EVOLUTION OF A GRAIN SYSTEM: FROM EARLY TO LATE STAGES*

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An analytical approach to the d-dimensional grain growth, which is a kind of the heterogeneous nucleation-and-growth phase transformation, is offered. The system is assumed to be driven by capillary forces. Another important operative assumption is that the system evolves under preservation of its hypervolume, which results in considering the process as a random walk in the space of grain sizes. A role of the initial condition imposed on the system behaviour, and how does the system behave upon a prescribed initial state, have been examined. A general conclusion appears, which states that this prescription does not affect the asymptotic system behavior, but may be of importance when inspecting the early-time domain more carefully, *cf.* the Weibull-type initial distribution. This study is addressed to some analogous theoretical descriptions concerning polycrystals as well as bubbles-containing systems. Some comparison to another modelling, in which a crucial role of local material gradients (fluxes) was emphasized, has been attached.

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1. Introduction

Nucleation-and-growth phase transitions belong to a class of the so-called heterogeneous phase transformations. Among quite many of them, one could list such as: grain growth (of normal and abnormal nature), recrystallization, some polymorphic as well as order–disorder phase transitions [1].

Throughout this study we wish to explore some kinetic effects characteristic of the grain growth, preferentially of the normal type [2]. An emphasis will be particularly put on revealing certain crucial points of the influence of Initial Conditions (IC) on the grain growth process studied, which is chosen by the authors as a landmark system. The reason for so doing comes from the fact, that up to now rather a little is known about the relation of how does some IC prescribed influence the overall system behavior [3].

Let us start with short characteristics of what really the grain growth means. For this purpose we may adopt a kind of definition proposed in a review paper by Weaire and McMurry. It can be summarized as follows [4]:

(*i*) grain growth process yields a cellular system of preserved hypervolume;

(*ii*) Grain Boundaries (GB's) are associated with a positive surface energy;

(*iii*) GB's migrate as to lower their energy;

(*iv*) grain structure evolves with time in the direction of increasing grain size;

(*v*) the above points would suffice to model the kinetics of the grain growth in a statistical way, which means, that such important notions, like grain texture and grain structure topology are unfortunately not involved explictly in this description.

From point (*v*) it immediately follows that grain growth as well as other related processes, like soap froth formation or evolution of bubbles-containing systems, can by modeled by the same means. There is, in fact, a long-standing analogy between polycrystals and soap froths, which has been exploited in various contexts, *cf.* [4,5].

As for the mechanism of grain growth, one is readily encouraged to see [2]. At this point, we may quite generally state that there are two types of forces driving the system and/or inhibiting the system evolution. Among a few principal types of driving forces we would just mention a deterministic one, inevitably connected with the surface (line) tension effect (for bubbles, according to the Laplace's law, with a pressure difference), and termed the capillary force. It is known [2], that this force may really be enhanced in a fluctuating system. Another type of forces is named the drag forces,

among which the Zener force, due to pinning, as well as the Mullins force, subjected to thermal grooves, emerging mostly in high-temperature limit, can be mentioned [6, 7].

Looking into the nature of GB's, being always of paramount importance while trying to study thoroughly the phenomenon, one may simply divide them into two basic types. These are as folows:

(*i*) low-angle GB's;

(*ii*) high-angle GB's.

For our description it is sufficient to mention that the first ones, occuring typically in some early stages of the grain growth, are equivalent to GB's of small curvature, whereas the second ones, emerging most probably in a late time zone, do not conform to this type of GB. They, in turn, are attributed to GB's with a big curvature, rather.

The systems mentioned above can be crudely classified by their kinetics, which can be best seen while looking at their asymptotic behavior. They mostly express the physical fact that their characteristic average linear size (grain radius) changes either powerly or logarithmically, except the system ceases to grow [8]. Under such circumstances, it would be interesting what would happen if we put various IC's just for having a look at how does it influence the global temporal system behavior and which are the details of it.

