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## Change in evaporation flux due to homogeneous condensation of vapour near the interfacial surface

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Received February 17, 2023

Revised March 14, 2023

Accepted March 14, 2023

An approach that allows to estimate the necessity of homogeneous nucleation process accounting/not accounting for evaporation intensity and to predict the direction (increase or decrease) of mass flux density change of evaporating matter has been proposed. A dimensionless parameter making it possible to determine the areas of increase/decrease of evaporation intensity in the presence of a homogeneous nucleation process is proposed. It is shown that for some substances (methanol, ethanol) the direction of process significantly depends on the nature of heat exchange between the formed droplets and the surrounding vapor droplets.

**Keywords:** homogeneous nucleation, evaporation, supersaturation degree.

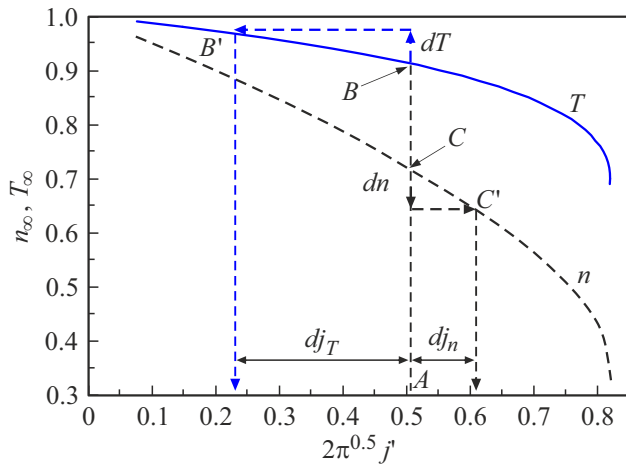
Evaporation and condensation processes are observed in various natural phenomena and manufacturing processes. If one has the capacity to adjust the intensity of evaporation and condensation, the efficiency of such processes as drying, chemical vapor deposition, vacuum distillation, cryo vacuum pumping, etc., may be enhanced considerably. It was demonstrated in [1] that vapor moving from the evaporation surface is supersaturated (i.e., the ratio of the partial vapor pressure to the saturated vapor pressure is higher than unity even at low evaporation intensities, and the supersaturation degree increases with the process intensity). It is common knowledge that homogeneous vapor condensation may proceed under these conditions [2]. The formation of a condensed phase within vapor leads to a reduction in density and an increase in temperature of vapor, thus affecting the evaporation intensity [3]. Therefore, the mutual influence of processes occurring within vapor and on the interfacial surface is an important feature of the considered problem.

Although the processes of evaporation and homogeneous nucleation have been studied for more than 100 years, the effect of spontaneous formation of liquid droplets near the interfacial surface on the evaporation intensity has hardly ever been discussed in literature. The data presented in [1] correspond to a distance of several free path lengths of vapor molecules from the phase boundary (i.e., to the outer boundary of the Knudsen layer). As was noted in [4,5], the stage of „removal“ of vapor molecules from the surface due to diffusion is an integral part of evaporation into a vapor–gas mixture. A diffusion vapor flux forms at a distance on the order of one free path length of vapor molecules from the evaporation surface (i.e., within the Knudsen layer). In view of this, it is crucial that homogeneous nucleation within the Knudsen layer be taken into account. The process of homogeneous nucleation near small evaporating particles

was examined in [6]. It was noted that „homogeneous vapor nucleation near evaporating droplets is a major issue for aerosol science and its practical application“. The formation of supersaturated vapor in the process of evaporation was reported in [7]. In addition, the observation of droplets forming in supersaturated vapor is one of the feasible experimental methods for determination of the nucleation rate [8,9].

A simplified approach, which allows one to estimate the necessity of taking into account the influence of homogeneous nucleation on the flow of vapor forming near the interfacial surface as a result of evaporation, was proposed in [10]. It was demonstrated in this study that homogeneous nucleation in certain substances (e.g., water) needs to be taken into account only at high supersaturation degrees (evaporation intensities). The influence of heat released as a result of the phase transition was neglected. The present study is a continuation of [10]. Here, we propose a method that allows one to determine (predict) the direction of variation (increase or decrease) of the mass flux density of evaporating matter as a result of homogeneous nucleation with both droplet formation and heat release taken into account.

