

ON THE KINETICS OF BINARY NUCLEATION

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Two problems of the theory of binary nucleation are solved: normalization of the equilibrium distribution function of nuclei and correct transition to the one-dimensional theory. Classification of multivariable nucleation processes is carried out and it is shown how to convert binary nucleation into a process with linked fluxes by means of the corresponding transformation of the variables describing a nucleus. Just the use of the variables (total number of monomers, composition) makes it possible to solve the given problems. Two transitions to the one-dimensional nucleation are described. One of them corresponds to the formation of nuclei with stoichiometric composition. The other transition is that to unary (single-component) nucleation.

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INTRODUCTION

The phenomena of binary and multicomponent nucleation are studied rather intensively both theoretically and experimentally. The examples of such processes include the formation of gas bubbles in a two-component solution of vacancies and gas atoms in solids under irradiation [1,2], the nucleation of two- and multicomponent precipitates in alloys and solid solutions, the formation of aerosols in atmosphere (nucleation in a gas mixture). The theory of these phenomena relates to more general multivariable theory of nucleation [3] which is an extension of Zel'dovich-Frenkel' one-dimensional theory [4,5] to the multivariable case. The phenomenological approach used in this theory is based on the expression for the work $\Delta\Phi(\xi_1, \xi_2, \dots, \xi_p)$ of the new-phase nucleus formation and the Fokker-Planck kinetic equation for the distribution function (DF) $f(\xi_1, \xi_2, \dots, \xi_p; t)$ in the space of the variables $\{\xi_i\}$ that describe a nucleus. Among the unsolved problems of binary nucleation, the following two can be singled out: normalization of the equilibrium DF and correct passage to the one-dimensional limit. Notice that the steady state nucleation rate and the DF can not be calculated correctly without exact value of the normalization constant. In the present report, both these problems are solved with the use of the results of general multivariable nucleation theory developed in Ref. [3].

MODEL AND MAIN RESULTS OF THE MULTIVARIABLE THEORY OF NUCLEATION

Near the saddle point ξ_* , where a nucleus is assumed to be a macroscopic subsystem, the work $\Delta\Phi$ can be represented as a quadratic form,

$$\Delta\Phi(\xi) = \Delta\Phi_* + \frac{1}{2} H(\xi_1, \xi_2, \dots, \xi_p),$$

$$H(\xi_1, \xi_2, \dots, \xi_p) = h_{ik} \xi_i \xi_k, \quad (1)$$

where $h_{ik} = \partial^2 \Delta\Phi / \partial \xi_i \partial \xi_k$ at $\xi = \xi_*$, $\Delta\Phi_* = \Delta\Phi(\xi_*)$,

and all variables are measured from their critical values; so, we have $\xi_* = 0$.

Upon being reduced to the sum of squares, this form has one negative term. This is a characteristic feature of the processes of multivariable nucleation; the corresponding variable is called "unstable". The nuclei which have passed over the energetic barrier in the vicinity of the saddle point as a result of Brownian motion in the space $\{\xi_i\}$ are the viable fragments of a new phase, so that the main problem of the theory is to calculate their flux over this barrier (the nucleation rate).

The work $\Delta\Phi(\xi)$ determines the equilibrium DF of nuclei as heterophase fluctuations [5]

$$f_0(\xi) = \text{const} \exp\left[-\frac{\Delta\Phi(\xi)}{kT}\right]. \quad (2)$$

As mentioned above, the evolution of the DF is described by the Fokker-Planck equation

$$\frac{\partial f(\xi, t)}{\partial t} = \frac{\partial}{\partial \xi_i} \left(d_{ij} \frac{\partial f(\xi, t)}{\partial \xi_j} - \dot{\xi}_i f(\xi, t) \right). \quad (3)$$

The condition of equality of the flux to zero in the equilibrium state makes it possible to obtain an equation for $\dot{\xi}_i$. Substituting $f_0(\xi)$ into eq. (3), we get

$$\dot{\xi}_i = -\frac{d_{ij}}{kT} \frac{\partial \Delta\Phi}{\partial \xi_j} = -\frac{d_{ij}}{kT} h_{jk} \xi_k \equiv -\frac{z_{ik}}{kT} \xi_k, \quad \mathbf{Z} = \mathbf{D}\mathbf{H}, \quad (4)$$

Hence

$$\mathbf{D} = \mathbf{Z}\mathbf{H}^{-1}. \quad (5)$$

This significant relationship shows that macroscopic equations of movement of a nucleus in its phase space $\{\xi_i\}$ (i.e. the matrix \mathbf{Z}) allow us to determine the matrix of diffusivities in the Fokker-Planck equation.