For sure, it would be equally interesting to provide a physically justified rationale for the use of the starting system of equations, *cf.* (1)–(3) in the next section. Let us sketch roughly both a physical motivation as well as mathematical reasoning for keeping the system (1)–(3) at work. First of all, we are of the opinion that we may start with a general kinetic equation of Avrami–Kolmogorov-type, which is customary in the theory of phase transformations [9]. Namely, we begin by stating that the rate of a small change in the average number of grains, designated by $d/dt\, f(\langle v\rangle; t)d\langle v\rangle$ [10], will be of interest. In other words, one is likely to look for the ensemble evolution over a set of states measured in subsequent time instants t. We take an average over the corresponding ensemble of grains, characterized by the distribution $f(\langle v\rangle; t)$, where time t stands for a parameter, and $\langle ...\rangle$ denotes the average over the statistical ensemble. Next, one should notice that the rate has to be proportional just to a small change in the average number of grains, *i.e.* to the overall (local) magnitude characterizing the process, namely $d/dt\, f(\langle v\rangle; t)d\langle v\rangle \propto f(\langle v\rangle; t)d\langle v\rangle$, which is by the way again consistent with philosophy of formulation of the above mentioned kinetic equation [9]. The second observation is a more physically motivated remark, namely, that the proportionality coefficient should be determined by the $(d-1)$-dimensional area of a d-dimensional grain [11–13]. It immediately results in proposing the following "averaged" equation, namely

$d/dt\, f(\langle v\rangle;t)d\langle v\rangle \propto \langle v\rangle^{1-1/d} f(\langle v\rangle;t)d\langle v\rangle$. Expanding the right-hand side of the proposed equation into the Taylor series around (*sic*!) the hypervolume of a single grain, v, neglecting the zeroth-term of the series (it could, by choice, be equal to zero), noticing negligible meaning of its first term, especially, when compared to the next one, and keeping as the most important the second term, one may safely arrive at the system (1)–(3).

The paper is structured in the following way. In the next section we introduce a statistical–mechanical model of grain growth, which for a specific choice of parameters, *i.e.* under constancy of total hypervolume of the system, reduces to the Mulheran–Harding (M–H) model for the random walk in the space of grain sizes. In Sec. 3 we sketch quite thoroughly the method of solving the grain-growth problem, whereas in Secs. 4–6 we provide exact result for the n-th statistical moment of the process, enabling one to include the entire analysis by splitting into both the main characteristics of the initial as well as late evolution stages, and discuss their physical meaning. Last section serves for conclusions and comparative analyses of related growing processes, for which the total hypervolume preservation is unfortunately not assured [14], with a certain comment on some experimental case, coming from physical metallurgy.

2. Mulheran–Harding model in *d*-dimensions

The M–H model [8, 11, 12] is a diffusion-type model which describes the size- and time-dependent evolution of a grain system. It has a form of the continuity equation, namely,

$$\frac{\partial}{\partial t} f(v,t) = \hat{L}_D f(v,t) = D_0 \frac{\partial^2}{\partial v^2} v^\alpha f(v,t) = -\frac{\partial}{\partial v} j(v,t)\,, \qquad v \in [0,\infty)\,, \quad (1)$$

where v is a volume of a grain, D_0 is a constant representing a random walk behavior in grain growth, $f(v,t)$ is the distribution function of grains at time t, *i.e.*, $f(v,t)\,dv$ is a relative number of grains of size in the volume range $[v, v+dv]$ and the parameter α depends on dimension of the system. We have introduced the "diffusion" operator $\hat{L}_D$ defined by the equality in (1). Below, we put $D_0 = 1$ because it enters only in the product $D_0 t$ which rescales time t. Assuming that the net flux of the migrating particles across the grain boundaries is proportional, for three-dimensional systems, to the surface of grains of volume v and, for two-dimensional systems, to the length of the circumference of crystallites of area s [8, 11], one provides for α the relation

$$\alpha = 1 - \frac{1}{d} \quad (2)$$

for systems of dimension $d > 1$.

The flux $j(v,t)$ reads, *cf.* (1),

$$j(v,t) = -\frac{\partial}{\partial v} v^{\alpha} f(v,t)\,. \tag{3}$$

After performing differentiation in (3) one sees that the flux $j(v,t)$ is decomposed into two parts, namely,

$$j(v,t) = -\alpha v^{\alpha-1} f(v,t) - v^{\alpha}\frac{\partial}{\partial v} f(v,t)\,. \tag{4}$$

These are respectively: the drift part and the diffusional part. Notice that the deterministic drift part is proportional to curvature $1/r$ of the grains, where r is the grain radius. Indeed, it is proportional to $v^{-1/d}$, but clearly v is proportional to r^d. The curvature-driven part is proportional to the surface tension change. The diffusional part conforms to a form of the first Fick's law and is proportional to the area of a single grain (because grains change their volume by gaining or losing atoms and the rate of attachment or detachment is proportional to the grain surface [15]) and to the gradient of the distribution function $f(v,t)$. Thus, one realizes that the surface properties, subjected to the $(d-1)$-dimensional hypersurface of a d-dimensional crystallite (bubble) are of crucial importance in understanding the mechanism of our modeling.

