Different approaches to describe depletion regions in first order phase transition kinetics

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During the first order phase transitions the objects of a new phase consume a mother phase which leads to some density gaps near existing embryos of a new phase. This fact is rather obvious [1], but complete theoretical description was given only in [2]. Now this fact is widely recognized [3] but still there are some attempts [4] to reconsider this approach which is now known as the model of depletion zones [3]. Here we shall analyze [4] and show the useless of reconsideration proposed in [3].

We don’t consider here the picture of the global evolution of a system - in [4] it is so primitive that can not give any reliable quantitative results but only some very rough estimates without any justifications. We needn’t to do it because this has been already done in [2] with all necessary justifications. So, we consider only the profile around the solitary embryo, which was the matter of interest in [4]. Why it is possible to consider only the solitary embryo? An answer has been given in [2], in [4] this question remains unsolved. Due to results of [2] we shall consider namely the solitary embryo.

We assume that the substance exchange regime is a diffusion one. Then there is a gap of a vapor density $n$ near an embryo. Then the density $n$ differs from a density $n(\infty)$. This difference initiates a variation in a nucleation rate $I$ and causes the effect of depletion zones. So, it is very important to get for $n(r) - n(\infty)$ ($r$ is a distance from a center of an embryo) a correct expression. This expression is a crucial point in consideration of an effect of depletion zones in nucleation kinetics. Certainly, it has to be a function of time $t$.

In [4] the following expression is proposed

$$ n(r, t) = n_s(r) + (n(\infty) + n(0)) \frac{R}{r} \Phi \left( \frac{r - R}{2\sqrt{Dt}} \right) $$

(1)
\[ n_s(r) = n(\infty) - \frac{n(\infty) - n(0)}{r} R \]  \hspace{1cm} (2)

Here \( n(0) \) is equilibrium concentration on the surface of nucleus, \( D \) is a diffusion coefficient, \( R \) is the nucleus radius, \( \Phi(x) \) is the Laplace probability integral.

Here the sign + was used instead of + in the second term of (1). Correct solution in the neighbourhood of an embryo is (3)

\[
\frac{n - n(\infty)}{n(0) - n(\infty)} = \frac{R}{r} \text{erfc}(\frac{r - R}{2\sqrt{Dt}}), \quad \text{erfc}(z) = \frac{2}{\sqrt{\pi}} \int_{z}^{\infty} \exp(-\xi^2)d\xi \]  \hspace{1cm} (3)

But even with this solution one can not construct the correct form of the nucleation rate profile. We shall outline this point.

There are following necessary suppositions to get this exact solution for concentration profile:

- The radius of an embryo is constant in time
- The boundary condition at \( r = R \) is constant in time.

Both assumptions are rather approximate. As a result the above solution is valid only near the surface of an embryo. It is easy to see from the following arguments. Let \( \delta t \) be the characteristic time when the relative variation of intensity of vapor consumption \( v \) is already essential

\[ |v(t + \delta t) - v(t)| \sim v(t) \]

Then it follows that only the distances

\[ r - R \sim 4D\delta t \equiv \delta R \]

or smaller ones can be considered on the base of solution (3). At all distances \( r - R > \delta R \) we have to take into account that earlier the intensity of vapor consumption was smaller. Really

\[ v \sim \frac{d\nu}{dt} \]

where \( \nu \) - a number of molecules inside the embryo. As it is known

\[ \frac{d\nu}{dt} = \text{const} \, \nu^{1/3} \sim t^{1/2} \]
and we see that the intensity isn’t constant in time.

It seems that the small distances are the main ones. Certainly the gap is greater the smaller the distance is. But the small distances aren’t essential because the functional form of the nucleation rate \[2\] can be approximated as

\[
I(r) = I(r = \infty) \exp\left(\Gamma(n(r) - n(\infty))/n(\infty)\right)
\]

where \(\Gamma\) is a big parameter. It is approximately equal to \(\nu\). Note, that the sense of nucleation rate as a probability to appear for an embryo was used here (details see in \[2\]). So, the distances where

\[-\Gamma(n(r) - n(\infty))/n(\infty) \approx 1\]

are the most interesting. When

\[|\Gamma(n(r) - n(\infty))/n(\infty)| \ll 1\]

we have \(I = I(\infty)\) and the is no gap of concentration. When

\[-\Gamma(n(r) - n(\infty))/n(\infty) \gg 1\]

we have \(I = 0\) and the is no nucleation. So, this region isn’t interesting.