In Ref. [3], the following equation for the steady state nucleation rate has been obtained:

$$I = N(2\pi kT)^{-1/2} \sqrt{|h_{11}^{-1}|} |\lambda_1| e^{-\frac{\Delta\Phi_*}{kT}}, \quad (6)$$

where h_{11}^{-1} is an element of the matrix \mathbf{H}^{-1} ; N is the number of monomers in unit volume of the initial phase; λ_1 is the negative eigenvalue of the matrix \mathbf{Z} .

An equation for the steady state DF is as follows [3]:

$$f_s(\xi) = f_0(\xi) \sqrt{\frac{|\kappa(\mathbf{e})|}{2\pi kT}} \int_{\frac{eH\xi}{\kappa(\mathbf{e})}}^{+\infty} e^{-\frac{|\kappa(\mathbf{e})|}{2kT} y^2} dy \equiv \frac{1}{2} f_0(\xi) \operatorname{erfc} \left(-\frac{eH\xi}{\sqrt{2kT} |\kappa(\mathbf{e})|} \right), \quad (7)$$

where $\operatorname{erfc}(\xi) \equiv 1 - \operatorname{erf}(\xi)$; \mathbf{e} is the eigenvector of the matrix \mathbf{Z} corresponding to λ_1 (this is the flux direction); $\kappa(\mathbf{e})$ is the curvature of the normal section of the saddle surface $\Delta\Phi$ along the direction \mathbf{e} .

Equations for the flux direction in processes of two-variable nucleation have been derived in Ref. [3] for different relationships between the nucleation parameters. In the case of binary nucleation, the following equation holds:

$$tg\theta = \frac{\gamma_{22} - h_{11}}{2h_{12}} - (\operatorname{sign}h_{12}) \sqrt{\left(\frac{\gamma_{22} - h_{11}}{2h_{12}} \right)^2 + \gamma}, \quad (8)$$

where $\gamma = d_{22} / d_{11}$.

CLASSIFICATION OF MULTIVARIABLE NUCLEATION PROCESSES

All multivariable processes of nucleation can be divided into two classes: (i) processes with independent changes of variables and (ii) processes with linked fluxes [6,7]. In the first case, a variation in the variable ξ_1 in an elementary event does not affect the value of the variable ξ_2 and vice versa; both the variables are physically equivalent. The processes of binary nucleation are of this type: a nucleus is characterized by the numbers of monomers of each kind, $\xi_1 \equiv n_1$ and $\xi_2 \equiv n_2$; the elementary event is the attachment or detachment of a monomer. More general case is the multicomponent nucleation; correspondingly, a nucleus is characterized by the numbers $\xi_1 \equiv n_1$, $\xi_2 \equiv n_2$, ..., $\xi_p \equiv n_p$ of the monomers of each species.

In the second case, a change in the variable ξ_1 in an elementary event leads to a change in the variable ξ_2 . The variation in ξ_2 can be represented as the sum of a regular part $\delta\xi_2^{(r)}$ and a fluctuating part $\delta\xi_2^{(f)}$. In addition, the fluctuations of ξ_2 independent of ξ_1 are possible. An example is non-isothermal nucleation in a mixture of a vapor and an inert gas [6]. The variables are the number of vapor molecules in a cluster ($\xi_1 \equiv n$) and the cluster energy ($\xi_2 \equiv \varepsilon$). When a molecule is attached to a nucleus, the average energy of the latter likewise increases.

Of course, the combined processes are also possible, e.g. when non-isothermal effects are taken into account for the condensation of a vapor mixture; in this case, the variables are n_1 , n_2 , and ε .

