t)); \\ T^l(x, 0) &= \varphi^l(x) > T^m, \quad x \in (s(t), \infty); \end{aligned} \quad (1.5)$$

Let us suppose now that  $T^m > T^d$ . In this case the concentration does not have the jump on the line  $x = s_d(t)$ . So the problems of finding the disperse concentration in the solid matrix  $C^s(x, t)$  and the disperse concentration in the liquid matrix  $C^l(x, t)$  consist of equations:

$$\frac{\partial}{\partial t} C^l + \frac{\partial}{\partial x} \left( C^l \left( L \frac{\partial T^l}{\partial x} + K g \right) \right) = 0, \quad x > s_m(t); \quad (1.6)$$

$$\frac{\partial}{\partial t} C^s = 0, \quad x < s_m(t); \quad (1.7)$$

and condition

$$\frac{ds_m}{dt} \left( C^l - C^s \right) = C^l \left( L \frac{\partial T^l}{\partial x} + K g \right), \quad x = s_m(t). \quad (1.8)$$

Functions  $T^l(x, t)$  and  $s_m(t)$  are the solution of the classic Stefan problem, mentioned above.

For the described systems we consider two problems: the problem of the concentration distribution determination in the solid part under given initial distribution of concentration in liquid, which we name "direct" and the problem of initial concentration distribution determination in liquid under given distribution of the concentration in the solid part which we name "inverse". The latter is the mathematical model for the problem of obtaining the composite with the given disperse phase distribution when the difference in densities is essential, that is impossible in the case of earth gravitation due to the floating effect. We will search for a classical solution of delivered problems with nonnegative, bounded functions  $C^s, C^{sl}, C^l$ .

**2. Conditions of admixture displacement by moving front of solidification.** We will be interested in the process "of directed solidification", i.e. solution of problem (1.4) - (1.5), satisfying the inequality  $\dot{s}_m(t) > 0$ . This requirement can be easily provided in terms of the Stefan problem input data ([2]).

Let us consider the relation of the disperse phase concentration on different sides of solidification boundary in the case  $T^m > T^d$ . Taking into account the condition of a gap for concentration (1.8), we conclude, that the necessary condition for solvability of direct and inverse problems has the form

$$\frac{ds_m}{dt} \geq L \frac{\partial T^l}{\partial x}(s_m(t), t) + Kg, \quad t > 0. \quad (2.1)$$

It provides the "right" slope of the characteristics and nonnegative sign of the concentrations on the solidification boundary. Inequality (2.1) means, that the disperse transport velocity in liquid phase, caused by thermocapillary effect and microgravitation, not greater than the velocity of the solidification. Under this condition in the case  $LT_x^l(s_m(t), t) + Kg \geq 0$  (Fig. 1,a) we have  $C^s(x) \leq C^l(x, s_m^{-1}(x))$ . In the case  $LT_x^l(s_m(t), t) + Kg < 0$  (Fig. 1,b) we obtain  $C^s(x) > C^l(x, s_m^{-1}(x))$ . In the case of realization of the identical equality  $\dot{s}_m(t) = LT_x^l(s_m(t), t) + Kg$ , the direct problem can have only trivial solution, the inverse one has no sense.

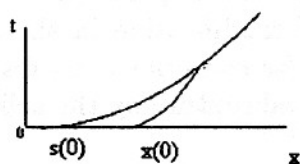


Fig 1 a

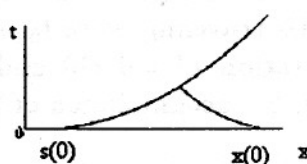


Fig 1 b

Note, that the conditions of the "right" slope for characteristics of two-front problem (1.3) are as follows:

$$\frac{ds_m}{dt} \geq Kg, \quad \left( \frac{ds_d}{dt} - LT_x^l(s_d(t), t) - Kg \right) \cdot \left( \frac{ds_d}{dt} - Kg \right) \geq 0.$$

**3. On resolvability of the problems.** In this section we formulate the sufficient conditions for existence of the bounded and nonnegative solution for the direct and inverse problems at any time interval ([4]).

**PROPOSITION 3.1.** *Let inequality*

$$-A \frac{ds_m}{dt}(t) \leq L \frac{\partial T^l}{\partial x}(s_m(t), t) + Kg \leq a \frac{ds_m}{dt}(t) \quad (3.1)$$

*is fulfilled for all significance of time t and for some constants A and a, such, that  $A \geq 0$ ,  $0 \leq a < 1$ . Besides that, let exists nonnegative constant N, such, that for all*

significance of  $t$ , inequality

$$\left| \int_0^t \left( L \frac{\partial T^l}{\partial x}(s_m(\tau), \tau) + Kg \right) \frac{\partial T^l}{\partial x}(s_m(t)(1-a) + s_m(\tau)a, \tau) d\tau \right| \leq N$$

is fulfilled.