The distances \(\Gamma(n(r) - n(\infty))/n(\infty) \approx 1\) because of \(\Gamma \sim \nu \to \infty\) corresponds to very small \(n(r) - n(\infty)\) and, thus, to big values of distances \(\sim \delta r\) from the embryo. Ordinary \(\delta r \gg \delta R\) and there is no possibility to use \(3\).

Instead of \(3\) in \[2\] another approach was used. This approach is based on the strong inequality

\[\delta r \gg R\]

Why this inequality takes place? When we consider classical nucleation, i.e. transition of a supersaturated vapor into a liquid phase everything is clear. We have a strong inequality

\[v_v/v_l \gg 1\]

where \(v_v\) is a partial volume for one molecule in a vapor, \(v_l\) is a partial molecule in an embryo. This inequality makes obvious that the final volume after the whole process of condensation is small. As an example of condensation of water vapor in normal external conditions. Suppose that supersaturation is somewhere \(\sim 5\). Then because \(v_v/v_l \sim 1000\) we see that
the final volume of a new phase is very small $\sim 1/200$ and the characteristic distance $\delta r \sim (200)^{1/3} R \gg R$.

We see that the value $\delta r$ has to take the same order of magnitude as the mean distance between objects of a new phase $r_{\text{mean}}$. Really, when $\delta r$ attains imaginary value $r_{\text{mean}}$ it means that the process of nucleation stops. So, namely the values at this moment are the final ones and the main ones. Then

$$r_{\text{mean}} \sim \delta r$$

The same will be valid for all other first order transitions.

Consider the opposite transition from the liquid to the vapor phase. We shall see the opposite inequality. But we know that compressibility of liquid is very low. So, the relative super stretching is rather low. It means that the vapor phase in a final state will occupy some rather small volume. And again we see that the mean distance between the objects of a new phase is many times greater than the size of objects. Again we get the same result.

We see that the following fact takes place: The relative final volume of a new phase will be small.

To see that (5) takes place we needn’t to consider the real final volume of a new phase after phase transition but can take the values at the end of nucleation. Because the relative quantity of surplus substance condensed in a new phase has an order $\Gamma^{-1}$ (at least under the collective regime of substance consumption [2]) we see that the relative volume occupied by a new phase is limited from above by a value of an order $\Gamma^{-1}$. So

$$r_{\text{mean}} \sim R\Gamma^{1/3}$$

and the required property is established.

So, we see the required property for moderate effects of depleted zones. When the effect of depletion zones is strong we can use at $r \sim (2 \div 3) R$ stationary solution and see that $\delta r > (5 \div 7) R$. So, here the required property is also observed which completes justification.

Then we can use profiles obtained on the formalism of Green function. We have [4]

$$n(\infty) - n(r) = \int_0^t \frac{\lambda x^{1/2}}{8(D\pi(t-x))^{3/2}} \exp\left(-\frac{r^2}{4D(t-x)}\right) dx$$

(6)

$$\lambda = (4\pi)^{3/2} \frac{\nu_l}{2\pi} \left(\frac{\nu_l}{2\pi}\right)^{1/2} (\zeta n_\infty D)^{3/2}$$

(7)
and $\zeta$ is a supersaturation.

This result was obtained for transition from vapor phase into a liquid phase but there is no difference because the kinetic mechanism remains the same.

One can see that expressions (1), (2) and (6), (7) are absolutely different in analytical structure. The same is valid when we compare (1), (2) and (6), (7) numerically.

On the base of profile we can easily get all results of the nucleation process.

