THERMODYNAMICS OF CONDENSATION IN THE FORMATION OF A FILM ON A SOLUBLE NUCLEUS

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A determination is made of the thickness of a liquid film between the residue of a soluble nucleus and a vaporgas medium surrounding a drop condensed onto the nucleus from the vapor. The thickness is determined with allowance for the disjoining pressure of the film and the maximum chemical potential of the condensing substance. The maximum of this potential is found in the region of incomplete dissolution of the nucleus. A determination is also made of the threshold value of vapor chemical potential which is necessary for condensation on the nucleus to occur without a barrier when the nucleus is either partially or completely dissolved.

In [1], condensation on soluble nuclei was examined for very large drops in the case when the dissolution of the nucleus was completed. The region of drop sizes in which dissolution of the nucleus in the drop was not yet complete was studied in [2]. In this region, a liquid film is formed between the incompletely dissolved nucleus (its residue) and the vapor-gas medium surrounding the drop. The role of the disjoining pressure of the liquid film was explained in [3, 4] in an examination of the problem of condensation on an insoluble nucleus. However, the same factor was not fully accounted for in [2], where condensation on a soluble nucleus was considered. The present study is devoted to determination of the thermodynamics on a soluble nucleus in the region of its incomplete dissolution in a drop with allowance for the disjoining pressure of the liquid film formed between the nucleus residue and the vapor-gas medium.

Of the most practical interest is the case of condensation nuclei large enough so that condensation can proceed at low levels of supersaturation of the vapor. It was shown in [2] that, in this case, the solubility of the nucleus in the drop must be low in order for the region associated with incomplete dissolution to be the main factor determining the chemical potential of the vapor at the threshold for barrier-less condensation. Thus, only under the circumstances just described will this region be important for the thermodynamics of condensation. It will therefore be sufficient to consider the solution of the substance of the nucleus in the drop to be weak in the given region. In this study, we will limit ourselves to the case [1] when the substance of the nucleus — which might be an electrolyte — has no surface activity and is incapable of micelle formation inside the drop. This is the case most characteristic of phase transformations in the earth's atmosphere which involve water. It is not important whether the nucleus is solid or liquid, as in [1, 2].

Chemical Potential of the Condensate in the Region of Incomplete Dissolution of the Nucleus. Following [1, 2]. we will use ν_n to designate the number of molecules or ions initially present in the vapor-gas medium of the nucleus, while R_n will denote the radius of the nucleus. We also use ν_n' to denote the number of molecules or ions in the still-undissolved portion of the nucleus (residue). The symbol R_n' will denote the radius of the residue. It is obvious that

$$v = 4\pi R_{*}^{3}/3v_{*}, v_{*}' = 4\pi R_{*}^{3}/3v_{*}$$

where v_n is the volume occupied by one molecule in the nucleus or the mean volume occupied by one ion in the nucleus.

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Following [1, 2], we use ν to denote the number of molecules of condensate in the drop (condensed in the drop from the vapor phase of the substance) and R to represent the radius of the drop. Assuming the solution of the substance of the nucleus in the drop to be weak, we have

$$v = 4\pi \left(R^3 - R_{*}^{3} \right) / 3v_{i}$$
⁽²⁾

where v_l is the molecular volume of the condensing fluid.

By the concentration of the dissolved substance in the drop, we mean its relative concentration — the number of molecules or ions of dissolved nuclear material per molecule of condensate. As in [2], we will designate this concentration as x and refer to it simply as the concentration of the solution. In the absence of adsorption, we then have

$$x = (v_n - v_n')/v \tag{3}$$

Using (1-2), we can write (3) as

$$x = \frac{v_i}{v_n} \frac{R_n^3 - R_n^{-3}}{R^3 - R_n^{-3}}$$
(4)