As is evident, the roles of variables are different in the processes with linked fluxes. Consequently, there is

no need to bring the quadratic form H to the canonical form in order to single out the unstable variable: the number $\xi_1 \equiv n$ of monomers in a nucleus or its size (radius, volume) is a natural unstable variable, as in the one-dimensional theory. Definitive signs of h_{ik} correspond to this fact. So, e.g. in the two-dimensional case, the following signs have to be: $h_{11} < 0$ and $h_{22} > 0$. So, the normalization of the equilibrium DF for these processes with respect to stable variables [3] is carried out with the use of the fluctuation theory [8]. The normalization in respect to the unstable variable n is carried out in the same way, as in the one-dimensional theory [5].

In the processes of binary, as well as p -component, nucleation, the situation is quite different. The variables n_1 and n_2 are physically equivalent. Accordingly, h_{11} and h_{22} have to be of the same sign. It is not difficult to establish from the analysis of directions of the flux of nuclei on the (n_1, n_2) -plane that the only possible physical situation is $h_{11} > 0$ and $h_{22} > 0$ at $h_{12} < 0$. These signs also can be obtained in direct calculations of h_{ik} for concrete mixtures using the experimental dates for the corresponding thermodynamic quantities. So, we can not apply the algorithm described above for the normalization of the function $f_0(n_1, n_2)$. The extension of the one-dimensional algorithm of normalization [5] to this case can not be performed also, so the normalization constant $N = N_1 + N_2$ equal to the total number of monomers of both kinds, which is widely used for binary-nucleation processes in literature, is incorrect. Consequently, in order to normalize the function $f_0(n_1, n_2)$ we have to convert the given process to a process with linked fluxes, i.e. to change the roles of variables by their corresponding transformation. Apparently, the total number of monomers in a nucleus, $n = n_1 + n_2 + \dots + n_q$, and compositions, $c_2 = n_2 / n$, ..., $c_p = n_p / n$, must be taken as new variables. In the variables (n, c_2, \dots, c_p) , the p -component nucleation will be a process with linked fluxes with the corresponding signs of h_{ik} (the variable n is unstable, as before; the variables c_i are stable).

NORMALIZATION OF THE EQUILIBRIUM DISTRIBUTION FUNCTION

As an example of a binary nucleation process, we consider the condensation of the mixture of vapors of two substances into ideal solution. The work of nucleus formation has the following form [9]:

$$\Delta\Phi(n_1, n_2) = \chi_1 n_1 + \chi_2 n_2 + kT n_1 \ln \frac{n_1}{n_1 + n_2} + kT n_2 \ln \frac{n_2}{n_1 + n_2} + \alpha (v_1 n_1 + v_2 n_2)^{2/3}, \quad (9)$$

$$\chi_i = v_i (P - P_i^0) + kT \ln(P_i^0 / P_i),$$

where v_i is the molecular volume of the i th species in a nucleus, α is the surface tension, $P = P_1 + P_2$ is the sum of partial pressures of vapors, and P_i^0 is the vapor pressure of pure i th component at the temperature T .

We pass to the new variables $n = n_1 + n_2$ and $c = n_2 / (n_1 + n_2)$. Eq. (9) takes the form

$$\Delta\Phi(n, c) = -n\Delta\mu(c) + \alpha s(c)n^{2/3} \quad (10)$$

similar to the one-dimensional one. Correspondingly, $h_{11}^{(n,c)} < 0$ and $h_{22}^{(n,c)} > 0$ now.

In the vicinity of the saddle point (n_*, c_*) , the quadratic form in eq. (1) can be identically transformed to the following form:

$$H(n, c) = \frac{\det \mathbf{H}^{(n,c)}}{h_{22}^{(n,c)}} n^2 + h_{22}^{(n,c)} (c - c^{(e)}(n))^2, \quad (11)$$

where $c^{(e)}(n) = -(h_{12}^{(c,n)} / h_{22}^{(c,n)})n \equiv \text{tg} \theta_c^{(e)} n$ is determined from the condition of equilibrium $\partial \Delta\Phi(n, c) / \partial c = 0$ with respect to composition c .