In order to solve Eq. (1) we have to support this equation by appropriate conditions. These are [8, 11]:

a) the initial condition

$$f(v,0) = f_0(v)\,, \tag{5}$$

where $f_0(v)$ is a given initial distribution of grains;

b) the boundary conditions

$$f(0,t) = f(\infty,t) = 0\,. \tag{6}$$

The physical interpretation of the boundary conditions is clear as well as unambiguous: the number of grains of zero volume $v = 0$ as well as of infinite volume $v = \infty$ at any time is zero.

3. Solution of the model

The solution $f(v,t)$ of the diffusion-type equation (1) can be presented in terms of a family of two-parameter evolution operators $\hat{P}(t,s)$ as follows

$$f(v,t) = \hat{P}(t,s)f(v,s)\,, \qquad t \geq s \geq 0\,. \tag{7}$$

This equation can be rewritten in the integral form, namely,

$$f(v,t) = \int_0^\infty P(v,t|w,s)f(w,s)dw\,, \tag{8}$$

where the integral kernel $P(v,t|w,s)$ of the time evolution operators $\hat{P}(t,s)$ forms a family of propagators. From the definition (8) of the propagators it follows that they obey the semigroup property

$$P(v,t|w,s) = \int_0^\infty P(v,t|z,\tau)P(z,\tau|w,s)dz\,, \qquad t \geq \tau \geq s \tag{9}$$

and the condition

$$\lim_{t\to s} P(v,t|w,s) = \delta(v-w)\,, \tag{10}$$

holds for any $t \geq s \geq 0$. Let us note that (9) is similar to the Chapman–Kolmogorov equation for the conditional probability distribution of stochastic Markovian processes. However, the distribution $f(v,t)$ is not normalized to unity and changes with time.

If the initial distribution $f(v,0)$ is given then from (8) it follows that

$$f(v,t) = \int_0^\infty P(v,t|w,0)f(w,0)dw\,. \tag{11}$$

It means that it is sufficient to construct the propagator $P(v,t|w,0)$. We seek a solution of (1) using the separation ansatz for $f(v,t)$, namely,

$$f_\lambda(v,t) = \mathrm{e}^{-\lambda t}\mathcal{G}_\lambda(v)\,. \tag{12}$$

It leads to the eigenvalue problem

$$\hat{L}_D\mathcal{G}_\lambda(v) = -\lambda\mathcal{G}_\lambda(v)\,. \tag{13}$$

Here $\mathcal{G}_\lambda(v)$ and λ are the eigenfunctions and eigenvalues of the "diffusion" operator $\hat{L}_D$ defined in (1). The eigenvalues may be discrete or continuous or both. One can show [16] that λ takes non-negative values in $[0, \infty)$.

From (1) and (12) it follows that $\mathcal{G}_\lambda(v)$ fulfils the ordinary differential equation of the second order

$$v^\alpha \mathcal{G}''_\lambda(v) + 2\alpha v^{\alpha-1}\mathcal{G}'_\lambda(v) + \left[\alpha(\alpha-1)v^{\alpha-2} + \lambda\right]\mathcal{G}_\lambda(v) = 0, \tag{14}$$

where the prime indicates differentiation with respect to v. The change of the independent variable

$$y = v^\alpha \tag{15}$$

transforms (14) into the equation

$$y^2 F''_\lambda(y) + ayF'_\lambda(y) + (by^m + c)F_\lambda(y) = 0\,, \tag{16}$$

where the new function $F_\lambda(y)$ is defined via the relation

$$F_\lambda(y) = \mathcal{G}_\lambda(v) \tag{17}$$

and $a = 3 - 1/\alpha$, $b = \lambda/\alpha^2$, $c = 1 - 1/\alpha$, $m = -1 + 2/\alpha$. Eq. (16) is the Bessel equation [17]. One can take an arbitrary set of two linearly independent particular solutions of this equation. Then a general solution is a linear combination of them. We choose it in the form [17]

$$\begin{aligned} F_\lambda(y) &= \mathcal{G}_\lambda(v) \\ &= v^{(1-2\alpha)/2}\left[C_1(\lambda)J_\nu\left(\frac{2\sqrt{\lambda}}{2-\alpha}v^{(2-\alpha)/2}\right) + C_2(\lambda)J_{-\nu}\left(\frac{2\sqrt{\lambda}}{2-\alpha}v^{(2-\alpha)/2}\right)\right], \end{aligned} \tag{18}$$