It was shown in [2] that when condensation begins without a barrier in the region of incomplete dissolution of the nucleus, the concentration of the solution in the drop can be considered to be identical to the equilibrium concentration at the surface of the residue. In accordance with the Ostwald-Freundlich formula, we have

$$x = x_{\rm sol} \exp\left(\frac{2\gamma_{\rm s} v_{\rm s}}{kTR_{\rm s}}\right)$$
(5)

where x_{sol} is the solubility of the substance of the nucleus at the plane interface with the surrounding medium: γ_n is the mechanical surface tension of the nucleus; T is temperature; k is the Boltzmann constant. The exponent in (5) accounts for the curvature of the surface of the residue. As we will see below, the curvature of the surface of the residue will be small in the interval of drop radii R that is important theoretically. Our use of the Ostwald—Freundlich formula (5) is therefore justified. The condition of weakness of the solution, corresponding to $x \ll 1$, is reduced by (5) to

$$x_{sol} \exp\left(\frac{2\gamma_{s}v_{s}}{kTR_{s}'}\right) << 1$$
(6)

With a positive value of mechanical surface tension of the nucleus, it is obvious that $x > x_{sol}$. Thus, condition (6) is more restrictive than the condition $x_{sol} << 1$.

Combining (4) and (5), we obtain

$$\frac{R_{a}^{3} - R_{n}^{3}}{R^{3} - R_{n}^{2}} = \frac{v_{a}}{v_{i}} x_{sol} \exp\left(\frac{2\gamma_{s}v_{a}}{kTR_{n}}\right)$$
(7)

which connects the radius \mathbb{R}^n of the residue with the radius R of the drop. Because of (2), ν will also be a function of R. We take R (rather than ν , as in [1, 2]) below as a variable to describe the drop in the region of incomplete dissolution of the nucleus.

In this region, the chemical potential of the condensate has three components [2]. The first component, always negative, describes the way in which the condensate is affected by the nuclear material dissolved in it. The second component, always positive, describes the way in which the condensate is affected by the capillary pressure created by the surface of the drop. The third component describes the manner in which the condensate is affected by the disjoining pressure of the liquid film between the residue and the vapor-gas medium surrounding the drop. This component is negative when disjoining pressure is positive.



Fig. 1. Qualitative dependence, on R, of chemical potential b_R (solid line) and the contributions of solution concentration (1), capillary pressure (2), and disjoining pressure (3) to b_R .

The first and second components of condensate chemical potential were found in [1, 2]. As was shown in [3, 4], the contribution of disjoining pressure can be obtained from the contribution of capillary pressure by replacing the latter by disjoining pressure taken with a minus sign. Thus, for a weak solution in the drop we have

$$b_{R} = -x + 2\varsigma v_{j}/kTR - v_{j} \Pi/kT$$
⁽⁸⁾

where b_R is the chemical potential of the condensate expressed in thermal energy units kT and reckoned from the value corresponding to the equilibrium of the vapor with the condensing liquid when their interface is planar (the designation b_{μ} was used in [1, 2] in place of b_R); σ is the surface tension of the drop; Π is the disjoining pressure of the film.

Having aqueous solutions and other highly structured liquids foremost in mind, we will use the following exponential approximation [5, 6] for the disjoining pressure of the film:

$$\Pi = K \exp\left(-h/l\right) \tag{9}$$

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(11)

where

$$h \equiv R - R_{\bullet}' \tag{10}$$

is the thickness of the film; K and *l* are empirical constants (*l* corresponds to the correlation length in the film). We will see later that the below inequality is observed in the range of drop radii R that is of theoretical importance

In this case, the film can be considered planar. Thus, use of the plane-film approximation in (9) is justified. Inequality (11), together with definition (10), also shows that the following is valid within the radius interval important for the theory

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$$R' = R \tag{12}$$

The below approximate equality exists for the constant K in approximation (9)

$$\zeta = s/l \tag{13}$$

where s is the spreading factor.

The solid and dashed lines in Fig. 1 show the effect of R on the chemical potential of the condensate b_R and the contributions to this potential from the concentration of the solution, capillary pressure, and disjoining pressure. The point of inflection on the solid line and on line 1 separate the regions of incomplete and complete dissolution of the nucleus in the drop (in the latter region, disjoining pressure makes no contribution to b_R). It is one of our goals here to determine which of the maximum value.

Maximum of the Chemical Potential of the Condensate in the Region of Incomplete Dissolution of the Nucleus. It is evident from the figure that the chemical potential of the condensate has a maximum in the region in which dissolution of the nucleus is incomplete. This maximum is of decisive importance in determining the conditions for barrier-less condensation in the given region, and we will now focus on its investigation. The interval of drop radii R that is important for the theory is also in the neighborhood of this maximum.

