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Influence of homogeneous nucleation on vapor parameters near the evaporation surface: a simplified approach

© V.Yu. Levashov[1], V.O. Mayorov[1], A.P. Kryukov[2]

[1] Institute of Mechanics of Lomonosov Moscow State University, Moscow, Russia
[2] National Research University „MPEI", Moscow, Russia
E-mail: vyl69@mail.ru, levashovvy@imec.msu.ru

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An approach that makes it possible to assess the need to take into account / neglect the process of homogeneous nucleation on the flow of vapor formed near the interface as a result of evaporation is presented. It is shown that estimations require information about the value of the surface tension coefficient of the critical size droplets. The influence of the homogeneous nucleation process on the evaporation of various substances: water, ethanol, and methanol is considered. The results show that for water, consideration of the process of homogeneous nucleation is necessary only for supersaturation ratio greater than 20, in contrast to the evaporation of methanol, when homogeneous nucleation must be taken into account at any supersaturation ratio

Keywords: homogeneous nucleation, evaporation, supersaturation degree.

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Condensation in the gas phase plays an important role both in natural phenomena and in various technological processes [1]. As noted in [2], the range of phenomena described by the condensation problem is enormous. The theory of the formation and growth of nuclei is becoming more and more relevant with the development of materials science, nanotechnology, and microelectronics. It was shown in the paper [3] that as the intensity of evaporation increases, the resulting vapor becomes more and more supersaturated. Even at low evaporation rates, vapor supersaturation ratio (i.e., the ratio of the partial vapor pressure to its saturation pressure) near the phase interface is greater than unity, and increases sharply with process intensity increasing. Under these conditions the process of homogeneous vapor condensation can be realized [4]. As examples of the drops formation over an evaporating surface, one can note the data of papers [5,6], in which the formation of a drop structure over the surface of evaporating liquid, called „drop cluster", was observed.

It is known that the intensity of the evaporation process is determined by the temperature of the evaporating surface and the environment parameters near this surface (temperature, relative humidity, the presence of non-condensable gases) [7]. The drops formed as a result of homogeneous condensation can lead to the environment parameters change in the immediate vicinity of the evaporation surface, which in turn can lead to the intensity change of the evaporation process.

The paper presents the approach that allows, based on simple relations of classical homogeneous nucleation theory to make preliminary estimates of the need to take into account / to neglect the effect of the nucleation process on the evaporation rate.

A one-dimensional stationary problem of the liquid evaporation into a semi-infinite space is considered, it is assumed that part of the supersaturated vapor condenses as it moves from x_1 to x_2. The diagram of the problem is shown in Fig. 1. The flow of vapor „released" from the condensation zone is linked to j_1 and j_{l2} by the relation $j_2 = j_1 - j_{l2}$. Consider that the condensation zone is shown schematically in the Figure. Information about its position and length (width) can be obtained, for example, by using solutions of the Boltzmann kinetic equation.

When vapor moves along the axis x, the value of the specific mass flow of the condensed (due to homogeneous nucleation) substance (j_l) can be determined using the following relationship:

$$\frac{dj_l}{dx} = m_{cr} J_{CNT}. \tag{1}$$

Here m_{cr} is the mass of critical drop J_{CNT} is the nucleation rate.

According to the classical theory [1,4], the nucleation rate can be determined as follows:

$$J_{CNT} = \beta \frac{\rho_v^2}{\rho_l} \sqrt{\frac{2\sigma}{\pi m^3}} \exp\left(-\frac{W}{kT}\right). \tag{2}$$