It follows from Fig. 1, which presents the results obtained in [1] in solving the problem of intense evaporation, that the evaporation intensity increases with decreasing number density of vapor at infinity  $n_\infty$ . At the same time, heat released as a result of the phase transition induces an increase in vapor temperature, thus reducing the evaporation intensity. Thus, it can be said that two opposing factors (vapor density reduction and vapor temperature  $T_\infty$  rise) affect the evaporation flux in the process of homogeneous nucleation. Therefore, the mass flux becomes functionally dependent on two variables: density and temperature. One



**Figure 1.** Interrelation between evaporation intensity  $j'$  and vapor parameters  $n_\infty$  and  $T_\infty$ . Here,  $j' = \frac{j}{\rho_s \sqrt{2RT_s}}$ ,  $R$  is the individual gas constant, and  $\rho_s$  is the density corresponding to surface temperature  $T_s$  along the saturation line.

may then write the following:

$$dj_{GN} = dj_T + dj_n, \quad (1)$$

where  $dj_{GN}$  is the variation of evaporation flux due to homogeneous nucleation,  $dj_T = \frac{\partial j}{\partial T_\infty} dT_\infty$  is the flux variation due to temperature change, and  $dj_n = \frac{\partial j}{\partial n_\infty} dn_\infty$  is the flux variation due to concentration change. It follows from Fig. 1 that  $n_\infty$  and  $T_\infty$  are functions of  $j$ ; i.e.,  $n_\infty = n(j)$ ,  $T_\infty = T(j)$ . The data from [1] are presented in Fig. 1 in a dimensionless form, which provides an opportunity to use these dependences in examining the process of evaporation of various substances.

Let us consider an evaporation process with a given intensity corresponding to a certain point (A) on the abscissa axis in Fig. 1. It can be seen that certain initial density and temperature values, which are denoted as C and B, respectively, in Fig. 1, correspond to this evaporation intensity. If the vapor density decreases by  $dn$  as a result of nucleation, it follows from Fig. 1 that vapor far from the interfacial surface goes to a new state, which is denoted by point C' and corresponds to a new evaporation flux being higher (by  $dj_n$ ) than the initial one. At the same time, when the vapor temperature increases by  $dT$ , the system goes to state B' with the evaporation flux reduced by  $dj_T$ . The values of  $dj_n$  and  $dj_T$  are shown schematically in Fig. 1. It is evident that the ratio between  $dj_n$  and  $dj_T$  dictates whether the flux remains the same, increases, or decreases.

Expression (1) for an arbitrary initial evaporation intensity may be written as

$$dj_{GN} = \frac{\partial j}{\partial T_\infty} dT_\infty + \frac{\partial j}{\partial n_\infty} dn_\infty. \quad (2)$$

The variation of number density and temperature of vapor due to nucleation may be determined using the following

expressions:

$$m_1 \frac{dn_\infty}{dt} = -\dot{M}, \quad (3)$$

$$m_1 n_\infty c_{p,g} \frac{dT_\infty}{dt} = \alpha L \dot{M}, \quad (4)$$

where

$$\dot{M} = \rho_l \frac{4}{3} \pi r_{cr}^3 J_{CNT}, \quad r_{cr} = \frac{\sigma}{\rho_l R T_\infty \ln S},$$

$$J_{CNT} = \beta \frac{\rho_g^2}{\rho_l} \sqrt{\frac{2\sigma}{\pi m_1^3}} \exp\left(-\frac{W_{cr}}{k_B T}\right) \text{ — homogeneous nucleation}$$

rate,  $W_{cr} = \frac{16\pi\sigma^3 m_1^2}{3(\rho_l R T \ln S)^2}$  — formation work of a droplet with critical radius  $r_{cr}$ ,  $L$  — phase-transition heat,  $\alpha$  — fraction of heat spent on vapor heating ( $0 < \alpha \leq 1$ ),  $m_1$  — mass of a vapor molecule.