where

$$\nu = \frac{1}{2-\alpha} = \frac{d}{1+d} \tag{19}$$

and $J_\nu(x)$ is the Bessel function [18]. Two "constants" $C_1(\lambda)$ and $C_2(\lambda)$ appearing in the linear combination of the particular solutions $J_\nu(x)$ and $J_{-\nu}(x)$ are determined by initial and boundary conditions. Let us notice that the eigenvalues λ are continuous and as it was stated above $\lambda \in [0, \infty)$. Therefore from the method of separation of variables (12) it follows that the general solution $f(v,t)$ is a linear combination of the solutions $f_\lambda(v,t)$ taken over all values of λ. It means that in the case of continuous eigenvalues $f(v,t)$ can eventually be represented by the expression:

$$f(v,t) = \int_0^\infty f_\lambda(v,t)\,d\lambda = \int_0^\infty \mathrm{e}^{-\lambda t}\mathcal{G}_\lambda(v)\,d\lambda\,. \tag{20}$$

We have to determine two "constants" $C_1(\lambda)$ and $C_2(\lambda)$. For this aim, let us rewrite (18) in the form

$$\mathcal{G}_\lambda(v) = \mathcal{G}_\lambda^{(1)}(v) + \mathcal{G}_\lambda^{(2)}(v)\,, \tag{21}$$

where

$$\mathcal{G}_\lambda^{(1)}(v) = C_1(\lambda) v^{(1-2\alpha)/2} J_\nu \left(\frac{2\sqrt{\lambda}}{2-\alpha} v^{(2-\alpha)/2} \right) \tag{22}$$

and

$$\mathcal{G}_\lambda^{(2)}(v) = C_2(\lambda) v^{(1-2\alpha)/2} J_{-\nu} \left(\frac{2\sqrt{\lambda}}{2-\alpha} v^{(2-\alpha)/2} \right). \tag{23}$$

For sufficiently small x ($x \ll 1$), the leading term of the Bessel function is

$$J_\nu(x) \sim x^\nu. \tag{24}$$

Therefore for small values of v the functions $\mathcal{G}_\lambda^{(1)}(v)$ and $\mathcal{G}_\lambda^{(2)}(v)$ behave as

$$\mathcal{G}_\lambda^{(1)}(v) \sim C_1(\lambda) \lambda^{\nu/2} v^{1-\alpha} \tag{25}$$

and

$$\mathcal{G}_\lambda^{(2)}(v) \sim C_2(\lambda) \lambda^{-\nu/2} v^{-\alpha}. \tag{26}$$

Accordingly, for sufficiently small v the distribution $f(v,t)$ depends upon v as

$$f(v,t) \sim v^{1-\alpha} \int_0^\infty \mathrm{e}^{-\lambda t} C_1(\lambda) \lambda^{\nu/2} d\lambda + v^{-\alpha} \int_0^\infty \mathrm{e}^{-\lambda t} C_2(\lambda) \lambda^{-\nu/2} d\lambda\,. \tag{27}$$

Two integrals in this expression are different from zero and should be finite if $f(v,t)$ exists. The first term in (27) tends to zero when $v \to 0$ while the second term tends to infinity. Hence, the first boundary condition $f(0,t) = 0$ holds only if $C_2(\lambda) \equiv 0$. The second boundary condition, $f(\infty,t) = 0$, is fulfilled because for large x the Bessel function behaves as $J_\nu(x) \sim x^{-1/2}$ which tends to zero as $x \to \infty$.

Because $C_2(\lambda) \equiv 0$, the expression (20) can be recast in the explicit form as

$$f(v,t) = 2 v^{(1-2\alpha)/2} \int_0^\infty dz\, z\, \mathrm{e}^{-tz^2} B(z) J_\nu \left(\frac{2z}{2-\alpha}\, v^{(2-\alpha)/2} \right), \tag{28}$$

where the new integration variable $z = \sqrt{\lambda}$ has been introduced and $B(z) \equiv C_1(z^2)$ is a function determined by an initial distribution $f(v,0)$. Let us take the limit $t \to 0$. Then (28) converges to

$$f(v,0) = 2v^{(1-2\alpha)/2} \int_0^\infty dz\, zB(z) J_\nu \left(\frac{2z}{2-\alpha} v^{(2-\alpha)/2} \right). \tag{29}$$