The position of the maximum on the axis of the variable R is determined by the equation

$$\left(\frac{\partial b_R}{\partial R}\right)\Big|_{R=R_0} = 0 \tag{14}$$

The zero subscript denotes quantities corresponding to the maximum (in contrast to [1], we are concerned with the maximum in the region of incomplete dissolution of the nucleus).

Expanding (14) by means of (8) and taking (5), (9) and (10) into account, we obtain

$$\left\{ \left[x_{unl} \frac{2\gamma_{n}v_{n}}{R_{n}^{\prime 2}} \exp\left(\frac{2\gamma_{n}v_{n}}{kTR_{n}^{\prime 2}}\right) - \frac{v_{n}KR_{n}^{\prime 2}}{lR^{2}} \exp\left(-\frac{h}{l}\right) \right] \frac{\partial R_{n}}{\partial R} - \frac{2\sigma v_{l}}{R^{2}} + \frac{v_{l}KR_{n}^{\prime 2}}{lR^{2}} \exp\left(-\frac{h}{l}\right) \right\} \right|_{R=R_{0}} = 0$$
(15)

Differentiating (17) (which connects R_n' and R) with respect to R, we obtain the following for the derivative $\partial R_n'/\partial R$ from the small quantity in the left side of inequality (6):

$$\frac{\partial R_{n}}{\partial R} = -x_{sol} \frac{v_{n} R^{2}}{v_{l} R_{n}^{2}} \exp\left(\frac{2\gamma_{n} v_{n}}{k T R_{n}}\right)$$
(16)

Again using (6) and taking (12) into account, we find from (16) that:

$$|\partial R_i'/\partial R| << j \tag{17}$$

We immediately conclude from (6-7) that the below equality is observed with a much higher degree of accuracy than approximate equality (12)

$$R_{\bullet}' = R_{\bullet} \tag{18}$$

This validates (17). The physical meaning of this is that for a low solution concentration, the thickness of the film and, accordingly, the radius of the drop are determined mainly by the transfer of substance to the film from the vapor rather than from the nucleus. This fact also explains why dissolution of the nucleus has little effect on solution concentration.

In the more interesting (for practical purposes) case in which the radius R_n of the condensation nuclei initially present in the vapor-gas medium is large enough so that condensation is possible at low levels of supersaturation of the vapor, the curvature of the surface of the residue will (in accordance with (18)) be small enough to justify use of the Ostwald—Freundlich formula (5).

It is evident from (6), (12), and (17) that the term with the derivative $\partial R_n'/\partial R$ in Eq. (15) does not play any significant tole. Ignoring it and solving the equation, we obtain the following equation with a high degree of accuracy when we consider (18)

$$h_0 = l \ln \left(\frac{KR_s^2}{2sl}\right) \tag{19}$$

which also determines the thickness of the film at maximum chemical potential of the condensate in the region of incomplete dissolution of the nucleus. Equality (19) agrees with Eq. (14) in [3].

Since the dependence of h_0 on K in (19) is logarithmic, then despite the appreciable relative error of (13) we obtain the following highly accurate result from (19), (13)

$$h_0 = l \cdot \ln \left(\frac{s R_s^2}{2 z l^2} \right) \tag{20}$$

In the most interesting practical case of nucleus radii R_n large enough so that condensation is possible at low levels of supersaturation of the vapor, we find that

$$R_{n} \gg l \tag{21}$$

The quantity in the right side of (20) increases logarithmically with an increase in R_n , i.e. it increases more slowly than R_n . Thus, in the case of large R_n , in accordance with (20-21) and $s \sim \sigma$ we have $R_n >> h_0$. The latter, together with (12) and (18), validates inequality (11) at the point corresponding to the maximum of chemical potential. Thus, these expressions also validate the inequality in the theoretically important neighborhood of this point.

In accordance with (5), (8), (9), (18) and (19), we have the following for the maximum value b_{R_0} of condensate chemical potential in the region of incomplete dissolution of the nucleus:

$$b_{R_0} = -x_{sol} \cdot \exp\left(\frac{2\gamma_s v_s}{kTR_s}\right) + \frac{25v_i}{kTR_s} - \frac{25v_i}{kTR_s^2}$$
(22)

The three terms in the right side of (22) represent the contributions to the maximum of chemical potential from the solution concentration, capillary pressure, and disjoining pressure, respectively. The constant K is dropped from Eq. (22).