Here β is condensation coefficient, m is vapor molecule weight, ρ_v is vapor density, ρ_l is liquid density, σ is surface tension coefficient, k is Boltzmann constant, T is ambient temperature, W is work of critical droplet formation (r_{cr}),

$$W = \frac{4\pi r_{cr}^2 \sigma}{3}. \tag{3}$$

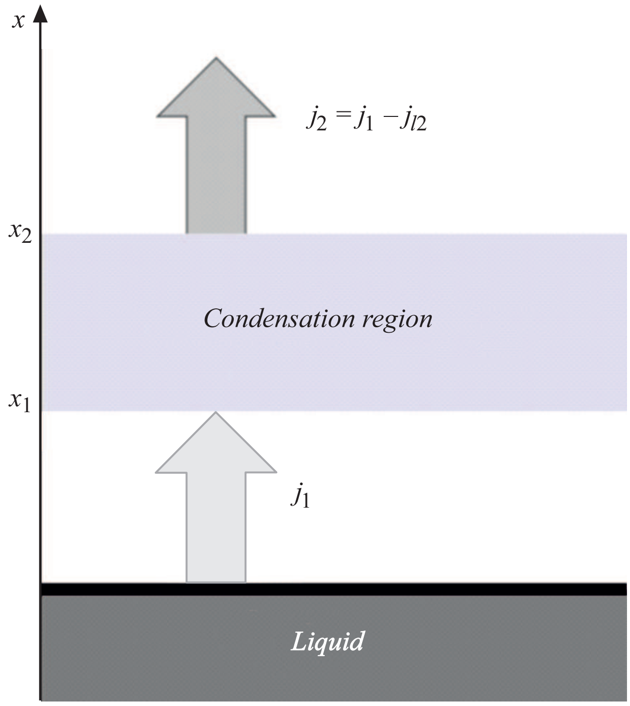

Figure 1. Task scheme. j_1 — mass flow density during evaporation from the liquid surface, j_{l2} — mass flow density of the condensed substance.

The supersaturation is related to the critical radius of the drop as follows:

$$\ln S = \frac{2\sigma M}{\rho_l r_{cr} k N_{\mathrm{A}} T}. \qquad (4)$$

Here S is supersaturation M is molecule mass of the substance, N_{A} is Avogadro's constant.

Equation (1) can be integrated and written as

$$j_{l2} = \frac{4}{3}\pi r_{cr}^3 \rho_l J_{CNT} \Delta x. \qquad (5)$$

From (5) it follows that the condensation mass flow is directly proportional to the width of the condensation zone $\Delta x = x_2 - x_1$.

To assess the influence of the homogeneous nucleation process on the flow parameters, it is necessary to have a relationship between the width of the condensation zone and the time of critical size droplet formation. Indeed, if the average time of critical size droplet formation (τ_f) is longer than the time the vapor stays in the supersaturation region (τ_d), then the process of homogeneous nucleation cannot be realized, i.e. the following condition must be met: $\tau_f \leqslant \tau_d$. Thus, to determine the possibility of the homogeneous nucleation process, it is necessary to estimate the time of critical size droplet formation (τ_f).

Expression (2) allows one to determine the number of drops of critical size formed in the volume V per time unit. Therefore, the characteristic time of one drop formation can be determined as follows:

$$\tau_f = \frac{1}{J_{CNT} V}. \qquad (6)$$

Here V is volume in which one drop of critical radius is formed.

Obviously, the volume V must contain at least the same number of vapor molecules as in the drop of critical radius, i.e. if we assume that all the vapor molecules in the volume V pass into the liquid phase with volume V_{cr}, we can obtain the relationship between the desired volume V and the volume of the critical drop: $V\rho_v = V_{cr}\rho_l$.

In this case, the time of formation of the drop of critical size can be found as follows:

$$\tau_f = \left(J_{CNT} \frac{4}{3}\pi r_{cr}^3 \frac{\rho_l}{\rho_v} \right)^{-1}. \qquad (7)$$

The width of the condensation zone is defined as the product of the vapor velocity and the time of the critical drop formation:

$$\Delta x = \frac{j_1}{\rho_v} \tau_f. \qquad (8)$$

The presented estimate gives the minimum length of the condensation zone, since in this case all vapor „coming“ from the evaporation surface must be condensed as a result of homogeneous condensation. Indeed, from (5), (7) and (8) one can obtain that $j_{l2} = j_1$.

Figs 2, 3 show the calculated dependences of the minimum length of the condensation zone Δx and the formation time of the nucleus of critical size τ_f for different substances on the supersaturation ratio. The range of change of the supersaturation degree was chosen based on the results of a kinetic analysis of the evaporation problem [3].

