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Finite volume effects in a model grain growth

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Abstract

Usually the distribution function of grains is defined on a semi-axis of all possible sizes of grains. It leads to some inconsistency, because volume of the individual grain can be arbitrary large and can be larger than the total volume of the system as a whole. Therefore in this paper we want to reconsider this problem assuming that the volumes of the individual grains are finite. We regard two classes of the boundary conditions: absorbing and reflecting. For the absorbing conditions, all statistical moments exponentially tend to zero, the stationary state does not exist and the grains-containing system is going to dissolve. In turn, the use of the reflecting boundary conditions realized in a finite size space reveals possibly a phenomenon of practical interest: Existence of a stationary state that depends upon space dimension.

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1. Introduction

The grain growth phenomenon in a variety of different materials attracts permanent attention because it becomes still a real challenge to give a satisfactory description of both its kinetics as well as dynamics whereas certain practical applications are desired, and very expected, mostly in nanotechnology, (bio)membrane engineering and microelectronics to mention but a few [1]. The grain growth of uniform boundaries, called most frequently the normal grain growth, is a complex phenomenon emerging

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after the recrystalization of a material is completed [2,3]. Its main landmarks as a model system are as follows: The process, taking very often place under a constant volume regime, relies on evolution of grains constituting a polycrystalline body in such a way that a mean radius of the grain has to grow in time whereas the total number of grains has to diminish. A basic physical tendency observed in the course of time is always a decrease of the grain surface free energy so that a low surface free energy microstructure is eventually formed.

It is accepted that the most important kinetic hindrance of the evolution in question appears to be the behavior of a single grain boundary (GB) which is a principal kinetic limiting factor of any evolution of the polycrystalline microstructure [1]. A GB-motion, in turn, can readily be examined as a process driven by deterministic as well as indeterministic sources, see Ref. [4, and refs. therein]. A deterministic source is represented by the capillary driving forces, where the motion of a GB towards its center(s) of curvature is performed. Another natural deterministic factor of interest is the total surface magnitude of the grain under evolution: The bigger the surface is the more vigorously the evolution can be, so that the grain under observation can absorb its neighbours by letting their material be transported through its GB into its internal space [5]. An indeterministic source of the GB-motion is of more hidden nature. Some authors claim that it is due to topological constraints, and their random arrangements during the evolution under study [4], some recall other valuable but 'noisy' sources [5,1].

In a series of papers, cf. Refs. [6,7] for example, we drew a statistical–mechanical picture of the grain-growth of a normal type based, similarly as in the nucleation theory [8,9], on a Fokker–Planck (FP)-type equation with suitable initial (IC) as well as boundary conditions (BC) [6,7]. The way by which we arrived at such a kinetic description, which models by means of the FP-equation the kinetics of the cellular system in a mesoscopic scale [8,9], was the following:

(A) Based on possibly realistic experimental as well as theoretical assumptions [5,1,4] we constructed, in a space of the grain sizes (but apparently not in a position space!) a local flux of grainy matter being composed of two basic terms: (i) A convection-driven term, proportional to the grain curvature; (ii) A diffusion-driven term, related via the surface magnitude of a single grain with a derivative of the grain distribution function: This term is of the well-known form of the 1st Fick's law.
(B) The constructed flux was plugged into a continuity equation, and the equation was solved subject to suitably chosen IC and BC.
(C) After getting the solution in its explicit form, the corresponding statistical moments were calculated; they represented a total (relative) number of grains in the system, its total volume, and when the latter is divided by the former, an expression for the average single grain volume can easily be obtained, which can eventually be related via a standard volume-to-radius relation with the mean grain radius.

In a few papers, cf. Ref. [6, and refs. therein], we explored thoroughly a dependence of the normal grain growth on the IC and we quite firmly arrived at a conclusion that

the long-time asymptotics does not depend upon the initial condition. Such a conclusion is justifiable as well as seems reasonable, especially when having a closer look into experiments [10,5,1,4], where no dependence upon the initial state is detected. What seems to us worth investigating is still a further exploration of the BC. Now, however, we wish to discuss which is the influence, if any, of two basic types of BC: The absorbing (Dirichlet) as well as the reflecting (Neumann) ones.

The paper is structured as follows. In Section 2, we formulate the extended Louat–Mulheran–Harding (LMH) model. In Sections 3 and 4, respectively, we discuss the two types of BC mentioned, operating in a finite phase space. Section 5 serves for conclusion and comparison with another model [11].

