

SPHERICAL TWO-PHASE INTERFACE IN A NEAR-CRITICAL FLUID. GRADIENT APPROACH

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Summary On macro-scales the liquid-gas interface is classically modelled by a surface of discontinuity. However, on micro-scales, there is a transition region, where the properties are changed continuously. This makes reasonable to describe the two-phase system, including the interface, within a hydrodynamic approach. In this approach, the new term proportional to density gradient is added to free energy. Near the critical point, where there is no strong difference between a liquid and its vapour, the gradient approach is quite reasonable. Based on this approach we considered the formation of nucleus in a closed cavity and studied its stability. The results are found to be in a good agreement with the classical Laplace approach, while considering the states far from the critical point. In near-critical region, the results are proved surprising. It is shown that within the framework of one-dimensional problem the system remains single-phase and uniform even for negative values of mechanical compressibility.

STATEMENT OF THE PROBLEM

We applied the gradient theory [1] for consideration of nucleus formation in a closed cavity, considering a spherical inclusion in a spherical container. The free energy of fluid in this approach is supposed to be a function of density gradient,

$$f = f^0(\mathbf{r}) + I \frac{(\nabla \mathbf{r})^2}{2}$$

where f is volumetric free energy, f^0 is classical part, and I is capillary coefficient.

The free energy of a spherical nucleus is then as follows

$$F = 4\pi \int_0^{a_0} \left[f_0 + \frac{I}{2} (\mathbf{r}')^2 + k \mathbf{r} \right] r^2 dr,$$

where k is Lagrangian multiplier responsible for condition that system is closed.

In equilibrium the functional of free energy should be minimal. Minimum of free energy is determined using the gradient method. The following equation is postulated:

$$\frac{\partial \mathbf{r}}{\partial t} = - \frac{dF}{d\mathbf{r}}$$

Here t is not real physical time, but just an auxiliary variable. However, the steady solutions of this equation correspond to the extremum of functional F .

This results in the following problem for density profile:

$$\begin{aligned} \frac{\partial \mathbf{r}}{\partial t} &= \frac{1}{a_0^2} \left(\mathbf{r}'' + \frac{2}{r} \mathbf{r}' \right) - \mathbf{r}^3 + \mathbf{r} - q - 3 \int_0^1 \mathbf{r}^3 x^2 dx, \\ \mathbf{r}'|_0 &= \mathbf{r}'|_1 = 0 \end{aligned}$$

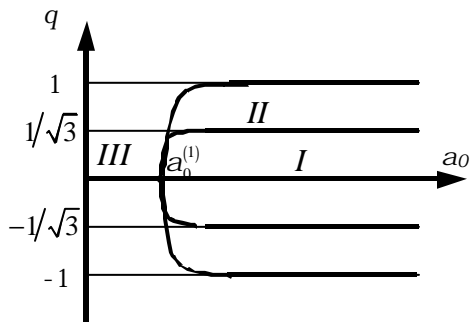


Fig. 1. Regions of existence for different types of solutions

uniform solutions can also realise in this region.

Isothermal compressibility determined by expression $(\partial p / \partial \mathbf{r})_T = -1 + 3q^2$ is negative inside region III and a physical system cannot exist here in a uniform state [2]. Introduction of capillary term into free energy results in surprising effect, in a sufficiently

We use here the following units of length and density: $L = \bar{a}_0$ (container radius), $[\mathbf{r}] = (a/b)^{-1/2}$; $a = -T (\partial a_T^2 / \partial T) > 0$,

$b = 1/6 (\partial^2 a_T^2 / \partial \mathbf{r}^2) > 0$. Two non-dimensional parameters appeared, $a_0 = \bar{a}_0 \sqrt{I \mathbf{r}_c / a}$ is dimensionless radius of a container, and $q = m / ([\mathbf{r}] L^3)$ is average density of a system.

Uniform solution of the problem is trivial $\mathbf{r} = q$. In region I (fig. 1)

$(|q| < \sqrt{3^{-1} \left(1 - (a_0^{(1)} / a_0)^2 \right)})$, where $a_0^{(1)} = 4.49$), these solutions are

unstable with respect to small disturbances, and develop to non-uniform (nucleus-type) solutions. Out of this region (in region II), uniform solutions

are stable with respect to small disturbances, however, they unstable in respect to finite perturbations. As a result, non-uniform

small container the system can be uniform even if $(\partial p/\partial r)_T < 0$. This critical radius is very small far from the critical point and cannot be considered within a hydrodynamic approach. Near the critical point this radius can be not small since $\bar{a}_0^{(i)} \sim 1/\sqrt{(-T)}$, i.e., diverge if $T \rightarrow 0$.