The classical theory often does not allow one to obtain experimentally observed nucleation rates, and the differences can reach several orders of magnitude. As noted in [8], one of the unsolved problems of classical models is the processing of small clusters. For example, in [9,10] it is shown that, depending on the substance, the drops size range, and the temperature the surface tension of small clusters can by 5–25% exceed the values obtained for large drops. The dashed lines in Fig. 2 show data on the size of the condensation zone, taking into account the assumption that the surface tension coefficient for drops of critical size can be by 25% greater than for large drops.

The estimates made show that for the case of methanol evaporation, the process of homogeneous nucleation should be taken into account at almost any supersaturation ratio (evaporation rates): the length of the condensation zone is fractions of a millimeter, while the surface tension does not significantly affect this conclusion. At the same time, this is not true for the case of water evaporation. For example, if we assume that the surface tension coefficient does not depend on the drop size (solid lines with circles

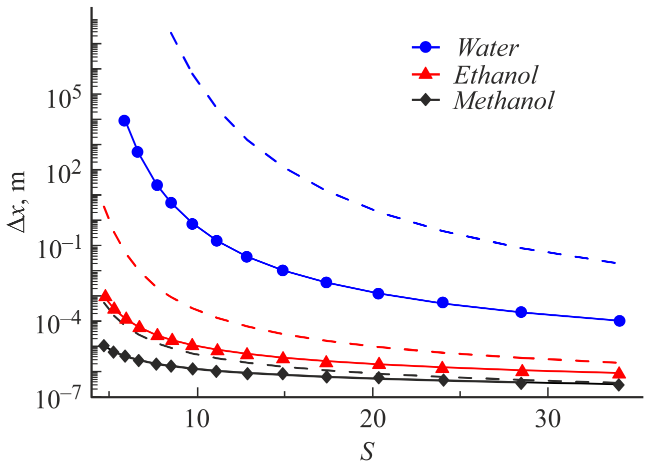

Figure 2. Mnimum width of the condensation zone vs. supersaturation ratio. Solid lines with symbols — surface tension coefficient for large drops; dashed lines — surface tension coefficient for drops of critical size (by 25% more than for large drops).

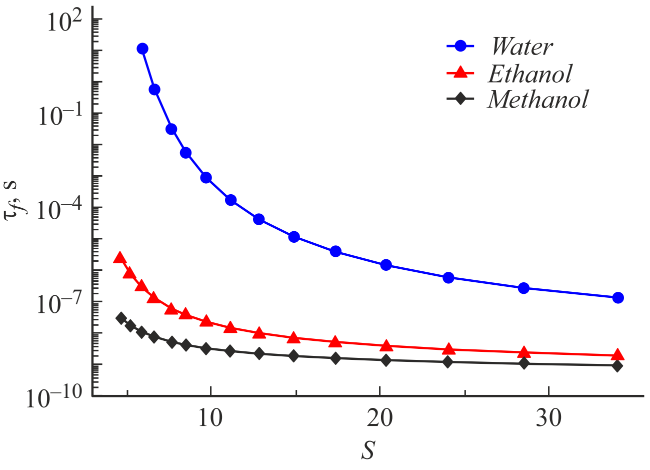

Figure 3. Formation time of critical size cluster vs. supersaturation degree.

in Fig. 2), the width of the condensation zone for the supersaturation 20 is 1 mm. If the surface tension becomes by 25% higher, then the width of the condensation zone in this case is $\simeq 2$ m, and for the supersaturation 35 it is $\simeq 1$ cm. The reliability of the obtained results of calculation estimates is justified by the legitimacy of using the classical homogeneous nucleation theory as a first approximation. Quite reasonable calculation data obtained for various values of the surface tension coefficients, shown in Figs 2 and 3, can be considered as an indicator of the adequacy of the model of reality.

The presented approach is evaluative one. In the general case, to correctly take into account the influence of the homogeneous nucleation process on the flow phenomena it is necessary to solve the complete problem of evaporation and condensation.

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Conflict of interest

The authors declare that they have no conflict of interest.

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