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The Effect of Elastic Strains and Depletion on Nucleation and Growth in Binary Solutions

As discussed in preceding papers /1,2/ elastic strains quantitatively and qualitatively modify the kinetics of growth of the supercritical clusters and the cluster evolution in the late stage of Ostwald ripening.

In this paper we restrict ourselves mainly to the initial stage of the phase transition including the formation of clusters and their growth up to an overcritical size.

The investigations are based on a thermodynamic analysis presented in the foregoing papers /3,4,5/.

1. Rate equation

We consider a binary system with the thermodynamic constraints

$$n = n_1 + n_2 = \text{const.}, \quad p = \text{const.}, \quad T = \text{const.} \quad (1.1)$$

where n is the total mole number in the system consisting of two components, p the external pressure and T the temperature. The molar fraction of the initial stage is introduced as $x = n_2/n$. In the heterogeneous state the total mole number is divided into the mole number of the cluster phase, denoted by α , and the mole number of the matrix phase indicated by β . Assuming as before /4,6,7/, that the clusters are formed only by particles of component 2 it yields $n_2 = n_{2\beta} + n_\alpha = \text{const.}$ and we may write for the molar fraction of the matrix:

$$x_\beta = \frac{n_{2\beta}}{n_\beta} = \frac{nx - n_\alpha}{n - n_\alpha} \quad (1.2)$$

The cluster phase shall be spatially distributed in clusters with the particle number j ($j \geq 2$)

$$n_\alpha = N_A^{-1} \sum_{j=2}^N j N_j, \quad (1.3)$$

It means that the most important contributions to the change of x_c with temperature are the temperature dependence of c_{2eq} and the dependence of x_p on x reflecting the influence of the total number of particles n .

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N_j being the number of clusters of size j and N_A the Avogadro number. Denoting the particle number of component 2 of the matrix by N_1 (that means free particles) it yields

$$n_{2\beta} = N_A^{-1} N_1 \quad (1.4)$$

and we obtain the following continuous equation resulting from the conservation of particles in the system /8/

$$\frac{\partial N_1}{\partial t} = - \frac{\partial}{\partial t} \sum_{j=2}^N j N_j \quad (1.5)$$

The change of the number of clusters of size j with time can be described by a rate equation. Assuming like in classical nucleation theory /9/ that the growth of clusters occurs only by the attachment of free particles, neglecting collisions of larger clusters, and assuming further that the clusters shrink detaching single particles, the rate equation can be written in the form:

$$\frac{\partial N_j}{\partial t} = I_{j-1} - I_j \quad j = 2, 3, \dots, N \quad (1.6)$$

I_j is the netto rate of formation of clusters of size j . It depends on time because the actual number of clusters of size j as well as the transition rates w^+ and w^- both depend on time:

$$I_j(t) = w_j^+(t) N_j(t) - w_{j+1}^-(t) N_{j+1}(t) \quad (1.7)$$

Eq. (1.6) represents a hierarchy of differential equations which must be solved successively to know the actual cluster distribution $N_i(t)$ ($i=1, 2, \dots, N$). For a calculation we have to determine first the transition rates w^+ and w^- .

2. Equilibrium cluster distribution

In order to determine the transition rates we make use of the condition of detailed balance /8/. That means for finite systems in equilibrium the netto rate I_j (eq. 1.7) must vanish for every j , resulting in:

$$w_j^+ N_j^0 = w_{j+1}^- N_{j+1}^0 \quad (2.1)$$

N_j^0 denotes the equilibrium cluster distribution which can be obtained from a mass action law by:

$$N_j^0 = N_1 \exp \left\{ - \frac{\Delta G_j}{k_B T} \right\} \quad (2.2)$$

$N_1 = N_A \cdot n_2$ gives the normalization. ΔG_j is the Gibbs free energy to form a cluster of size j (not the thermodynamic potential of the whole system). It is given by /9/

$$\Delta G_j = - \Delta g \cdot j + \sigma \cdot j^{2/3} \quad (2.3)$$

The first term of eq. (2.3) describes the binding energy in the cluster (bulk phase), the second term the surface energy caused by the formation of a (spherical) surface with the surface tension σ . The binding energy per particle in the cluster can be approximated classically by /9/:

$$\frac{\Delta g}{k_B T} = \ln \frac{cx}{c_{2eq}(T)} \quad (2.4)$$

Here c is the total particle density in the system being constant, x is the initial molar fraction of component 2 and $c_{2eq}(T)$ is the equilibrium concentration of component 2 in the matrix depending on temperature by /10/

$$c_{2eq}(T) = c_{2eq}(T_0) \exp \left\{ \frac{q}{k_B T} \left(\frac{1}{T_0} - \frac{1}{T} \right) \right\} \quad (2.5)$$

q means the solution heat per particle.

The ratio cx/c_{2eq} is a measure of the supersaturation in the classically infinite system.

For the finite system we have to consider firstly the depletion of the matrix, that means, x must be replaced by the actual molar fraction x_B /6,7/. Secondly, the existence of elastic strains caused by the cluster formation in the matrix decreases the effective supersaturation by a certain amount ϵv_{2d}^0 , as has been discussed in the foregoing paper /4/. ϵ is a measure for the elastic strains. Assuming strains of a Nabarro-type /5/, ϵ depends only on the elastic modul, the Poissonian number, the molar volumes of the pure components 1,2, of the phases α and β (see /4/). v_{2d}^0 is the molar volume of component 2 in the cluster. With respect to depletion and elastic strains in the con-

sidered system we introduce now instead of eq. (2.4):

$$\frac{\Delta g^e}{k_B T} = \ln \frac{c x_B}{c_{2eq}(T)} - \varepsilon \frac{v_{2d}^0}{k_B T} \quad (2.6)$$

Considering the temperature dependence of c_{2eq} (eq. 2.5) we arrive at:

$$\frac{\Delta g^e}{k_B T} = \ln \frac{c x_B}{c_{2eq}(T_0)} - \frac{q}{k_B T} \left(\frac{T - T_0}{T_0} \right) - \frac{\varepsilon v_{2d}^0}{k_B T} \quad (2.7)$$

In the following Δg^e (eq. 2.7) gives a measure of the effective actual supersaturation in the system in contrast to the initial supersaturation (eq. 2.4).

The formation energy ΔG_j (eq. 2.3) has got a maximum for the critical cluster size $r \sim j^{1/3}$:

$$r_{cr} = \frac{2 \sigma v_{2d}^0}{k_B T} \left(\ln \frac{c x_B}{c_{2eq}(T)} - \frac{\varepsilon v_{2d}^0}{k_B T} \right)^{-1} = \frac{2 \sigma v_{2d}^0}{\Delta g^e (k_B T)^2} \quad (2.8)$$

This equation for the critical radius r_{cr} agrees obviously with the equilibrium condition derived before /4/ for the considered system with respect to elastic strains.

3. Transition rates

Because the equilibrium cluster distribution N_j^0 is known, now, we need a kinetic assumption for one of the transition rates w^+ , w^- in order to determine the transition rate for the opposite process from the condition of detailed balance /8/. We decide for an ansatz for the transition rate of attachment of free particles.

The Fickian law gives a relation between the flux of free particles through a spherical surface and the gradient of the concentration c_1 of free particles:

$$\frac{dN_1}{dt} = - D 4\pi R_j^2 \left. \frac{dc_1(r)}{dr} \right|_{r=R_j} \quad (3.1)$$

Here R_j means the radius of the spherical cluster, r is the distance from the cluster and D the diffusion constant of free particles of component 2 in the matrix consisting of components

1 and 2. The gradient $\partial c_1 / \partial r$ must be determined from the diffusion equation

$$\Delta c_1(r) = 0 \quad (3.2)$$

We assume the boundary conditions

$$1. c_1(r \rightarrow \infty) = c_{2\beta} \quad 2. c_1(r=R_j) = 0 \quad (3.3)$$