Then the solution  $C^s(x)$  of problem (1.5)–(1.8) with the initial condition  $C^l(x, 0) = C_0^l(x) > 0$ ,  $x > s_m(0)$  is nonnegative and bounded by the value  $\max C_0^l(x) \exp\{Lk_m^{-1}((\varphi^l(x) - T^m)\rho_m^l \lambda_m^l)\}(1 + A)$ .

**PROPOSITION 3.2.** Let condition (3.1) fulfilled. Then the solution of problem (1.5)–(1.8) with the condition  $C^s(x, s_m^{-1}(x)) = C_0^s(x)$  is nonnegative and bounded by  $\max C_0^s(x) \exp\{Lk_m^{-1}N\rho_m^l \lambda_m^l\}/(1 - a)$ .

Note, that, the qualitative picture of the behavior of the solution for the problem (1.6) - (1.8) is rather well represented by the cases, when the functions  $T^l$  and  $T^s$  are "travelling wave" or self-similar ( $g = 0$ ) solutions of appropriate Stefan problem.

**4. Some special solutions.** The exact solutions for problem (1.4), (1.6)–(1.8) with the special conditions of solidification (traveling wave with the solidification front equation  $s(t) = Vt$ , and self-similar with  $s(t) = \beta\sqrt{t}$ ,  $g = 0$ ) are obtained and studied in [4].

Fig.2 shows the graph of the exact solution for the disperse phase concentration of the emulsion Al-Pb with traveling wave type of solidification in the moment  $t = 2c$  with the initial concentration  $C = 0.006$  under the conditions: a - displacement of the admixture to the liquid, b - accumulation of the admixture by the solidified part.

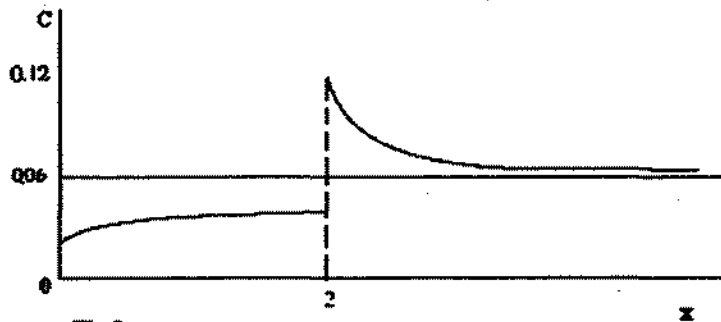


Fig 2 a

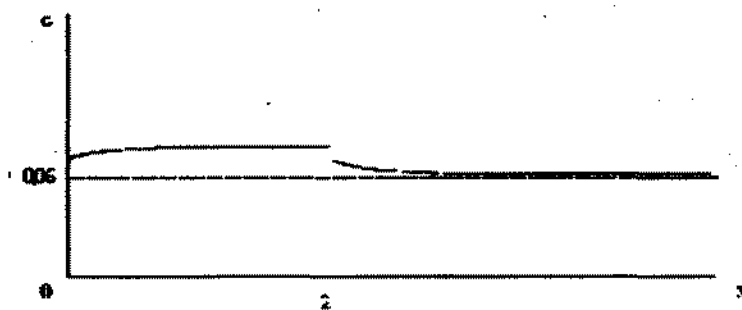


Fig 2 b

The exact solution for two-front problem is also obtained ([4]). It should be noticed that the presence of the second line of the concentration discontinuity and the intermediate layer, representing the liquid matrix with solid inclusions, does not render essential influence to the qualitative picture of the solution behavior in the liquid emulsion and in the completely solidified part.

**5. Spherical symmetry problem of solidification of emulsion.** In the case of the spherical symmetry (solidification of the sphere with unit radius,  $g = 0$ ) equations (1.6)-(1.8) are replaced by

$$\begin{aligned} \frac{\partial}{\partial t} C^l + \frac{1}{x^2} \frac{\partial}{\partial x} \left( x^2 C^l L \frac{\partial T^l}{\partial x} \right) &= 0, & x < s_m(t); \\ \frac{\partial}{\partial t} C^s &= 0, & s_m(t) < x < 1, \\ \frac{ds_m}{dt} (C^l - C^s) &= C^l \left( L \frac{\partial T^l}{\partial x} + Kg \right), & x = s_m(t). \end{aligned}$$

Temperature  $T^l(x, t)$  and free boundary  $s_m(t)$  are preliminary found by solving the spherical symmetry two-phase Stefan problem. Note, that in this case  $\dot{s}_m(t) < 0$ . This requirement can be easily provided in terms of initial and boundary conditions for Stefan problem.