So, the equilibrium DF $f_0(n, c)$ splits into two parts, $f_0(n, c) = \rho_0(n) \psi_{fl}(c)$:

$$\rho_0(n) = (N_1 + N_2) e^{-\frac{\Delta\Phi_* + (\det \mathbf{H}^{(n,c)} / 2h_{22}^{(n,c)})n^2}{kT}},$$

$$\psi_{fl}(c) = \sqrt{\frac{h_{22}^{(n,c)}}{2\pi kT}} e^{-\frac{h_{22}^{(n,c)}(c - c^{(e)}(n))^2}{2kT}}. \quad (12)$$

The ‘‘fluctuating’’ part $\psi_{fl}(c)$ of the equilibrium DF is normalized according to the theory of fluctuations [8], whereas the function $\rho_0(n)$ is normalized at $c = c^{(e)}(n)$ as in the one-dimensional theory [5], acquiring the factor $N = N_1 + N_2$. As it is known, $\langle (c - c^{(e)}(n))^2 \rangle = kT / h_{22}^{(n,c)}$. This equality will be used below for describing the limiting cases.

If the variables (n_1, n_2) are used nevertheless, then the DF $f_0(n_1, n_2)$ is got by the reverse transition $(n, c) \rightarrow (n_1, n_2)$:

$$f_0(n_1, n_2) = \frac{N_1 + N_2}{n_1 + n_2} \sqrt{\frac{h_{22}^{(n,c)}}{2\pi kT}} e^{-\frac{\Delta\Phi(n_1, n_2)}{kT}}, \quad (13)$$

where the multiplier $1/(n_1 + n_2)$ is the Jacobian of this transition. This equation solves the problem of normalization of the function $f_0(n_1, n_2)$.

The matrix \mathbf{D} has the simplest form in the variables (n_1, n_2) : there are only diagonal elements d_{11} and d_{22} . The matrix $\mathbf{D}^{(n,c)}$ in the variables (n, c) can be obtained by means of the transformation of initial movement equations (4) to these variables. Doing so, we find:

$$\mathbf{D}^{(n,c)} = \begin{pmatrix} d_{11} + d_{22} & -c_* d_{11} + (1 - c_*) d_{22} \\ -c_* d_{11} + (1 - c_*) d_{22} & c_*^2 d_{11} + (1 - c_*)^2 d_{22} \end{pmatrix}$$

so that $\det \mathbf{D}^{(n,c)} = \det \mathbf{D}^{(n_1, n_2)} / n_*^2$. Also, the following relationships can be derived: $\det \mathbf{H}^{(n,c)} = n_*^2 \det \mathbf{H}^{(n_1, n_2)}$, $\det \mathbf{Z}^{(n,c)} = \det \mathbf{Z}^{(n_1, n_2)}$, $\lambda_1^{(n,c)} = \lambda_1^{(n_1, n_2)}$. Hence, we also can calculate the steady state nucleation rate in the variables (n_1, n_2) using the normalizing constant from (13); the nucleation rate value (6) is invariant with respect to the transformations $(n_1, n_2) \leftrightarrow (n, c)$, as it must from the physical point of view.

LIMITING CASES

The equations $\partial \Delta\Phi(n_1, n_2) / \partial n_i = 0$, $i = 1, 2$, define the lines $L_e^{(1)}$ and $L_e^{(2)}$ of the equilibrium of a nucleus with respect to the variables n_1 and n_2 . They have the following directions in the vicinity of the saddle point:

$$\text{tg} \theta_e^{(1)} = -\frac{h_{11}}{h_{12}}, \quad \text{tg} \theta_e^{(2)} = -\frac{h_{12}}{h_{22}}. \quad (15)$$

There are kinetic and thermodynamic limits. Let us consider the former, $\gamma = d_{22} / d_{11} \rightarrow 0$ and $\gamma \rightarrow \infty$. The fact of the same signs of h_{11} and h_{22} reflects the certain symmetry of a system in respect to both the species. It leads to the symmetry of expression (9) with respect to n_1 and n_2 as well as to the following symmetry. In the limit $\gamma \rightarrow \infty$, we have