Using (3) and (4), one may rewrite expression (2) as

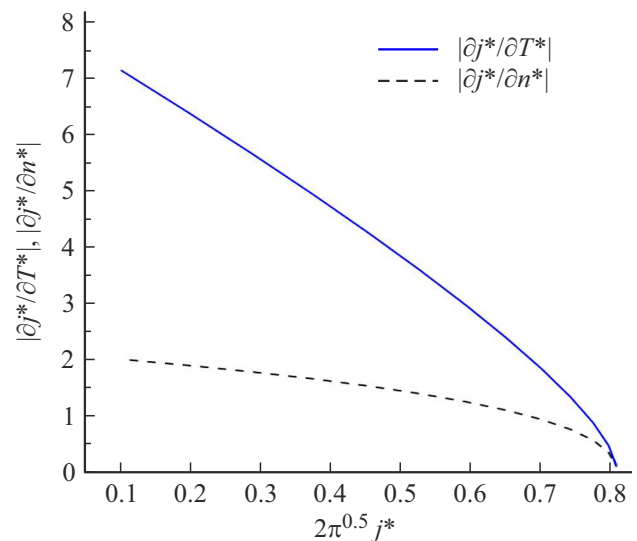
$$dj_{GN} = J_{CNT} \left( \frac{\alpha L}{n_\infty c_{p,g}} \frac{\partial j}{\partial T_\infty} - \frac{\partial j}{\partial n_\infty} \right) \frac{\rho_l \frac{4}{3} \pi r_{cr}^3}{m_1} dt. \quad (5)$$

It can be seen from Fig. 1 that derivatives  $\frac{\partial j}{\partial T_\infty}$  and  $\frac{\partial j}{\partial n_\infty}$  always assume negative values; i.e., the flux decreases with increasing temperature and concentration. It is then convenient to rewrite expression (5) in the following form:

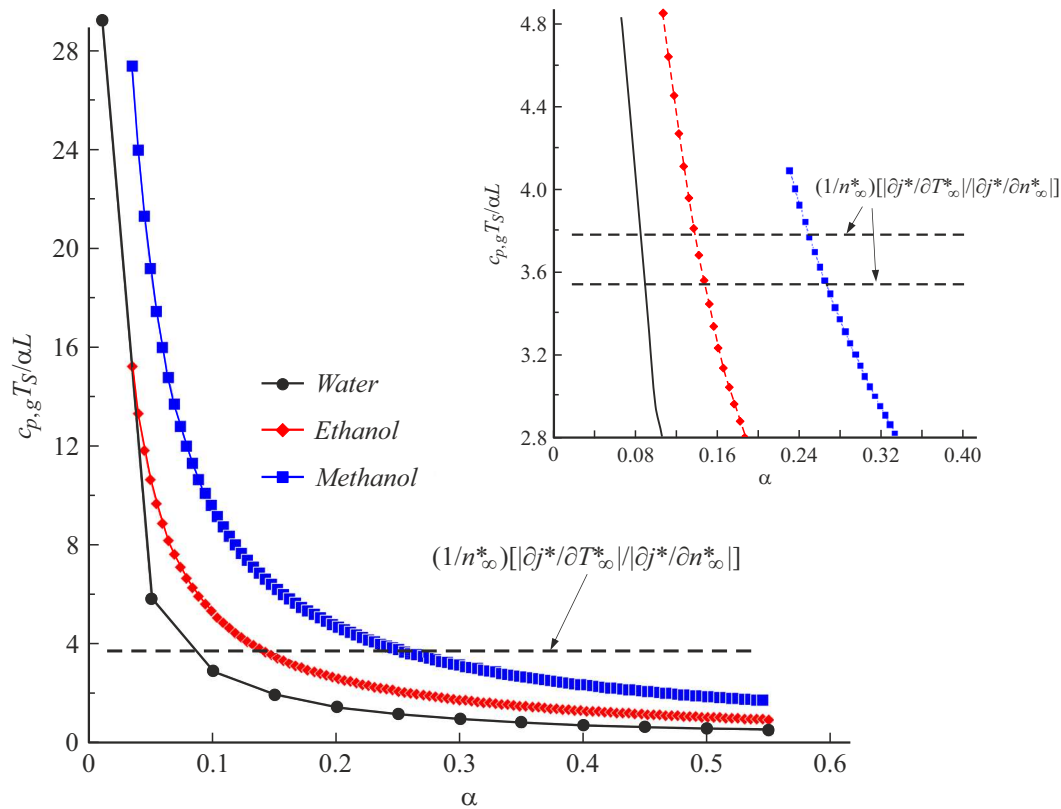
$$\frac{dj_{GN}}{dt} = J_{CNT} \left( \left| \frac{\partial j}{\partial n_\infty} \right| - \frac{\alpha L}{n_\infty c_{p,g}} \left| \frac{\partial j}{\partial T_\infty} \right| \right) \frac{\rho_l \frac{4}{3} \pi r_{cr}^3}{m_1}. \quad (6)$$