The first condition reflects that the concentration of free particles apart from the cluster agrees with the actual concentration of component 2 in the matrix. The second condition means that all free particles which arrive at the cluster surface will be bound in the cluster - that is the meaning of the transition rate of attachment. With eqs. (3.3) we find from eq. (3.2):

$$c(r) = c_{2\beta} \left(1 - \frac{R_j}{r}\right) \quad (3.4)$$

Inserting this result into eq. (3.1) we arrive at:

$$\frac{dN_j}{dt} = -D \, 4\pi R_j \, c_{2\beta} \quad (3.5)$$

It seems to be sensible now to make the following ansatz for w_j^+ in agreement with the solution (3.5) of the Fickian law:

$$w_j^+ = \alpha D \, 4\pi r_j \, c_{2\beta} \quad (3.6)$$

$c_{2\beta}$ means the actual concentration of free particles of component 2 in the matrix which form the clusters, $r_j \sim j^{1/3}$ is the cluster radius and α is a constant which scales the time and should reflect the properties of the surface, like surface tension, sticking coefficient and so on. Using the condition of detailed balance (eq. 2.1) with respect to eqs. (2.2), (2.3), (2.6) we find the transition rate for the detachment of free particles from the cluster in the form:

$$w_j^- = \alpha \, 4\pi D \, r_j \, c_{2eq} \exp \left[\left(\frac{2\sigma}{3k_B T} \right) j^{-1/3} \right] \left(\frac{\epsilon v_{2d}^0}{k_B T} \right) \quad (3.7)$$

We choose for the time the unknown parameter α to be arbitrary equal to one and introduce the capillary length $d_0 = 2\sigma v_{2d}^0 / k_B T$ and the equilibrium concentration of free particles above the curved cluster surface by

$$c_{eq}(r_j) = c_{eq2}(T) \exp \left\{ \frac{d_0}{r_j} \right\} \quad (3.8)$$

After a division of the eqs. (3.6) and (3.7) by the constant $\exp(-\xi v_{2d}^0/k_B T)$ the transition rates to describe the cluster growth and shrinkage with respect to elastic strains and to the depletion of the matrix are obtained finally in the form:

$$\begin{aligned} w_j^+ &= 4\pi D r_j c_{xp} \exp \left\{ -\frac{\xi v_{2d}^0}{k_B T} \right\} \\ w_j^- &= 4\pi D r_j c_{eq}(r_j) \end{aligned} \quad (3.9)$$

In contrast to a classical description the transition rate of attachment of free particles depends on the whole cluster distribution because of the dependence on x_p . This fact has been obtained also in isochoric gases /8,11/. Because we have in the considered case a diffusion-controlled cluster growth (remember eq. (3.3) and $d=1$) and not an interface-controlled growth like in isochoric gases, w^+ is here only proportional to r_j and not to r_j^2 . Moreover, it becomes clear that elastic strains decrease the transition rate of cluster growth to a certain extent and the clusters will form slower.

4. Deterministic growth equation for supercritical clusters

It is known from stochastic simulations of the nucleation process /8/ that for clusters with an overcritical size the probability to shrink becomes smaller compared with the one describing the further growth. Therefore, the evolution of overcritical clusters can be well described by a deterministic growth equation which neglects the probability of shrinkage of supercritical clusters. For the evolution of the cluster distribution then the following Liouville equation is held /12/

$$\frac{\partial N_j}{\partial t} = -\frac{\partial}{\partial j} (N_j v_j) + I_j(t) \quad (4.1)$$

Here I_j denotes a source term which agrees in the considered case with the netto rate (eq. 1.7) of cluster formation. The first term of the r.n.s. reflects the deterministic cluster growth and shrinkage. v_j means the deterministic velocity which can be derived from /12/

$$v_j = \frac{dj}{dt} = w_j^+ - w_j^- \quad (4.2)$$

Inserting the known transition rates w^+ , w^- (eq. 3.9) it yields for the considered case:

$$v_j = 4\pi D r_j \left[c x_p \exp\left(-\frac{\xi v_{2d}^0}{k_B T}\right) - c_{2eq} \exp\left(\frac{d_0}{r_j}\right) \right] \quad (4.3)$$