$\lambda_1 \rightarrow d_{11} \det \mathbf{H} / h_{22}$, and $\text{tg} \theta \rightarrow -h_{12} / h_{22} \equiv \text{tg} \theta_e^{(2)}$ (see eq. (8)). The kinetics of the second species predominates in this case, so that the equilibrium in respect to the variable n_2 has a chance to be established. Therefore, the flux vector tends to the line $L_e^{(2)}$. In the opposite case $\gamma \rightarrow 0$, we have $\lambda_1 \rightarrow d_{22} \det \mathbf{H} / h_{11}$, and $\text{tg} \theta \rightarrow -h_{11} / h_{12} \equiv \text{tg} \theta_e^{(1)}$, i.e. the replacement of indexes $1 \leftrightarrow 2$ in the expression for λ_1 takes place. So, the flux vector is enclosed between the equilibrium lines $L_e^{(1)}$ and $L_e^{(2)}$, as it must from the physical point of view.

Now let us pass to the variables (n, c) and consider the thermodynamic limits. In binary nucleation, two transitions to the one-dimensional theory are possible. The first of them, $h_{22}^{(n,c)} / kT \rightarrow \infty$ (the variance of c tends to zero), is general for the processes with linked fluxes [3]. In this limit, the variable c has the same, equilibrium, value $c^{(e)}(n)$ for all the nuclei of size n . It is seen from (12), that f_0 can be represented in this case as $f_0(n, c) = \rho_0(n) \delta(c - c^{(e)}(n))$; also, $c^{(e)}(n) \rightarrow 0$, and $(\det \mathbf{H}^{(n,c)} / h_{22}^{(n,c)}) \rightarrow h_{11}^{(n,c)}$, i.e. $\rho_0(n)$ converts to the DF of the one-dimensional theory. So, the variable c has the same, critical, value c_* for all the nuclei in this limit, i.e. it converts to the constant and falls out from consideration. This transition corresponds to the formation of the two-

component embryos with the fixed (stoichiometric) composition $c = c_*$.

The second transition is peculiar to this process. This is the passage to unary nucleation, when one of the components vanishes. Let us assume $N_2 \rightarrow 0$. Since $P_2 \sim N_2$, and $d_{22} \sim P_2$, we have: $\gamma \rightarrow 0$, and consequently $tg\theta \rightarrow 0$ in this limit. Using (10), it is not difficult to obtain the following equation for c_* :

$$\frac{c_*^\omega}{1-c_*} = g \frac{P_2^\omega}{P_1}, \quad \omega \equiv v_1/v_2,$$

$$g \equiv P_1^0 (P_2^0)^{-\omega} e^{\frac{v_1(P_2^0 - P_1^0)}{kT}}. \quad (16)$$

Taking an interest only in qualitative picture let us put $\omega = 1$. Then one follows from (16):

$$c_* = \frac{(P_2/P_1)g'}{1 + (P_2/P_1)g'}. \quad (17)$$

So, at $N_2 \rightarrow 0$, or, what is the same, at $P_2 \rightarrow 0$, we have $c_* \rightarrow 0$, i.e. the saddle point "drives down" to the n -axis.

Further, in the same approximation, $v_1 = v_2 \equiv v$, one obtains from eq. (10):

$$h_{11}^{(n,c)} = -\frac{2}{9} \alpha v^{2/3} n_*^{-4/3}, \quad h_{22}^{(n,c)} = \frac{kTn_*}{c_*(1-c_*)},$$

$$h_{12}^{(n,c)} = \Delta\chi + kT \ln \frac{c_*}{1-c_*}, \quad (18)$$

where $\Delta\chi \equiv \chi_2 - \chi_1$.

From these expressions one follows: $h_{22}^{(n,c)} \rightarrow \infty$ at $c_* \rightarrow 0$, and $(h_{11}^{(n,c)})^2 / h_{22}^{(n,c)} \rightarrow 0$, i.e. $\det \mathbf{H}^{(n,c)} / h_{22}^{(n,c)} \rightarrow h_{11}^{(n,c)}$; $h_{11}^{(n,c)}$ has the same form as in the one-dimensional theory. Thus, this transition formally proceeds in accordance with the general way, $h_{22}^{(n,c)} / kT \rightarrow \infty$, however, at $c \rightarrow 0$, i.e. this is the transition to unary nucleation in the framework of the saddle-point theory.