Formally, it is an integral equation with respect to the unknown function $B(z)$. In fact, it is a Bessel transform [19]. The inverse Bessel transform yields $B(z)$ as an integral of $f(v,0)$. As a result we obtain

$$B(z) = \frac{1}{2-\alpha} \int_0^\infty dv\, v^{1/2} f(v,0) J_\nu \left(\frac{2z}{2-\alpha} v^{(2-\alpha)/2} \right). \tag{30}$$

If we insert (30) into (28) and change the integration order, we obtain the relation (11) in which the propagator $P(v,t|w,0)$ has the form

$$\begin{aligned} P(v,t|w,0) \;=\; & \frac{2v^{(1-2\alpha)/2} w^{1/2}}{2-\alpha} \int_0^\infty dz\, z\, \mathrm{e}^{-tz^2} J_\nu \left(\frac{2z}{2-\alpha} v^{(2-\alpha)/2} \right) \\ & \times J_\nu \left(\frac{2z}{2-\alpha} w^{(2-\alpha)/2} \right). \end{aligned} \tag{31}$$

The integration can be carried out [20] with the result

$$P(v,t|w,0) = \frac{v^{(1-2\alpha)/2} w^{1/2}}{(2-\alpha)t} \exp\left(-\frac{v^{2-\alpha} + w^{2-\alpha}}{(2-\alpha)^2 t} \right) I_\nu \left(\frac{2(vw)^{(2-\alpha)/2}}{(2-\alpha)^2 t} \right), \tag{32}$$

where $I_\nu(x)$ is the modified Bessel function [18]. Thus, the solution $f(v,t)$ is fully determined and now can be analyzed.

4. Main characteristics of the process

For any state function $G(v)$ one can determine the statistical characteristics of it like a mean value or fluctuations. For the special case when $G(v) = v^n$, $n = 0, 1, 2, \ldots$ one can obtain statistical moments $m_n(t)$ which we define by the integrals

$$m_n(t) = \int_0^\infty v^n f(v,t)\, dv, \quad n = 0, 1, 2, \ldots\ . \tag{33}$$

In virtue of (11) and (32) and after integration over the variable v, they take on the form:

$$m_n(t) = (2-\alpha)^{\frac{2(n-1)}{2-\alpha}} \frac{\Gamma\left(\frac{n-\alpha+2}{2-\alpha}\right)}{\Gamma\left(\frac{3-\alpha}{2-\alpha}\right)} t^{\frac{n-1}{2-\alpha}} \times \int_0^\infty dw\, f(w,0)\, w \exp\left(-\frac{w^{2-\alpha}}{(2-\alpha)^2 t}\right) \times M\left(\frac{n-\alpha+2}{2-\alpha};\, \frac{3-\alpha}{2-\alpha};\, \frac{w^{2-\alpha}}{(2-\alpha)^2 t}\right), \tag{34}$$

where $M(a,b,x)$ is the Kummer (confluent hypergeometric) function and $\Gamma(x)$ is the Euler gamma function [18].

The first two moments $N(t) \equiv m_0(t)$ and $V(t) \equiv m_1(t)$ are the most important ones because of their physical interpretation. The zero-order moment

$$N(t) = \int_0^\infty f(v,t)\, dv \tag{35}$$

is the relative number of grains at time t. From (34) one gets

$$N(t) = \frac{\beta^{1/(2-\alpha)}(t)}{\Gamma\left(\frac{3-\alpha}{2-\alpha}\right)} \int_0^\infty F(w,t)\, dw\,, \tag{36}$$

where

$$F(w,t) = w \exp\left(-\beta(t) w^{2-\alpha}\right) M\left(1;\, \frac{3-\alpha}{2-\alpha};\, \beta(t) w^{2-\alpha}\right) f(w,0) \tag{37}$$

and

$$\beta(t) = \frac{1}{(2-\alpha)^2 t}\,. \tag{38}$$

The first-order moment $V(t)$ is the *average total volume* of the system. Using (11) and (32), one can show by an explicit evaluation of the integrals that

$$V(t) = \int_0^\infty v f(v,t)\, dv = \int_0^\infty v f(v,0)\, dv = V(0). \tag{39}$$

It means that statistically the total volume of the system is preserved in time.