Use of power expansion for the exponential functions up to the first order leads to:

$$v_j = 4\pi D r_j c_{2eq} \left[\ln \frac{c x_p}{c_{2eq}(T)} - \frac{\xi v_{2d}^0}{k_B T} - \frac{d_0}{r_j} \right] \quad (4.4)$$

Finally, we introduce the critical cluster size given by eq. (2.8) and arrive at the deterministic growth equation:

$$v_j = 4\pi D d_0 c_{2eq}(T) r_j \left(\frac{1}{r_{cr}(t)} - \frac{1}{r_j} \right) \quad (4.5)$$

In eq. (4.5) the critical radius $r_{cr}(t)$ (eq. 2.8) acts as an selection value. Only clusters with a radius r_j larger than r_{cr} are able to grow, clusters with a size $r_j < r_{cr}$ must shrink and diminish again. In contrast to classical nucleation theory the critical radius depends on time now, because x_p depends on time. It increases from an initial value

$$r_{cr}(t=0) = d_0 \left(\ln \frac{c x}{c_{2eq}} - \frac{\xi v_{2d}^0}{k_B T} \right) \quad (4.6)$$

which agrees with r_{cr} for the classical infinite system, up to the value of the stable cluster size (compare Fig. 2 in the foregoing paper /4/) and v_j becomes equal to zero in stable equilibrium.

Note that eq. (4.5) is related to known equations describing the selection of different species in self-organizing systems /5,13/. Deterministic equations for crystal growth in visco-elastic and elastic media derived from a quite different ansatz but in conclusion similar to eq. (4.5) are given in /1,2/.

In order to solve the kinetic equations (1.6), (1.7), (4.1) for nucleation and growth of clusters we use now the following boundary conditions:

- (i) $N_j(t=0) = 0$ for $j \geq 2$; that means initially only free particles of component 2 exist in the system.
- (ii) Clusters with a size $j > j^* > j_{cr}$ are no longer considered in terms of the rate equation. Classically their further growth will be neglected. We describe the cluster growth for $j > j^*$ by a deterministic growth equation (4.1) where for $j = j^*$ the source term is given by the netto rate $I_j(t)$ (eq. 1.7). For $j > j^*$ the source term results from the cluster growth only.
- (iii) $N_j(t) = N_j^0$ for $j < u$, that means the cluster distribution is equal to the equilibrium distribution (eq. 2.2) up to a small cluster size u (about five particles).
- (iv) The conservation of the total particle number of component 2 $\sum_{j=1}^u jN_j = \text{const.}$ is always fulfilled.

The given kinetic equations are applied now to cluster growth in a solid solution. The parameters are related to a silver halogenide/natrium borate solution where AgCl-clusters are formed, but we are not engaged here in a comparison between theory and experiment. Only the initial stage of the phase transition is investigated. For results in comparison with measurements for larger times we refer the reader to refs. /1,2,14/.

5. Solution of the kinetic equations

The investigation of nucleation and growth in the solid solution shall demonstrate

- (i) the influence of the depletion of the matrix and
(ii) the influence of elastic strains

on the kinetics of phase transition during the first stage.

(i) The effect of depletion is demonstrated in Fig. 1 which presents the netto rates of cluster formation in the limit of vanishing elastic strains ($\epsilon = 0$). Curve a is obtained considering a phase transition without any depletion as assumed in classical nucleation theory. In this case a stationary nucleation rate is established ($I_j = I_{j+1} = \dots = I^s = \text{const.}$) which can be approximated by /9/:

$$I^s = \left\{ \sum_{j=1}^{j^*} \frac{1}{w_j^+ N_j^0} \right\}^{-1} \approx I_0 \exp \left\{ \frac{4\pi}{3} \frac{\sigma}{k_B T} r_{cr}^2 \right\} \quad (5.1)$$

I_0 is a constant (about $10^{31} \text{ m}^{-3} \text{ s}^{-1}$) which is derived in classical nucleation theory /9/. r_{cr} means the critical radius being classically constant because the depletion is neglected. It is obtained from eq. (4.6) keeping in mind that in Fig. 1 $\xi = 0$. In a quasi-stationary approximation /5,15/ the classical formulae for the nucleation rate (eq. 5.1) is used, but now taking into account the depletion of the matrix by the already formed clusters. This results in a time dependence of the critical radius which is now given by eq. (2.8) with $\xi = 0$. Curve b (Fig. 1) presents the quasi-stationary nucleation rate. Caused by the depletion, a slight decrease of I is obtained in the initial stage of the phase transition.