In closing, let us find the contours of constant ratio $f_s / f_0 = r$ [10] using $f_s(n_1, n_2)$ from eq. (7). Applying the expansion $erf(x) \approx (2/\sqrt{\pi})x$, we find that in the vicinity of the saddle point the following equality holds:

$$\frac{f_s}{f_0} = \frac{1}{2} \left[1 - \frac{2}{\sqrt{\pi}} (a_1 n_1 + a_2 n_2) \right] = r,$$

$$a_1 = -\frac{1}{\sqrt{2kT|\kappa|(1+tg^2(\theta))}} (h_{11} + h_{12}tg\theta),$$

$$a_2 = -\frac{1}{\sqrt{2kT|\kappa|(1+tg^2(\theta))}} (h_{12} + h_{22}tg\theta). \quad (19)$$

From this equation one obtains:

$$n_2 = -\frac{a_1}{a_2} n_1 + \frac{\sqrt{\pi}}{2a_2} (1-2r). \quad (20)$$

So, we have the set of parallel straight lines (for different r) with slope tangent

$$tg\tau = -\frac{a_1}{a_2} = -\frac{h_{11} + h_{12}tg\theta}{h_{12} + h_{22}tg\theta}. \quad (21)$$

As it was stated above, in the limit $\gamma = d_{22}/d_{11} \rightarrow 0$, $tg\theta \rightarrow -h_{11}/h_{12}$. It follows from (21) that $tg\tau \rightarrow 0$ in this case. In other words, in the case of slow kinetics of the second species the contours of $f_s / f_0 = r$ are almost parallel to the n_1 -axis. So, the theory confirms the results obtained by the authors of Ref. [10] via numerical solution of binary-nucleation equations.

CONCLUSIONS

At first glance it would seem that processes of binary (multicomponent) nucleation and those with linked fluxes are physically different. However, the passage to the variables (total number of monomers, composition) converts binary nucleation into a process with linked fluxes. So, we conclude that the latter is the general case of nucleation processes.

Representation of binary nucleation as a process with linked fluxes makes it possible to normalize the equilibrium DF and thereby to calculate correctly the stationary nucleation rate and size distribution of nuclei. Also, it allows us to investigate the transitions to the one-dimensional nucleation.

The normalization factor for the equilibrium DF has been obtained in the framework of this approach. The nucleation rate value does not depend on the variables used, as it must from the physical point of view.

The use of the variables (total number of monomers, composition) makes it possible to reveal and correctly describe all the limits, both the kinetic and thermodynamic ones. Two one-dimensional limits for binary nucleation have been described here. One of them corresponds to the formation of nuclei with stoichiometric composition. The second limit is the transition to unary (single-component) nucleation.

The results of the theory which concern the steady state DF confirm the results of numerical solution of the binary-nucleation equations given in literature.

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О КИНЕТИКЕ БИНАРНОЙ НУКЛЕАЦИИ

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Решены две задачи теории бинарной нуклеации: нормировка равновесной функции распределения зародышей и корректный переход в одномерную теорию. Проведена классификация многомерных процессов зарождения и показано, как представить бинарную нуклеацию в виде процесса со связанными потоками посредством соответствующего преобразования переменных описания зародыша. Именно использование переменных (полное число мономеров, состав) позволяет решить данные задачи. Описаны два перехода в одномерную нуклеацию. Один из них соответствует образованию зародышей стехиометрического состава. Другой есть переход в однокомпонентную нуклеацию.

ПРО КІНЕТИКУ БІНАРНОЇ НУКЛЕАЦІЇ

М.В. Алексеечкин

Вирішено дві задачі теорії бінарної нуклеації: нормировка рівноважної функції розподілу зародків і коректний перехід в одновірну теорію. Проведено класифікацію багатомірних процесів зародження і показано, як перетворити бінарну нуклеацію в процес зі зв'язаними потоками за допомогою відповідного перетворення перемінних опису зародка. Саме використання перемінних (повне число мономерів, склад) дозволяє вирішити дані задачі. Описано два переходи в одновірну нуклеацію. Один з них відповідає утворенню зародків стехіометричного складу. Другий є перехід в однокомпонентну нуклеацію.