2. Extended Louat–Mulheran–Harding (LMH) model

The LMH model [12,6] is a sort of a random walk (RW)-like model where the RW is to be realized by means of a random hoping that is not done in a position space but clearly along the grain size axis. Thus, it usually describes the size- and time-dependent rearrangement of a polycrystalline system by means of the following continuity equation [7]

$$\frac{\partial}{\partial t} f(v,t) = -\frac{\partial}{\partial v} J(v,t) \,, \tag{1}$$

where v is the volume of a grain, D is a constant reflecting a RW behavior in grain growth, to be named as diffusion–migration reference constant, $f(v,t)$ is the distribution function of the grains at time t (having the meaning of the number density), i.e., $f(v,t)\,\mathrm{d}v$ is a relative number of grains of size in the volume range $[v, v+dv]$. The suitable IC reads

$$f(v,t=0) = f_0(v) \,, \tag{2}$$

where $f_0(v)$ is a given initial distribution of grains. The flux $J(v,t)$ in Eq. (1) is assumed to be

$$J(v,t) = -\sigma v^{\alpha-1} f(v,t) - D v^{\alpha} \frac{\partial}{\partial v} f(v,t) \,, \tag{3}$$

which means that it is decomposed into two parts. These are respectively: The drift and the diffusion terms. (Such a decomposition realized, however, on a mesoscopic level, resembles the Pandé's construction of the GB-speed term made of a capillary as well as random parts, respectively [4,5]. Notice that Pandé's presumption is invented to mimic a dynamic behavior of the GB as a separate unit but investigated on a microscopic level.) The parameter $\alpha \geqslant 0$ depends on dimension d of the system,

$$\alpha = 1 - \frac{1}{d} \,. \tag{4}$$

Notice that for $d = 1$ one provides $\alpha = 0$, so that it implies that the approximation invented does work effectively for $d > 1$, and in this work is confined to d being of integer value. The parameter α reflects the fact that the net flux of the particles wandering across the grain boundaries is proportional to the area of the surface $s \propto v^{2/3}$

of grains of volume v (for three-dimensional systems), and to the length $l \propto s^{1/2}$ of the circumference of crystallites of area s [5] for two-dimensional systems. The drift term is proportional to curvature $1/R$ of the grains, where R is the grain radius. It is so indeed because it is proportional to $v^{-1/d}$, but clearly v is proportional to R^d. Certainly, after Kelvin, Young and Laplace the curvature-driven part is proportional to the surface tension associated with a GB [5]. The diffusional term, in turn, takes the form of the phenomenological 1st Fick's law, with an appropriate modification however, namely that the flux is proportional to the area of a grain. Now σ and D can be independent parameters. It means that the surface tension mechanism of growth is independent of the mechanism of the migration of particles through boundaries of grains. We can rescale time and it means that we can put for simplicity $D=1$ in further considerations.

In previous studies [13], it was assumed that the volume of a grain can take on arbitrary non-negative values $v \in [0, \infty)$ and Eq. (1) has been solved with the BC of Dirichlet type, namely

$$f(v=0,t)=0, \quad f(v=\infty,t)=0\ . \tag{5}$$

For the LMH model (when $\sigma=\alpha$) the average total volume of the whole system is constant in time and is finite. It leads to some inconsistency, because volume of the individual grain can be arbitrary large and can be larger than the total volume of the entire system. Therefore in this paper we want to reconsider this problem assuming that the volumes of the individual grains are finite.

3. Dirichlet boundary conditions

We assume that all grains are finite and their volumes are not larger than a given overall volume $V_0 < \infty$. Now, $v \in (0, V_0)$. It seems that the simplest boundary conditions can be postulated as the only slight modification of those for the unlimited phase-space, i.e., in the form, cf. Eq. (5)

$$f(v=0,t)=0, \quad f(v=V_0,t)=0\ . \tag{6}$$

To investigate to what consequences it may eventually lead we solve the evolution equation (1) with the boundary conditions (6). To accomplish the task, we exploit the separation variable method assuming that