But the quasi-stationary approximation still neglects the further growth of the already formed clusters. This effect increases the depletion of the matrix considerably, it predominates the depletion caused by the cluster formation only. Therefore, the nucleation process cannot longer be described as a stationary process. Instead of approximation (5.1) we now have

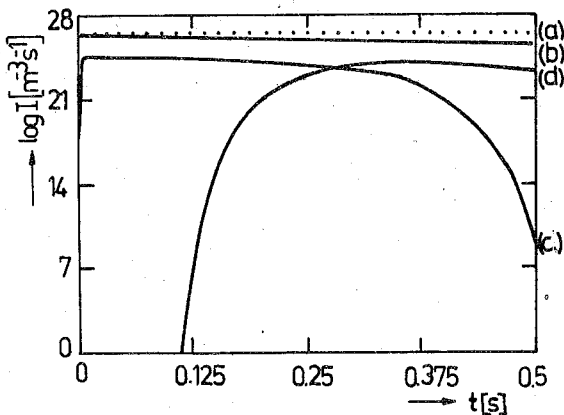


Fig. 1: Rate of cluster formation I (clusters/ m^3s) in dependence on time (s)

(a) classical nucleation rate (eq. 5.1), (b) quasi-steady state approximation, (c) $I(j_{cr})$ (eq. 1.7), (d) $I(j=250)$ (eq. 1.7)

The parameters are related to a silver halogenide/sodium borate solution, $c = 2.1 \cdot 10^{29} \text{ m}^{-3}$, $x = 0.02$, $T = 730 \text{ K}$, $\xi = 0$

to solve the system of rate equations (1.7) because both nucleation of critical clusters and growth of supercritical clusters depend reciprocally and should be described simultaneously /15/. The nonstationary nucleation rate of critical clusters (cf. also /16/) is given in curve c (Fig. 1). After a certain time lag, known already from classical nucleation theory, $I(j_{cr})$ is nearly constant in a small range of the time - only for this range a steady state approximation should be satisfied.

But we note that I_{cr} is always smaller than the value given by the classical formulae. This fact can be understood only from a kinetic point of view: Because we start with a distribution of only free particles first during the time lag a metastable equilibrium cluster distribution is established which decreases in a very short time the initial supersaturation to a certain value /8/ (compare Fig. 3). Only this value gives the real "initial" supersaturation for the nucleation of critical clusters. The classical nucleation theory ignores this relaxation into the metastable state for a calculation of I^s . Curve c shows that $I(j_{cr})$ after a small time decreases rapidly caused by the depletion. Keep in mind, that in the same time the critical cluster size j_{cr} is increased, too.

Curve d of Fig. 1 gives the nucleation rate to form clusters with 250 particles. $I(j=250)$ is used to be the source term for the growth equation (4.1). Clusters with a size $j > 250$ are always supercritical for the considered times ($t \leq 0.7$ s), therefore, their further growth here is described by the deterministic growth equation. It is shown that the time lag to form clusters of 250 particles becomes much greater. As discussed before, also for $I(j=250)$ no stationary value could be obtained - after a certain time it decreases again (not clearly to be seen in Fig. 1.).

The largest cluster obtained in the system after $t = 0.64$ s has got a size of 1.9 nm that means nearly 2400 particles. It becomes clear that such a decrease of the free particles in the system should be not ignored in the initial stage of the phase transition.