The next important characteristics of the process is the *average volume of a single grain.* It is defined as:

$$\langle v(t)\rangle = \frac{\int\limits_0^\infty v f(v,t)\,dv}{\int\limits_0^\infty f(v,t)\,dv}\,. \tag{40}$$

By virtue of (39), the mean single grain volume can be expressed via the average number of grains. Indeed, Eq. (40) can be rewritten as

$$\langle v(t)\rangle = \frac{V(t)}{N(t)} = V(0)N^{-1}(t)\,. \tag{41}$$

In this way, the main characteristics are determined by the zero-order moment $N(t)$ only.

5. Early stages of evolution

Now, let us investigate the influence of the initial condition $f(v,0)$ on evolution of the system. As a first example we consider the Dirac-delta initial distribution,

$$f(v,0) = N_0\delta(v-v_0) = \frac{V(0)}{v_0}\,\delta(v-v_0)\,. \tag{42}$$

Physically, it means that at initial time $t=0$ there are N_0 grains each of non-zero volume v_0 and in consequence the total volume of the system is $V(0)=v_0N_0$. In this case one gets

$$f(v,t) = N_0P(v,t|v_0,0) \tag{43}$$

and the zero-order moment

$$N(t) = V(0)\frac{\beta^\nu(t)}{\Gamma(\nu+1)}\exp\left(-\beta(t){v_0}^{1/\nu}\right) M\left(1;\nu+1;\beta(t){v_0}^{1/\nu}\right), \tag{44}$$

where ν is given by (19). The second initial distribution $f(v,0)$ is chosen to be the Weibull distribution (see Fig. 1),

$$\begin{aligned} f(v,0) &= N_0(2-\alpha)v^{1-\alpha}\exp\left(-v^{2-\alpha}\right) \\ &= \frac{V(0)}{\Gamma\left(\frac{1}{2-\alpha}\right)}(2-\alpha)^2v^{1-\alpha}\exp\left(-v^{2-\alpha}\right). \end{aligned} \tag{45}$$

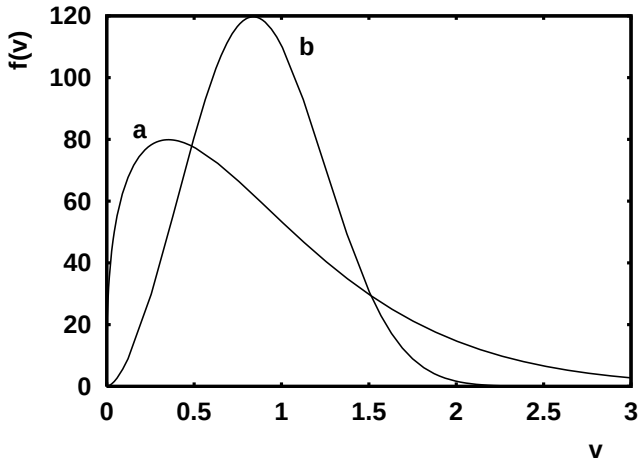

Fig. 1. Plot of the initial distributions $f(v,0) = f(v)$ versus v for a – the Weibull function defined by Eq. (45) and b – the Weibull-type function defined by Eq. (47). The parameters $V(0) = 100$ and $\alpha = 2/3$ (which corresponds to the three-dimensional case, $d = 3$).

In this case, the zero-order moment is expressed by an elementary function, namely,

$$N(t) = N_0 \left[\nu^{-2} t + 1\right]^{-\nu} = \frac{V(0)}{\nu \Gamma(\nu)} \left[\nu^{-2} t + 1\right]^{-\nu}. \tag{46}$$

The third example of the initial distribution $f(v,0)$ is the Weibull-type distribution (see Fig. 1),

$$\begin{aligned} f(v,0) &= N_0 (4-2\alpha) v^{3-2\alpha} \exp\left(-v^{4-2\alpha}\right) \\ &= \frac{V(0)}{\Gamma\left(\frac{1}{4-2\alpha}\right)} (4-2\alpha)^2 v^{3-2\alpha} \exp\left(-v^{4-2\alpha}\right). \end{aligned} \tag{47}$$

The corresponding zero-order moment is more complicated than in the second case and reads

$$\begin{aligned} N(t) &= N_0 2^{-\nu} \nu^{2\nu} t^{-\nu}\, U\left(\frac{\nu}{2}, \frac{1}{2}, \frac{\nu^4}{4t^2}\right) \\ &= \frac{V(0)}{\Gamma(\frac{\nu}{2})} 2^{1-\nu} \nu^{2\nu-1} t^{-\nu}\, U\left(\frac{\nu}{2}, \frac{1}{2}, \frac{\nu^4}{4t^2}\right), \end{aligned} \tag{48}$$

where $U(a,b,z)$ is the Tricomi (confluent hypergeometric) function. In Fig. 2, we visualize the influence of the initial distributions on the kinetics of the mean number of grains in the system. In all three cases we assume