The condition of the "right" slope of the characteristics and nonnegative sign of the concentrations on the solidification boundary takes the form

$$\dot{s}_m(t) \leq LT_x^l(s_m(t), t).$$

Note, that in the absence of gravitation the concentration of disperse on the front from the liquid part is always greater than from the solid.

The problem accepts the exact solution

$$\begin{aligned} s(t) &= \beta \sqrt{t^* - t}, & T^l(x, t) &= b \left( t^* - t - \frac{x^2}{6a_l} \right), \\ T^s(x, t) &= b \left( t^* - t - \frac{x^2}{6a_l} \right) - \frac{\beta^3}{2} \gamma \cdot \exp \left( \frac{-\beta^2}{4a_s} \right) \cdot \int_{\beta}^{\frac{x}{\sqrt{t^* - t}}} \frac{\exp(x^2/(4a_s))}{z^2} dz, \end{aligned}$$

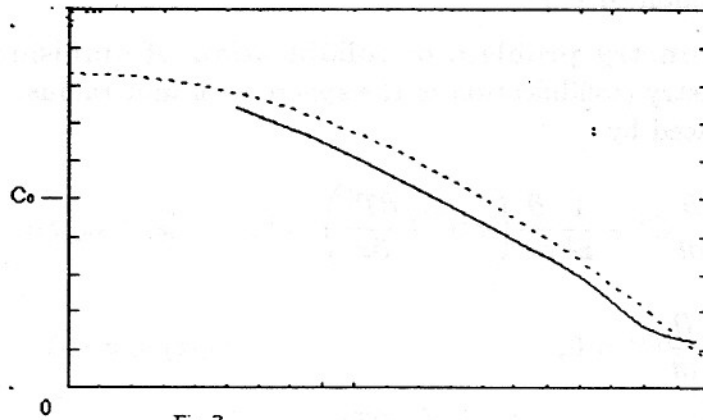
where  $b$  is an arbitrary positive constant,  $a_l, a_s$  are the thermal conductivity of liquid and solid;  $\beta = \sqrt{6a_l}$ ,  $t^* = 1/6a_l$  is the time of full solidification of the sphere;

$$C^l(x, t) = \exp(bLt/a_l) C_0^l \left( x \cdot \exp(bLt/3a_l) \right), \quad C_0^l = C^l(x, 0).$$

$$C^s(y(t), t) = C_0^s \exp(bLt/a_l) \cdot \left( 1 - \frac{bL}{9a_l^2} + \frac{2bLt}{3a_l} \right).$$



Fig.3 demonstrates the distribution of the concentration in the solidified part, dotted line represents the exact solution, continuous line represents the numerical solution under the input data from the exact solution, given above. The error is about 0.01. Redistribution of the disperse can be estimated as 10% of initial concentration.



Calculations were conducted under different initial and boundary regimes for the Stefan problem and showed that the degree of redistribution increased together with the increasing of the sphere surface cooling velocity.

The corresponding problem can be easily formulated and the exact solution found for the case of one-dimensional motion with cylindrical waves.

**6. Numerical study.** The numerical research of the linearized problem consists of consecutively solving the Stefan problem and the transport equation. For the solution of the two-phase Stefan problem, the Riccati transformation is used, which allows to calculate the free boundary without additional iterations. Moreover, it permits to solve the modified Stefan problem with surface tension and supercooling conditions on the solidification front. This method was offered by Gunter Meyer ([3]). In the heat equation, the time derivative is replaced with backward finite difference, and the temperature for time step number  $n$  is found in the form

$$T^n(x) = R(x) \frac{\partial T^n}{\partial x} + W(x)$$

or

$$\frac{\partial T^n}{\partial x} = R(x) T^n(x) + W(x) \quad (6.1)$$

for the boundary conditions of the first and the second order correspondingly. Functions  $R(x)$ ,  $W(x)$  are found as the solutions of the boundary problems for the Riccati and linear equations correspondingly. The free boundary position for the time step  $n$  is found from the algebraic equation obtained from the Stefan condition, where the time derivative is replaced with backward finite difference and the temperature gradients are taken from (6.1). The temperature is restored from solving the ordinary differential equation with given conditions on the free boundary, which position was found before.