The rate of nucleation can be found according to (4). Then we have

$$I(r) = I(r = \infty) \exp(\Gamma n(0) - n(\infty)\frac{R}{r}erf\left(\frac{r - R}{2\sqrt{Dt}}\right)/n(\infty))$$ (8)

for analytical solution with fixed boundary of embryo and

$$I(r) = I(r = \infty) \exp(-\Gamma \int_0^t \frac{\lambda x^{1/2}}{8(D\pi(t - x))^{3/2}} \exp(-\frac{r^2}{4D(t - x)})dx/n(\infty))$$ (9)

for solution on the base of Green functions. Note that the analogous transformation in [4] was done with partial shift of coordinate $r - R$ to $r$ which is wrong and beside this isn’t necessary.

We see that these approaches give absolutely different results.

The next step is an obvious remark that the probability $dp$ that an embryo appears during elementary interval $dt$ at the distance from $r$ up to $r + dr$ from the center of the already appeared embryo is

$$dp = dt 4\pi r^2 I(r, t)$$

Then the probability $dP$ that the embryo appears in the layer from $R$ up to $r$ is

$$dP = dt \int_R^r 4\pi r'^2 I(r', t)dr'$$ (10)

This expression differs from [4] where the following expression was presented

$$dP = dt \int_r^R 4\pi r'^2 I(r', t)dr'$$

The last expression is wrong and we shall follow (10).

The next step used in [4] is to come to an integral value $P$ which is the probability that in a sphere of radius $r$ around already existing embryo there will be no appearance of a new embryo. Certainly we can start from (10).
and integrate it. But it is necessarily to take into account that the zone of depletion will grow according to the absolutely precise law. This law was established in [2]. But here we suppose that one can act in another manner and write

\[ P = \exp\left(-\int_0^t \int_R 4\pi r'^2 I(r', t') dr' dt'\right) \] (11)

The last expression analogous to those which forms the base for further constructions in [4] is rather doubtful. The problem appears because now we are coming to characteristic of a whole process of nucleation (earlier there was a profile around a solitary object of a new phase).

For a ”solitary subsystem” the derivation of (11) is evident: Let

\[ dp = dt I \]

be a probability that during an elementary interval \( dt \) there will be appearance of a new embryo. The value of \( dt \) is rather small. Then \( dp \) is proportional to \( dt \) and to the rate of nucleation \( I \) (here the sense of the nucleation rate as a probability is used). Then the probability for the absence of appearance is

\[ dp' = 1 - dp = 1 - dt I \]

If we have two elementary intervals \( dt_1 \) and \( dt_2 \) which don’t overlap then the probability of the absence of embryo appearance is

\[ dp'_1 dp'_2 = (1 - I_1 dt_1)(1 - I_2 dt_2) \]

The same is valid for arbitrary number of intervals.

To fulfill multiplication in the r.h.s. one can present \( dp' \) as

\[ dp' = (1 - I dt) = \exp(-I dt) \]

Then the total probability of the absence of appearance is

\[ P = \prod_i dp'_i = \exp(-\sum_i dt_i I_i) = \exp\left(-\int I(t) dt\right) \] (12)

and we come to (11).

\(^1\)To be close to [3].

\(^2\)In [4] an integral over \( r \) is from 0 up to \( r \).
The real problem is how to take into account the overlapping of density profiles initiated by different embryos. This problem was solved in [2]. The exponential form (11) for $P$ is some rather arbitrary interpretation of the value “the free volume” used in [2]. Now we shall discuss this interpretation in frames of the approximation of solitary droplet.

The overlapping of exhausted regions (ER) requires to use instead of (8) - (9) another approximation where instead of one profile there is a superposition of profiles. Certainly it is impossible to calculate this superposition precisely. Then one will come to some models analogous to formulated in [2].

One can directly use the results from [2] here. When the free volume is calculated as function of time one can determine the total nucleation rate as a ratio of the free volume to the whole volume of a system and use then (12).

The real problem is what consequences can we make from a knowledge of $P$. On one hand it seems that the knowledge of $P$ solves all problems in kinetics of the global nucleation stage. Really, having presented $P$ in a form

$$P = \exp(-L(r, t))$$

where $L(r, t)$ is some expression one can approximately estimate the average radius of a sphere where there will be no appearance by relation

$$L(r, t \rightarrow \infty)|_{r = r_0} = 1$$

This construction analogous to [4] needs two remarks.