The position of the chemical potential maximum determined by Eq. (19) is determined entirely by the competition between the contributions to the derivative $\partial b_R/\partial R$ from capillary pressure and disjoining pressure (the contribution to $\partial b_R/\partial R$ from the solution concentration, accounted for in (15) by the term with the derivative $\partial R_n'/\partial R$, turned out to be relatively small and was therefore omitted in the derivation of Eq. (19)). Nevertheless, in accordance with (21), the contribution to the maximum value b_{R_0} of condensate chemical potential (as determined by Eq. (22)) from capillary pressure is much greater than the corresponding contribution from disjoining pressure. We will explain this next.

Capillary pressure is much less dependent on R than is disjoining pressure. After differentiation with respect to R, the role of a quantity which depends weakly on R is diminished compared to the role of a quantity which is heavily dependent on R. The contributions of capillary and disjoining pressure to the derivative $\partial b_R/\partial R$ at the point $R = R_0$ cancel one other and are thus equal in absolute value. But then the contribution to b_{R_0} from capillary pressure will be much greater than the corresponding contribution from disjoining pressure. Conversely, the contribution of capillary pressure to $(\partial^2 b_R/\partial R^2)$ will be much smaller than the corresponding contribution from disjoining pressure.

The foregoing explains why the extremum of condensate chemical potential at the point $R = R_0$ is a maximum: the main contribution to $(\partial^2 b_R / \partial R^2) |_{R=R_0}$ (from disjoining pressure) is negative. This also explains why the contribution from the quantity which is the least dependent on R in the neighborhood of $R = R_0$ — the concentration of the solution — is kept in (22) but omitted from (19).

Let us change over from γ_n and σ to dimensionless values of surface tension a_n and a for the nucleus and the drop

$$a_n \equiv (4\pi \gamma_n/kT)(3v_n/4\pi)^{23}, \quad a \equiv (4\pi c/kT)(3v_n/4\pi)^{23}$$

(the definition of a is consistent with its definition (4) in [1]). Using (23) and the first equality of (1) and, in accordance with (21), ignoring the contribution of disjoining pressure in (22), we obtain

$$b_{R_0} = -x_{m1} \cdot \exp\left(\frac{2a_n}{3v_n^{V_3}}\right) + \frac{2a}{3v_n^{V_3}}$$

where for simplicity we have put $v_n = v_l$. Equation (24) expresses the maximum of condensate chemical potential in the region f incomplete dissolution of the nucleus in terms of the external parameters of the problem x_{sol} , a_n , a, and v_n .

Threshold Value of the Chemical Potential of the Vapor. The maximum value of condensate chemical potential 1 the region of complete dissolution of the nucleus was found in [1] (and previously in [7])). It determines the threshold value tr of the chemical potential of the vapor necessary for condensation to occur on soluble nuclei without a barrier in the region f complete dissolution of the nucleus. As the chemical potential of the condensate, we express the chemical potential of the apor in thermal energy units kT and reckon it from the value corresponding to equilibrium of the vapor with the condensing luid when their interface is planar. In accordance with Eq. (11) from [1], we have

$$b_{\mu} = 2 \ (2a)^{3/2} / 27 v_{\mu}^{3/2} \tag{25}$$

The true threshold value of vapor chemical potential necessary for condensation on soluble nuclei to occur without a arrier in both the region in which nucleus dissolution is complete and in the region in which it is incomplete is obviously etermined by the greater of the quantities in the right sides of Eqs. (24) and (25). If

$$x_{mol} > \frac{2a}{3v_{n}^{1/2}} \exp\left(-\frac{2a_{n}}{3v_{n}^{1/2}}\right) \left[1 - \frac{2(2a)^{1/2}}{9v_{n}^{1/2}}\right]$$
(26)

ien the value in the right side of (25) turns out to be the largest. Otherwise, the value in the right side of (24) becomes the urgest. In either case, Eqs. (24) and (25) make it possible to easily find the true threshold value of vapor chemical potential.