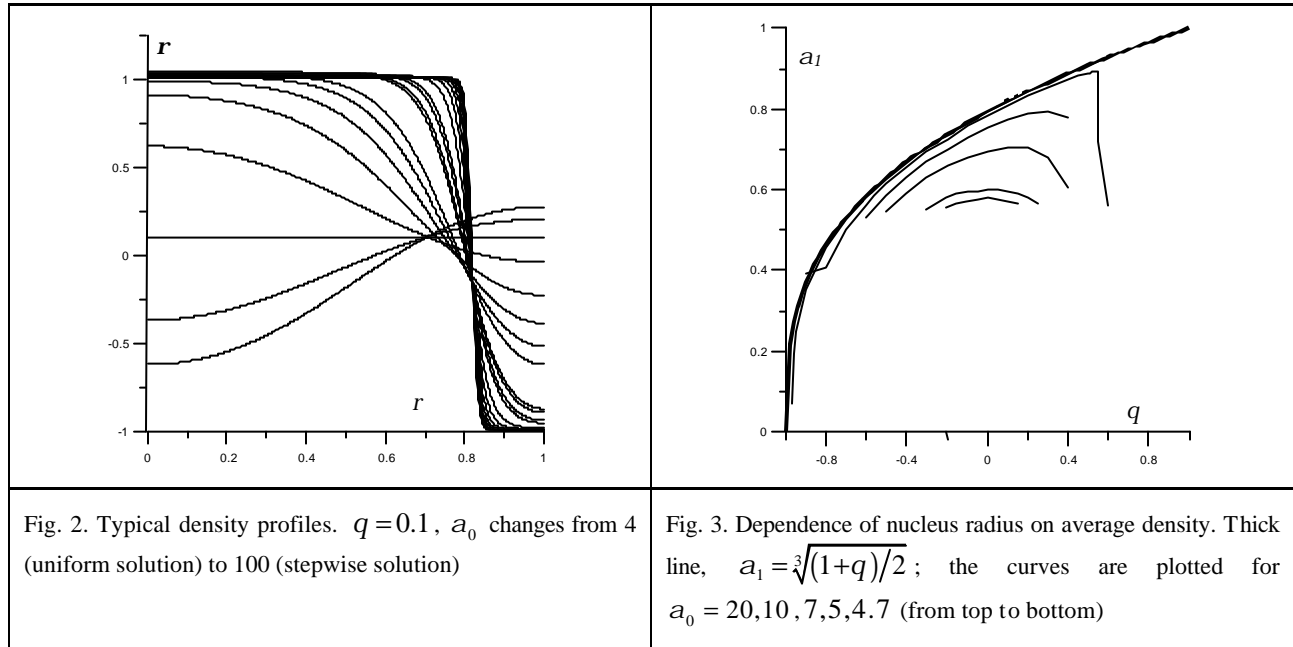
NUMERICAL RESULTS

The typical density profiles obtained by finite-difference method are given in fig. 2. The thickness of a transitional layer, the so-called capillary length, equals $d_l = 1/a_0 = 1/\bar{a}_0 \cdot (\mathbf{I}r_c/a)^{1/2}$.

Radius of nucleus and coefficient of surface tension were calculated with the help of the following effective formulas:

$$a_1^3 = 3 \int_0^1 \frac{r - r(1)}{r(0) - r(1)} r^2 dr, \quad s = \frac{1}{a_0 a_1^2} \int_0^1 \left(\frac{\partial r}{\partial r} \right)^2 r^2 dr.$$

Dependences of nucleus radius on average density are given in fig. 3.



CONCLUSION

The gradient approach is convenient to study the fluid behaviour, since the same system of equations is sufficient to find the flows throughout the two-phase region; the phase transitions, shape and evolution of phase interface are solutions of this system [3]. This description is particularly worth near the critical point where the gradient approach is more reasonable than the classical Laplace theory.

The problem considered was just a preliminary stage of the investigation with the objective in studying the hydrodynamics of near critical two-phase media.

The results obtained are in a good agreement with the classical approach for the states far from the critical point, and for large containers and nuclei. In near critical region the results are intriguing; system can be uniform even for negative values of the derivative $(\partial p/\partial r)_T$, which is absolutely impossible within the classical approach.

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References

- [1] Gouin H. Utilization of the second gradient theory in continuum mechanics to study the motion and thermodynamics of liquid-vapor interfaces // From Physicochemical Hydrodynamics, Interfacial phenomena, p.p. 667-682, Plenum Publishing Corporation, 1987.
- [2] Landau L.D., Lifshiz È. Theoretical Physics. vol. V. Statistical Physics. - M.: Nauka, 1964.
- [3] Jamet D. et al. The second gradient method for the direct numerical simulation of liquid-vapor flows with phase change // J. of Computational Physics 169, 624-651, 2001.