This estimate makes no difference between embryos appeared earlier or later during the nucleation stage. It means that this approximation supposes that the spectrum of droplets sizes is monodisperse one. It isn’t correct assumption and can be treated only as a rough estimate.

It is known that sometimes the overlapping of profiles is important and makes the main contribution in nucleation kinetics. This phenomena can occur when the substance exchange regime is going to be a free molecular one and when the long tails of profiles are important. The first situation can not be realized here. The last situation was a matter of separate consideration in [6].

In [2] an integral definition of a boundary of ER allowed to take into account the situation of long tails. Here this situation can not be considered, because the level type definition is used. It means that the value of $r_0$ is determines as a value when $L$ attains some level ($= 1$). The definition of
level type can not take into account long tails and it is more convenient to use approach from [2]. Here this approach is considered because it is analogous to [4].

When the value of \( r_0 \) is determined it is easy to estimate the total average number of droplets \( N \) as

\[
N = \frac{3}{4\pi} r_0^{-3}
\]

(the coefficient \( \frac{3}{4\pi} \) can be omitted, this depends on interpretation of \( r_0 \)).

To fulfill concrete calculations one can use the following approach. Note that \( P \) can be in any case presented as

\[
P = \exp\left(- \int_0^\infty dt' \int_{r_l}^r dr' 4\pi r'^2 \exp(f(r', t'))\right)
\]

The lower limit \( r_l \) of integration has to be put \( r_l = R \) for models with explicit boundary of an embryo and for models with Green function \( r_l = 0 \). But because \( \delta r \gg R \) the effect of the lower limit will be small and we can put \( r_l = 0 \) in all situations. Certainly, for \( P \) due to the exponential form the effect is essential, but when we are calculating the mean distances the essential dependence on \( r_l \) disappears due to \( \delta r \gg R \). In the last equation \( f \) is some function with explicit form given by solution of diffusion equation.

One can see that in \( f(r, t) \) two variables has to appear in combination \( \beta = t/r^2 \) or \( \beta' = t/(r - R)^2 \). The dependence on \( t \) and \( r \) via \( \beta \) is the main one (in special regimes of mother phase consumption there may be dependence on \( t, r \) in another combination, but this dependence will be much more smooth than via \( \beta \)). Because \( \delta R \gg R \) the difference in substitution \( \beta' \) instead of \( \beta \) will be small (not in \( P \), but on mean distances and times).

Then in integration \( \int_0^\infty \) we have to substitute \( dt' \) by \( d\beta' \) which gives

\[
P = \exp\left(- \int_0^r dr' 4\pi r'^4 \text{const}\right)
\]

where \( \text{const} \) comes from

\[
\text{const} = \int_0^\infty d\beta' \beta'^2 \exp(f(r', t'))
\]

because

\[
\int_0^\infty dt' = r'^2 \int_0^\infty d\beta'
\]

It leads to

\[
P = \exp(-r^5 \gamma) \quad (13)
\]
where $\gamma = 4\pi \text{const}/5$ is some constant.

One can show that

$$\int_0^\infty dx \exp(-x^5) \approx 1$$

This approximate equality is very important. Namely this equality allows to determine the mean distance between droplets as $N^{-1/3}$, or as $r_0$ or according to the sense of $P$. Three ways are available. They have to give similar results. This fact is ensured by this approximate coincidence. Then the third way gives

$$\bar{r} = \int_0^\infty dr Pr = \frac{C_0}{2\gamma^{2/5}} \int_0^\infty d\alpha \exp(-\alpha^{5/2})$$

where $C_0$ is the normalizing factor of distribution $P$. One can calculate $C_0$ according to

$$C_0 \int_0^\infty dr \exp(-\gamma r^5) = 1$$

and

$$C_0 = \frac{\gamma^{1/5}}{\int_0^\infty dx \exp(-x^5)}$$

Then

$$< r > = \frac{1}{2\gamma^{1/5}} \frac{\int_0^\infty d\alpha \exp(-\alpha^{5/2})}{\int_0^\infty dx \exp(-x^5)}$$

Because

$$\int_0^\infty d\alpha \exp(-\alpha^{5/2}) \approx 1$$

$$\int_0^\infty dx \exp(-x^5) \approx 1$$

we see that $\bar{r}$ is two times smaller than the mean distance between embryos. It is clear because the mean distance between embryos is two mean distances until the boundary of exhaustion zone $r_0$. So, we observe a coincidence. Certainly, one can calculate integrals more precisely.