$$f(v,t)=\mathrm{e}^{-\lambda t}F_\lambda(v)\ . \tag{7}$$

It leads to the eigenvalue problem for the function $F_\lambda = F_\lambda(v)$,

$$v^\alpha F_\lambda'' + (\alpha+\sigma)v^{\alpha-1}F_\lambda' + \sigma(\alpha-1)v^{\alpha-2}F_\lambda = -\lambda F_\lambda \tag{8}$$

with the boundary conditions

$$F_\lambda(0)=F_\lambda(V_0)=0\ . \tag{9}$$

The prime denotes the derivative with respect to the argument. The solution of this differential equation can be presented in terms of the Bessel functions $J_\nu(z)$,

namely,

$$F_\lambda(v) = v^{(1-a)/2}\left[AJ_\nu\left(\frac{2}{m}\sqrt{\lambda}v^{m/2}\right) + BJ_{-\nu}\left(\frac{2}{m}\sqrt{\lambda}v^{m/2}\right)\right] , \tag{10}$$

where A and B are constants and

$$a = \alpha + \sigma, \quad m = 2 - \alpha, \quad \nu = \frac{1 + \sigma - \alpha}{2 - \alpha} > 0 . \tag{11}$$

In order to fulfil the boundary condition at zero, $F_\lambda(0) = 0$, one has to put $B = 0$. The second boundary condition $F_\lambda(V_0) = 0$ is satisfied if

$$J_\nu\left(\frac{2}{m}\sqrt{\lambda}V_0^{m/2}\right) = 0 . \tag{12}$$

It determines the eigenvalues

$$\lambda = \lambda_n = \frac{1}{4}m^2 V_0^{-m}\beta_n^2, \quad n = 1, 2, 3, \ldots , \tag{13}$$

where β_n are the positive zeros of the Bessel function,

$$J_\nu(\beta_n) = 0 . \tag{14}$$

The eigenvalue $\lambda_0 = 0$ has to be rejected because Eq. (1) does not possess the stationary solution which fulfils the boundary conditions (6). A general solution of (1) is a linear combination of the functions of (7),

$$f(v,t) = \sum_{n=1}^{\infty} A_n \mathrm{e}^{-\lambda_n t} F_{\lambda_n}(v) , \tag{15}$$

where the constants A_n are determined from the IC (2). Their explicit form is not needed because we are interested in the long-time asymptotics ($t \gg 1$) of the statistical moments

$$\langle v^k \rangle = \int_0^{V_0} v^k f(v,t)\,\mathrm{d}v, \quad k = 0, 1, 2, \ldots . \tag{16}$$

One can see that all moments tend to zero as $t \to \infty$. In particular, the number of grains (the zero moment) decays exponentially to zero as well as the average volume of all grains (the first moment) drops exponentially to zero. It means that all grains would disappear. In consequence, the boundary conditions (6) cannot describe correctly any grain growth process. It remains to comment right here that, quite amazingly, one may again appreciate the fact that the infinite size of the phase space helps to get some fruitful results, as often anticipated by statistical–mechanical theories, mostly while studying phase transformation kinetics. Put it another way, a 'slight' modification in a BC, but without changing the BC type, can even provide drastic changes in a system behavior.

4. Neumann boundary conditions

In the standard LMH model [12,6], the average total volume of the system is constant in time what corresponds to the constant first moment. It immediately suggests to

introduce the auxiliary function

$$g(v,t) = v f(v,t)\ . \tag{17}$$

Then this function is non-negative and normalizable in the standard LMH model, cf. Ref. [12, and refs. therein]. It satisfies the equation

$$\frac{\partial}{\partial t} g(v,t) = -\frac{\partial}{\partial v} j(v,t) + (\alpha - \sigma) v^{\alpha-2} g(v,t), \quad D = 1\ , \tag{18}$$

where the auxiliary current is given by

$$j(v,t) = (2-\sigma) v^{\alpha-1} g(v,t) - v^{\alpha} \frac{\partial}{\partial v} g(v,t)\ . \tag{19}$$

Eq. (18) looks like a diffusion equation with the source $(\alpha - \sigma) v^{\alpha-2} g(v,t)$. For the standard LMH model, $\alpha = \sigma$ and the source becomes zero. Let us consider this case. We postulate the following boundary conditions of Neumann type

$$j(v=0,t) = 0, \quad j(v=V_0,t) = 0\ . \tag{20}$$

It guarantees the normalization of $g(v,t)$. In the stationary state, the current $j_{st}(v) = 0$ and it implies that the stationary distribution

$$g_{st}(v) = N v^{2-\alpha}\ , \tag{21}$$

where N is the normalization constant determined from the condition

$$\int_0^{V_0} g_{st}(v)\,\mathrm{d}v = \int_0^{V_0} v f_{st}(v)\,\mathrm{d}v = V_0\ . \tag{22}$$