For the numerical study of the direct problem, the implicit scheme with directed differences is used. Space and time steps were chosen such that the inequalities

$$\left| L \frac{\partial T}{\partial x} + Kg \right| < \frac{\Delta x}{\Delta t} < \left| \frac{ds}{dt} \right|$$

would be fulfilled. In dependence on the signs of the expression  $LT_x + Kg$  the value of the liquid phase concentration  $C_i^n$  for the time step  $n$  and the space step  $i$  is determined from equation

$$C_i^{n+1} = C_i^n - \frac{\Delta t}{\Delta x} \left\{ \left( L \frac{\partial T_{i+1}^n}{\partial x} + Kg \right) C_{i+1}^n - \left( L \frac{\partial T_i^n}{\partial x} + Kg \right) C_i^n \right\}$$

if the expression is negative, or from equation

$$C_i^{n+1} = C_i^n - \frac{\Delta t}{\Delta x} \left\{ \left( L \frac{\partial T_i^n}{\partial x} + Kg \right) C_i^n - \left( L \frac{\partial T_{i-1}^n}{\partial x} + Kg \right) C_{i-1}^n \right\}$$

if the expression is positive. Fixed mesh with uniform time and space steps was used for calculations. The values on the free boundary in the case of difference in the free boundary position and the mesh points are calculated by using the Newton interpolation polynomial of the 4-th order. The boundary value problems for the first order ordinary equations are solved with the Runge-Kutt method of the 4-th order.

The calculations were carried out on the example of the mixture aluminum - lead. As the test for one-dimensional problem the exact solution of the emulsion solidification with temperature mode such as a traveling wave was used. The calculations were carried out for the different values of  $Kg$ . Fig.4a shows the profiles of concentration for  $Kg = 0$ , Fig.4b - for  $Kg = -0.003$ .

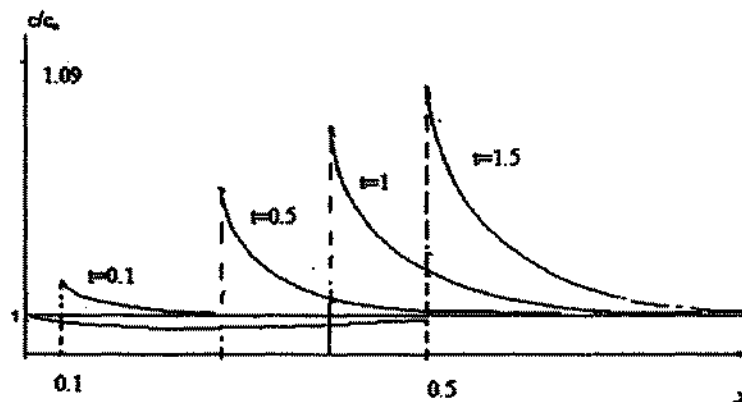


Fig.4 a



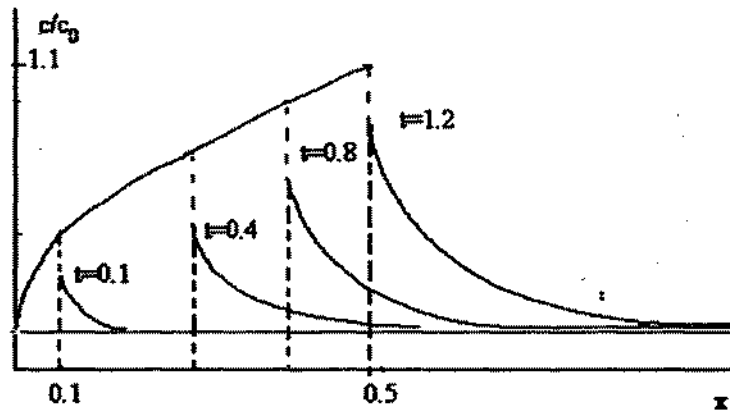


Fig 4b

#### REFERENCES

1. Pukhnachov V.V., Voinov O.V., *Mathematical model of motion of emulsion under effect of thermocapillary forces and microacceleration*, Abstracts of Third Siberian Congress on Applied and Industrial Mathematics (Berlin - 1995), 32-33.
2. Petrova A.G., *Monotonicity of the free boundary in two-phase Stefan problem*, Dynamics of the continuous medium, Novosibirsk 67 (1984), 97-99. (Russian)
3. Meyer G.H., *On computing free boundaries which are not level sets*, Proc. of the 1987 IRSEE Conference.
4. Petrova A.G., Pukhnachov V.V., *One-dimensional motion of emulsion with solidification*, J. of Appl. Mech. and Tech. Phys. 40 (1999), no. 3, 128-136.

ALTAI STATE UNIVERSITY, BARNAUL, RUSSIA

LAVRENTYEV INSTITUTE OF HYDRODYNAMICS, NOVOSIBIRSK, RUSSIA

ALTAI STATE UNIVERSITY, BARNAUL, RUSSIA