The average time of waiting fro the appearance of new embryo near the already existing one is directly and elementary connected with a mean distance between embryos and we needn’t to calculate it separately\(^4\).

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\(^3\)This value is calculated in \(\text{[4]}\) in a wrong way.

\(^4\)The integral in expression for this value is calculated in \(\text{[4]}\) in a wrong way.
As for calculation a quadratic mean deviation from the average distance it is given\(^5\) by
\[
< (r - < r >)^2 > = < r >^2 \left( \frac{I_2 I_0}{I_1^2} - 1 \right)
\]
where
\[
I_i = \int_0^\infty dx x^i \exp(-x^5)
\]
But we have to stress that this result has practically no meaning because as it is stated in [7] the effect of interaction leads to decrease of fluctuation of the total number of droplets. The right numerical value of this effect is given in [3]. Here we have absolutely no interaction between droplets and this result has to be seriously changed due to interaction. An example of such account is given in [9].

To close this question we shall construct now more precise solutions of diffusion equation. They have to be used in (4) and then in (11).

We shall start with the following model. There exists a more general solution [5].

For a domain \( R < r < \infty \) where
\[
n = f(r) \quad \text{at} \quad t = 0
\]
and
\[
n = g(t) \quad \text{at} \quad r = R
\]
the solution of diffusion equation
\[
\frac{\partial n}{\partial t} = D \left( \frac{\partial^2 n}{\partial r^2} + \frac{2}{r} \frac{\partial n}{\partial r} \right)
\]
(this the diffusion equation with spherical symmetry) is [4]

\[
n = \frac{1}{2r\sqrt{\pi Dt}} \int_R^\infty \xi f(\xi) [\exp(-\left(\frac{r - \xi}{4Dt}\right)^2) - \exp(-\left(\frac{r + \xi - 2R}{4Dt}\right)^2)]d\xi
\]
\[
+ \frac{2R}{r\sqrt{\pi}} \int_z^\infty g(t - \left(\frac{r - R}{4Dt}\right)^2) \exp(-\tau^2)d\tau
\]

where
\[
z = \frac{r - R}{2\sqrt{Dt}}
\]
\(^5\)Result in [4] is wrong.
This solution allows to formulate the following approximation. Let $R$ be constant
\[ R = R(t) \]
at the current moment when we want to know the density profile. But the boundary condition $n_b$ at $R(t)$ in the previous moment $t - t'$ will be recalculated according to the stationary solution:
\[ n_b = n(\infty) - \frac{n(\infty) - n(0)}{R(t - t')} R(t) \] (15)
Really the rate of embryo growth is so small that the stationary distribution can be regarded as practically precise one. So, this approximation is practically precise and takes into account the variation of mother phase consumption by an embryo.

Note that diffusion equation
\[ \frac{\partial w}{\partial t} = D \left[ \frac{\partial^2 w}{\partial r^2} + \frac{2}{r} \frac{\partial w}{\partial r} \right] \]
in the system with spherical symmetry can be transformed by substitution
\[ u(r, t) = rw(r, t) \]
to an ordinary one dimensional diffusion equation
\[ \frac{\partial u}{\partial t} = D \frac{\partial^2 u}{\partial r^2} \]
We shall consider this equation in future.