In accordance with the Ostwald-Freundlich formula, the equality

$$x_{n} = x_{\text{sol}} \exp\left(\frac{2a_{n}}{3v_{n}^{1/3}}\right)$$
(27)

etermines the equilibrium concentration x_n of solution at the surface of a curved nucleus (such a nucleus being present initially n the vapor-gas medium). This concentration was referred to in [2] as the solubility of the nucleus. We can use (27) to write ordition (6) as

$$x_n \ll 1 \tag{28}$$

nd write inequality (26) as

$$x_{n} > \frac{2a}{3v_{n}^{13}} \left[1 - \frac{2(2a)^{12}}{9v_{n}^{16}} \right]$$
(29)

It was shown in [2] that inequality (29) places a lower bound on x_n . This value is sufficient for barrier-less ondensation on soluble nuclei to occur in the region of incomplete dissolution of the nucleus at all of the vapor chemical votentials at which barrier-less condensation occurs in the region of the nucleus' complete dissolution (and, thus, the true hreshold value of vapor chemical potential is determined by the quantity in the right side of (25)). Condition (28) need not we observed in this case, but is is necessary that the solution be weak at concentrations corresponding to the lower bound mposed on x by (29). Such a prerequisite is obviously met when

$$v_{a}^{\nu_{3}} >> 2a/3$$
 (30)

condition (19) in [2]). The significance of (29) thus turns out to be the same here as in [2]: the region of complete dissolution of the nucleus is the main factor determining the threshold value of vapor chemical potential.

As was noted in [2], condition (30) is entirely applicable in the case of the greatest practical interest. Specifically, it is notes for the case of condensation nuclei large enough so that condensation can take place at low levels of supersaturation of he vapor. If condition (30) is satisfied, then we can reach conclusions that are more far-reaching than those made in [2].

When condition (29) is satisfied, the true threshold value of vapor chemical potential is determined by the quantity in the right side of (25). In the opposite case (when (30) will ensure that condition (28) is observed), the true threshold value of vapor chemical potential is determined by the quantity in the right of (24).

Thus, if (30) is observed, the condition (29) found in [2] is a necessary as well as a sufficient condition for barrier-less condensation to occur on soluble nuclei in the region of incomplete dissolution at any vapor chemical potential at which such condensation would occur in the region of complete dissolution. The reason (29) turns out be be not only a sufficient but a necessary condition is the smallness of the contribution of disjoining pressure to b_{R_0} , as shown by (22). This circumstance was not accounted for explicitly in [2] (although it was assumed in [2] that this contribution is negative).

Numerical Calculations and Discussion of Results. When (30) is observed and $a_n \sim a$, we can write (24) as follows with a high degree of accuracy

$$b_{K_0} = -x_{sc} + \frac{2a}{3v_c^{13}}$$
(31)

where we have ignored the dependence of the solubility of the nucleus on the curvature of its surface as described by the Ostwald—Freundlich formula (although this is difficult to establish, due to the lack of reliable data on the parameter a_n). Here, obviously, (29) practically amounts to the limitation

$$x_{\text{sol}} > \frac{2a}{3v_{1}^{13}} \left[1 - \frac{2(2a)^{12}}{9v_{1}^{16}} \right]$$
(32)

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Let us consider a case typical of the winter atmosphere: condensation of water vapor on nuclei of NaCl. Na₂SO₄, and CaSO₄ — substances mentioned in decreasing order of their solubility at the temperature T = 273 K characteristic of the lower winter atmosphere. For water, we find from the second definition of (23) that a = 10 at T = 273 K. Being written in approximate form rather than as a strong inequality, condition (30) for the use of limitation (32) thus implies that $p_n^{1/3} \ge 10$ (the solution can be considered weak, as required for the use of (32), at the concentration corresponding to the lower bound of x_{sol} under (32)). The condition $\nu_n^{1/3} \ge 10$ is readily observed within the range 5 $10 \le \nu_n^{1/3} \le 5 \cdot 10^3$ characteristic of the condensation nuclei present in the winter atmosphere.