The stationary grain distribution exists and has the form

$$f_{st}(v) = N v^{1/d}, \quad N = \left(2 + \frac{1}{d}\right) \Big/ V_0^{1+1/d}\ , \tag{23}$$

where d is the dimension of the space. The time-dependent solution of Eq. (18) for $\alpha = \sigma$ can be obtained by using the same method as in the previous section and the time-dependent grain distribution $f(v,t)$ is again of the form of Eq. (15) where the constants A_n are determined from the IC (2) and now the eigenvalues λ_n are determined from the equation

$$J_{\nu+1}\left(\frac{2}{m}\sqrt{\lambda_n} V_0^{m/2}\right) = 0\ . \tag{24}$$

In this case, the eigenvalue $\lambda_0 = 0$ should be taken into account and it corresponds to the stationary distribution (23). Under such physical circumstances, the growth process has radically different properties: The grains evolve exponentially in time to a stationary state (23). This function is a monotonically increasing function of v, or the radius R via $v \propto R^d$, and it means that the large number of grains has big volume (or radius). But also small grains survive in the stationary state. This is because the average total volume of the system does not depend on time and the reminder of the volume which cannot be filled by large grains is completely filled by smaller and smaller grains. In this case, the boundary conditions (20) are physically acceptable. It remains, however,

to be checked whether such a scenario is plausible from the experimental viewpoint, and can be met, e.g. in Refs. [1,5].

5. Concluding address

The main conclusion can be presented as follows:

(1) We have looked more carefully at the grain growth phenomenon by examining how do certain BC affect its kinetics.
(2) We have regarded two classes of the boundary conditions: absorbing and reflecting assuring, contrary to LMH description, that the grain size axis (space) is finite.
(3) For the absorbing boundary conditions (6), a quite hardly expected effect that we have noticed is that the physically motivated assumption about finiteness of the grain size space leads to an undesirable physical system behavior: the system under study is, unfortunately, going to dissolve and no real growth signatures can be observed; on the contrary, something like a polycrystalline phase separation [1] effect [14] can be suspected; by the way, when formulating the kinetic problem in a semiinfinite space, we arrive at a very interesting physical behavior, first known in physical metallurgy as the LMH statistical grain growth description [12], the main landmark of which is presence of the power laws (in the long-times regime), containing a characteristic exponent, presumably reflecting a certain 'fingerprint' of the random close-packing, a notion well known in physics of amorphous as well as polycrystalline materials.
(4) While applying the reflecting boundary conditions (20) taken at both ends of the finite size space, and for a suitably reformulated kinetic problem a phenomenon of possibly prior technological interest has been revealed, i.e., the existence of a stationary state that depends exclusively upon space dimensionality.
(5) Some criticism of the FP-description of the grain growth of uniform boundaries due to Mullins [2], pointing to a possible absence of the diffusion term cannot be appreciated because the argumentation he offered (his assumption about infinite GB-mobility seems to be unphysical [4]) presents a rationale valid for a standard RW case realized in a position space; in turn, an extended FP-description of (poly)crystallization by Reguera et al. [8], including the useful notions of out-of-equilibrium thermodynamics looks promising, mostly when applied to complex (bio)polymeric systems.

[1] Phase separation often accompanies the specimen ripening effect. According to the description of Lifshitz and Slezov (LS) the spherically shaped grains have to be well separated [11], then they may grow and coalesce until impingment, and this is called the ripening effect, first described by Ostwald. It seems to us that the main differences between the LS approach and our model are pronounced twofold: (i) the dependence upon dimension d is manifested in ours but not in the LS description, e.g. they arrived at an $1/3$-asymptotic power law for the critical radius whereas in our description we offer consequently $1/(d+1)$ [13] but in other studies [2,4], in turn, the $1/2$-exponent kinetic law can be encountered as if the process was either of diffusive or surface tension-mediated characters; (ii) the diffusion–migration coefficient depends upon the grain surface magnitude [7] while the LS approach suffers from constancy of the analogous quantity.

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