Then we can make a shift $x = r - R(0)$. The problem
\[ w = f(x) \]
at $t = 0$
\[ w = g(t) \]
at $x = 0$ has solution
\[ w(x, t) = \frac{1}{2\sqrt{\pi Dt}} \int_0^\infty \left[ \exp\left(-\frac{(x - \xi)^2}{4Dt}\right) - \exp\left(-\frac{(x + \xi)^2}{4Dt}\right) \right] f(\xi) d\xi \]
\[ + \frac{x}{2\sqrt{\pi}} \int_0^t \exp\left(-\frac{x^2}{4D(t - \tau)}\right) \frac{g(\tau)}{(t - \tau)^{3/2}} d\tau \] (16)
It is also possible to go to variable
\[ \rho = r - R(t) \]
where \( R(t) = At^{1/2} \) is a boundary of an embryo. Then in the diffusion equation beside
\[ \hat{Q}u \equiv \frac{1}{D} \frac{\partial u}{\partial t} - \frac{\partial^2 u}{\partial \rho^2} \]
there appear some additional terms. Having considered \( \hat{Q} \) as a main operator we come to an iteration procedure where all other terms are assumed to be known in a previous approximation (in zero approximation they are omitted). So, at every step of iterations we have to solve a problem
\[
w = f(x) \]
at \( t = 0 \)
\[
w = g(t) \]
at \( \rho = R(0) \) for equation
\[
\frac{1}{D} \frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial \rho^2} + \Phi'(\rho, t) \]
may be with renormalized \( D \). Here \( \Phi' \) is a known (at the previous step) function. After the mentioned shift \( x = \rho - R(0) \) we came to a problem
\[
w = f(x) \]
at \( t = 0 \)
\[
w = g(t) \]
at \( \rho = R(0) \) for equation
\[
\frac{1}{D} \frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial x^2} + \Phi(x, t) \]
with known \( \Phi \). The solution of the last problem is
\[
u = \int_0^\infty G(x, \xi, t)f(\xi)d\xi + \\
\frac{x}{2\sqrt{D\pi}} \int_0^t \exp\left[-\frac{x^2}{4D(t-\tau)}\right] \frac{g(\tau)}{(t-\tau)^{3/2}} d\tau + \int_0^t \int_0^\infty G(x, \xi, t-\tau)\Phi(\xi, \tau)d\xi d\tau \]
(17)
where
\[ G(x, \xi, t) = \frac{1}{2\sqrt{\pi Dt}} \left[ \exp\left( -\frac{(x - \xi)^2}{4Dt} \right) - \exp\left( -\frac{(x + \xi)^2}{4Dt} \right) \right] \]

The last relation solves the problem to construct iterations on every step. Thus, we come to solution of diffusion equation which has to be used in our previous constructions.

1 Numerical results

We have calculated the worst situation when \( v_v/v_l = 10 \) for vapor-liquid transition. Here we have taken \( G = 13 \) which is the minimal value for macroscopic description of the critical embryo. when \( v_v/v_l \) and \( G \) increase the accuracy of approach (6) and partially the accuracy of approach (15). The accuracy of (6) decreases because the error in (6) depends mainly on the property of relative stationarity which has no connection with values of \( \Gamma \). It isn’t so evident that the property \( v_v/v_l \gg 1 \) also has no influence on the error of (6) because here only the relative variation of radius is important. But when \( \Gamma \) increases it means that the main important region of solution now corresponds to the asymptote where the qualitative behavior of (6) is wrong. So, we came to conclusion that it isn’t possible to use (6). The error produced by (6) is seen already at \( \Gamma = 13 \). Figure 1 illustrates the behavior of different profiles in this situation. One can see four lines there. One line is apart from all other and it has the different asymptotic behavior. This is solution (6). Other lines are solutions of diffusion problem and some approximations. One of them (the highest) is thick because actually there are two formally coinciding solutions: precise solution and approximation (15). The coincidence is formal. In the middle of this group one can find numerical solution with prescribed law of droplet radius growth \( R = At^{1/2} \) (it is obtained by linear extrapolation of results of precise solution). The difference between these curve and precise self consistent solution is caused by numerical errors and corrections on radius growth. the lowest curve in this group is approximation (6). One can see that both (6) and (15) give good results. for natural situations with big values of \( \Gamma \) and \( v_v/v_l \) the accuracy will even increase.
Figure 1.
Different approximations for the form of density profile.

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