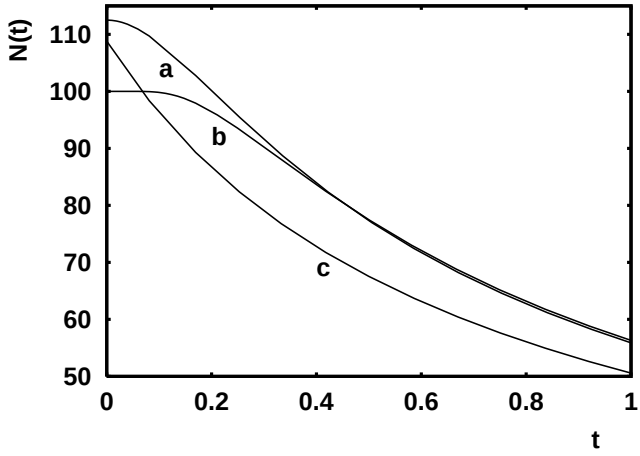

Fig. 2. The mean number $N(t)$ of grains as a function of time t in the 3-d systems and for three various initial distributions: $a - f(v,0)$ is the Weibull-type function given by (47), $b - f(v,0)$ is the Dirac-delta distribution (42) with $v_0 = 1$, $c - f(v,0)$ is the Weibull function (45). The average total volume of the system is $V(0) = 100$.

the same value of the total volume $V(0)$ of the system, *cf.* (42), (45) and (47). All three functions $N(t)$ monotonically decrease with time. For the Dirac-delta (42) and Weibull-type (47) initial distributions, after a relatively short transient regime, the zero-order moments approach almost the same values. In the case of the Weibull initial distribution (45), the mean number $N(t)$ of grains is, after very short time, smaller than in two previous cases. In turn, because the average total volume is the same in all cases, it means that the average radius of grains is larger in the third case.

6. Late stages of evolution

The behavior of the system for long time can be studied by analyzing the statistical moments in (34). If time $t \to \infty$, the integrand in (34) tends to the function $wf(w,0)$ and the integral tends to a constant value $V(0)$. Therefore one can conclude that for long time, $t \gg 1$, the statistical moments $m_n(t)$ behave as

$$m_n(t) \sim t^{(n-1)/(2-\alpha)}. \tag{49}$$

In particular, the zero-order moment exhibits the asymptotics

$$N(t) \sim t^{-1/(2-\alpha)} = t^{-d/(d+1)}. \tag{50}$$

The important feature is that the asymptotics do not depend on the initial state of the system which is characterized by the initial distribution $f(v,0)$ of the grains.

One of the basic characteristics of the normal grain growth is the average radius $\langle r(t)\rangle$ of the single grain. Because the volume $v(t) \sim r^d(t)$, the average radius is

$$\langle r(t)\rangle \sim \left\langle {}^{1/d}(t)\right\rangle \equiv \frac{\int\limits_0^\infty v^{1/d} f(v,t)\, dv}{\int\limits_0^\infty f(v,t)\, dv}. \tag{51}$$

Although the exponent $1/d$ is not a natural number, nevertheless one can exploit the expression (34) to calculate the average radius. As a result one obtains

$$\langle r(t)\rangle \sim t^{\mu} \tag{52}$$

with the exponent

$$\mu = \frac{1}{d+1}. \tag{53}$$

So, the average radius of grains increases with time and independently of the initial conditions, it depends powerly on time. It is interesting that the mean-field type definition

$$\langle r(t)\rangle \sim \langle v(t)\rangle^{1/d} \sim N^{-1/d}(t), \tag{54}$$

used in other papers [14], leads to the same result (52).

Now, let us integrate (1) over the phase space $v \in [0,\infty)$. Then one gets

$$\frac{dN(t)}{dt} = j(0,t). \tag{55}$$

One can check that $j(0,t) < 0$ and the flux at infinity $j(\infty,t) = 0$. From this relation it follows that the diminishing of the grain number $N(t)$ is related to the flux across the absorbing boundary at $v = 0$. This is why there are less and less grains: infinitesimally small grains disappear attaching some bigger ones.