In the case of nuclei of NaCl, according to [8] we have (with allowance for complete dissociation in the solution) $x_{sol} = 0.29$ at T = 273 K — the solubility of the substance of the nucleus is high. Then limitation (32) is realized for nearly all $v_n^{1/3} \ge 50$. The true threshold chemical potential of the vapor is accordingly determined by the quantities in the right side of (25) and (at a = 10) is roughly equal to $6.63v_n^{-1/2}$. This also applies fully to highly soluble nuclei of CsJ, which together with NaCl nuclei may enter the winter atmosphere in large quantities due to natural processes or disasters.

According to [8], for Na₂SO₄ nuclei we have (with allowance for complete dissociation in the solution) $x_{sol} = 0.012$ at T = 273 K — the solubility of the substance of the nucleus is low. In this case, limitation (32) is realized only when $p_n^{1/3} \ge 5.6 \cdot 10^2$. Then the true threshold chemical potential of the vapor is determined by the quantity in the right side of (25) and. as already noted, is equal roughly to $6.63v_n^{-1/2}$. Conversely, limitation (32) is not realized at $v_n^{-1/3} < 5.6 \cdot 10^2$. In this case, the true threshold chemical potential of the vapor is determined by the quantity in the right of (31) and (at a = 10) is equal to approximately $-0.012 + 6.67v_n^{-1/3}$. As was noted in [2], the fact that it might already be substantial at $p_n^{1/3} < 5.6 \cdot 10^2$ (reaching values close to 0.12 at $v_n^{-1/3} = 50$ and exceeding the threshold value predicted by (25) by a factor of six) might be one explanation for the fact that nuclei of Na₂SO₄ — in which $v_n^{-1/3} < 5.6 \cdot 10^2$ (R_n $\le 1.1 \cdot 10^{-5}$ cm) — remain in the winter atmosphere for a long time.

The above remarks regarding the relative importance of the region of incomplete dissolution of the nucleus in the formation of vapor chemical potential at the threshold for barrier-less condensation are even truer in the case of nuclei of CaSO₄. Here, we have (with allowance for complete dissociation in the solution) $x_{sol} = 4.6 \cdot 10^{-4}$ at T = 273 K — the solubility of the nuclear material is low. Limitation (32) is realized only when $\nu_n^{1/3} \ge 1.5 \cdot 10^4$. It is certain that (32) is not satisfied in the range $5 \cdot 10 < \nu_n^{1/3} < 5 \cdot 10^3$ characteristic of condensation nuclei present in the winter atmosphere. The true value of the threshold chemical potential of the vapor in this range is determined by the quantity in the right side of (31) and (at a = 10) is approximately equal to $-4.6 \cdot 10^{-4} + 6.67\nu_n^{-1/3}$ (it reaches 0.13 at $\nu_n^{1/3} = 50$, exceeding the threshold value predicted by (25) by a factor of seven).

It should be noted that, in contrast to the cases of NaCl and $CaSO_4$, the solubility of Na_2SO_4 rapidly decreases with a decrease in temperature [8]. As is now clear, in the presence of a temperature gradient, this might in turn lead to stratification of the vapor-gas medium with respect to the size and composition of the condensation nuclei.

The results obtained here on the thermodynamics of condensation on soluble nuclei in the region of incomplete dissolution of nuclei in drops involve the use of an important physico-chemical concept: the solubility of the substance of the nucleus. This notion bridges the gap between the theory of condensation on soluble nuclei and the theory constructed in [3, 4] to explain condensation on insoluble nuclei. In both theories, competition between the contributions to the chemical potential of the condensate from the capillary and disjoining pressures in the liquid film ensures that this potential will have a maximum. It will be the only maximum in the case of an insoluble nucleus, while for a soluble nucleus it will be in addition to the maximum of condensate chemical potential in the region of complete dissolution of the nucleus (see Fig. 1).

The role of the disjoining pressure of the film in the formation of the potential maximum in the region of incomplete dissolution of the nucleus has not been addressed in the literature. The expediency of allowing for disjoining pressure in problems concerning condensation on an incompletely dissolved nucleus was discussed in general terms in the conclusion to [9], where the author took up the question of the manner in which condensate chemical potential is affected by the dependence of the solubility of the residue on the curvature of its surface. As is now clear, to raise this question is pointless if no allowance is made for the disjoining pressure of the liquid film. In fact, a maximum of condensate chemical potential is impossible even in principle in the absence of disjoining pressure from the film.

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