The conditions for evaporation intensity reduction or growth in the presence of homogeneous nucleation may be derived from (6):

$$\begin{aligned} \frac{c_{p,g} T_s}{\alpha L} > \frac{1}{n_\infty^*} \left| \frac{\partial j^* / \partial T_\infty^*}{\partial j^* / \partial n_\infty^*} \right|, & \quad \frac{dj_{GN}}{dt} > 0, \\ \frac{c_{p,g} T_s}{\alpha L} < \frac{1}{n_\infty^*} \left| \frac{\partial j^* / \partial T_\infty^*}{\partial j^* / \partial n_\infty^*} \right|, & \quad \frac{dj_{GN}}{dt} < 0, \end{aligned} \quad (7)$$



**Figure 2.** Dependences of derivatives  $\left| \frac{\partial j^*}{\partial T^*} \right|$  and  $\left| \frac{\partial j^*}{\partial n^*} \right|$  on the value of  $j^*$ .



**Figure 3.** Dependences of  $\frac{c_{p,g}T_S}{\alpha L}$  on  $\alpha$  for various substances.

Dimensionless variables (denoted with an asterisk) were introduced here. Evaporation surface temperature  $T_S$  and number density  $n_S$  corresponding to this temperature along the saturation line were used as the base values for  $T_{\infty}$  and  $n_{\infty}$ ; the mass flux density was made dimensionless in the following way:  $j^* = \frac{j}{m_1 n_S \sqrt{2RT_S}}$ . Our estimates demonstrate that the value of dimensionless complex  $\frac{1}{n_{\infty}^*} \frac{|\partial j^* / \partial T_{\infty}^*|}{|\partial j^* / \partial n_{\infty}^*|}$  varies within the range from 3.53 to 3.77. The plots of  $|\frac{\partial j^*}{\partial T_{\infty}^*}|$  and  $|\frac{\partial j^*}{\partial n_{\infty}^*}|$ , which were drawn using the data from Fig. 1, are presented in Fig. 2 (index  $\infty$  is omitted).

Figure 3 shows the dependences of  $\frac{c_{p,g}T_S}{\alpha L}$  for various substances (water, methanol, ethanol) on fraction  $\alpha$  of heat released as a result of the phase transition and spent on vapor heating (a close-up of the dependences is shown in the inset of Fig. 3). Dashed lines in the inset of Fig. 3 denote the range of variation of dimensionless complex  $\frac{1}{n_{\infty}^*} \frac{|\partial j^* / \partial T_{\infty}^*|}{|\partial j^* / \partial n_{\infty}^*|}$ . It is evident that even if the fraction of released heat spent on vapor heating is relatively small, homogeneous nucleation reduces the evaporation intensity in all the examined substances. The evaporation flux may increase only if fraction  $\alpha$  of heat released as a result of the phase transition and spent on vapor heating does not exceed a certain threshold. For example, this limit value is approximately 0.08, 0.14, and 0.25 for water, ethanol, and methanol, respectively. If the fraction of heat absorbed

by vapor exceeds this limit, the evaporation flux decreases. Thus, dimensionless parameter  $\frac{c_{p,g}T_S}{\alpha L}$  introduced here may be used to identify the regions of evaporation intensity growth and reduction in the presence of homogeneous nucleation. The reported data are valid only at the initial stage of volume condensation, which is significantly shorter than the process of subsequent droplet growth in the process of vapor condensation on the surface of droplets. Data listed in the table below were used to draw the plots in the present study.

Properties of substances

Substance	$c_{p,g}$ , J/(kg · K)	$L$ , $10^6$ J/kg	$T_S$ , K
Water	2200.0	2.26	300
Methanol	3663.8	1.17	300
Ethanol	1605.5	0.92	300

### Funding

This study was supported by grant No. 22-19-00044 from the Russian Science Foundation.

### Conflict of interest

The authors declare that they have no conflict of interest.

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*Translated by D.Safin*