7. Conclusions

The following conclusions can be listed as being of prior importance for the kinetics of d-dimensional grain growth as well as soap froth evolution:

(i) For the growth process with constant total hypervolume the evolution does not asymptotically depend upon the initial state $f(v,0)$;

(ii) Influence of the initial condition may sometimes be more pronounced (*cf.* the initial state in the form of Weibull functions of v), which suggests some possible application of the modeling proposed, mostly towards designing a fine-grained material;

(*iii*) Main characteristics of the evolution show up an explicit dependency upon dimensionality;

(*iv*) Evolution goes in a self-similar way both in the regimes of the space of grain sizes as well as time;

(*v*) If the total hypervolume of the system remains unconserved, one may expect abnormalities, *cf.* [2, 6].

Some comparison to another approach, proposed to describe a phase change or microstructure formation, under a set of physical conditions juxtaposed below is worth making. The conditions are [14]:

(*i*) the kinetic equation is of diffusion type, but the physical mechanism is readily manifested *via* some domination of local fluxes in the space of grain sizes; by the way, right at this point it is worth to realize a perennially alive discussion by Van Kampen on which kind of diffusion equation, suitable for description of kinetic processes in inhomogeneous media, one is likely to work under concrete physical circumstances to be modeled, *cf.* [21];

(*ii*) the diffusion (migration, mutation, *etc.* [14]) is a state-dependent process and the diffusion function $D(v)$ does depend powerly upon the physical state of the system, so that the role of the boundary is very much pronounced in this case;

(*iii*) there is no domination of the smallest as well as largest grains;

(*iv*) there exists an explicit dependence of the problem in question upon dimensionality by means of (2);

(*v*) total volume (hypervolume in a d-dimensional case, or simply area in a $2d$ case) increases powerly with time, which is in apparent contradiction with what is presented here. The first and last above stated conditions differ substantially while comparing with the normal grain growth conditions (formally, it should be realized that also the number of grains decreases slower in time than in the case of normal grain growth, namely as $\sim t^{-1/(d+1)}$ [8, 14]) while conditions (*ii*)–(*iv*) are exactly the same. It must also be noticed that the average radius of the grain or domain follows the same asymptotics in the both cases mentioned.

It is also worthy to argue that the phase transformation process, described in the present paper, proceeds in $2d$ as well as in $3d$ systems similarly from the qualitative point of view (*cf.* [8, 14]). There are, however, certain

quantitative differences. Namely, the distribution function, subjected to the same values of parameters, reaches its maximum value somewhat higher in $3d$ than in $2d$ case. The first two moments behave also in a slightly different manner, that means, the number of grains in the system (the 0-th moment of the process) decreases somewhat slower in $2d$ than in $3d$ case, whereas the total volume of the system (the first moment of the process) remains always conserved, no matter whether the system under consideration is two- or three-dimensional. The average radius, in turn, evolves a bit slower in a $3d$ system, which is also the case represented by the diffusion-type model mentioned above. Moreover, it is interesting to notice here that all the basic physical quantities that we have analyzed (the two first moments and the average radius of the microdomain) do scale perfectly with time t, with certain exponents being less than one, which are going to become exclusively dimensionality d-dependent.

In a final word, let us draw some attention to the fact, that the afore presented comparison could, to a certain extent, be exemplified by the recrystallization as well as grain growth processes in the single phase *b.c.c.* iron [22]. In the former, grains of the pre-strained iron phase after primary crystallization do not perfectly feel the available space and the remaining amorphous part competes with the crystalline one, which is however the case characteristic of earlier annealing times. In the latter, that means, when the annealing time rises considerably, the crystalline phase prevails, which eventually results in perfect space-feeling by the formed polycrystal. Then, the evolution of the polycrystal is reported to proceed in a uniform way, rather (no signatures of abnormality are detected in [22]). It appears to be interesting, that the growth exponents got from measurements differ, while compared with ours, mostly in the (stationary) grain growth case. It may thus imply, that the offered model would be more suitable for bubbles-containing systems, where such low-valued fractional exponents can be met [3]. It cannot, in fact, be discarded in total for polycrystals, since for such systems one anticipates quite often (for instance, for ceramic ferroelectric polycrystals) the exponents about 1/2 or lower, just around 1/4 for three-dimensional systems ($Ba\ Ti\ O_3$) [1,2,23]. It may, however, follow from the presented modeling, that no hope for inclusion of some necessary information about the type of crystallinity (perhaps, about the mechanism of appearence/disappearence of the grain boundaries as well as grain shapes alterations [22]) into the offered statistical-physical description would as usually result, at least in some specific cases, in misfits between theory and practice.

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