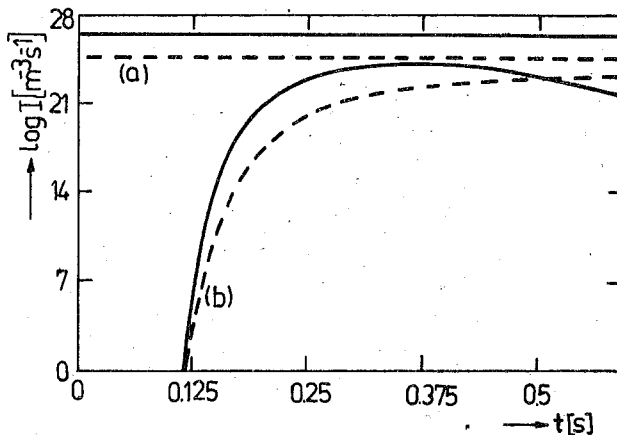


Fig. 2: Rate of cluster formation I (clusters/ m^3s) in dependence on time (s)
 (a) classical nucleation rate (eq. 4.1), (b) $I(j=250)$ (eq. 1.7)
 solid lines: $\epsilon = 0$, dashed lines: $\epsilon = -6.1 \cdot 10^7 Jm^{-3}$
 For the parameters see Fig. 1.

(ii) In order to investigate the influence of elastic strains we calculate both the classical nucleation rate $I^s = \text{const.}$ and the rate $I(j=250)$ for two values of ϵ . The solid lines in Fig. 2 give the nucleation rates without elastic strains, the dashed lines consider elastic strains. The curves (a) represent the classical steady-state nucleation rate (eq. 5.1). It is shown that elastic strains decrease I^s considerably because the effective supersaturation $\Delta g^e/k_B T$ (eq. 2.7) becomes smaller in dependence on ϵ . In contrast to (a) the curves (b) give the non-stationary nucleation rate $I(j=250)$ obtained from a solution of the system of rate equations (eqs. 1,6, 1.7). After the time lag the netto rate for $\epsilon = 0$ increases faster, but also decreases earlier than the rate in presence of ϵ . This fact must be understood also kinetically:

Fig. 3 gives the effective supersaturation (eq. 2.7) in dependence on time. Because the initial concentration of free particles in the system, c_x , is equal in both cases, the existence of elastic strains decreases the initial supersaturation compared with $\epsilon = 0$. A lower supersaturation relaxes slower into its equilibrium value. This fact has been discussed also for

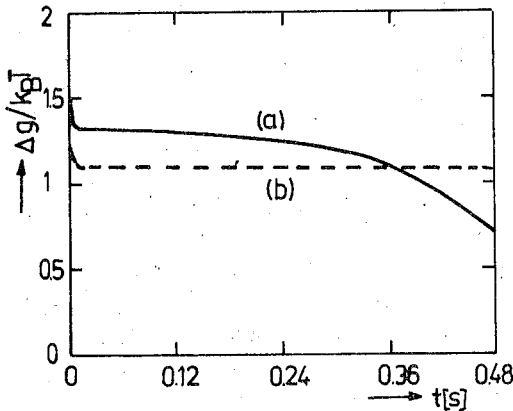


Fig. 3:
Effective supersaturation
 $\Delta g / k_B T$ (eq. 2.7) in dependence on time t (s),
(a) $\xi = 0$, (b) $\xi = 6.1 \cdot 10^7 \text{ Jm}^{-3}$
For the parameters see Fig. 1.

stochastic simulations of the nucleation process /8,5/. It becomes clear that after a certain time the supersaturation for $\xi = 0$ is more decreased because it reduces faster and the nucleation rate becomes smaller compared with $\xi > 0$. That means the period of formation of clusters becomes longer for systems where elastic strains exist, because the relaxation processes proceed slower.

We summarize the results for the kinetics of phase transition obtained for the first period, now, as follows:

1. In finite systems a depletion of the matrix is obtained caused by the formation of new clusters and the growth of already formed clusters. Therefore, the critical radius of cluster formation depends on time.

2. Since this depletion cannot be neglected, a nonstationary nucleation rate must be calculated from a system of rate equations. A deterministic growth equation describes the further growth of the already formed supercritical clusters.

3. The nucleation process of formation of critical clusters starts with a supersaturation smaller than the initial supersaturation, because the system first relaxes in a metastable equilibrium state.

4. Elastic strains lead to a decrease of the supersaturation. Smaller supersaturations relax slower into the equilibrium value, therefore, the nucleation period becomes longer in the presence of elastic strains.

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