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Fractal-type relations and extensions suitable for systems of evolving polycrystalline microstructures

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Abstract

We report on two possible physically justified situations concerning the evolution of polycrystalline microstructures. The first is an observation that a polycrystalline microstructure formation process, considered as a random walk in the space of the crystallites' sizes, can be equivalent to the anomalous kinetic problem in a continuum percolation space. The second, in turn, regards some extension of the random walk process while occurring on a fractal substrate and/or when a single grain boundary can presumably be treated as a random walk trajectory, i.e. a situation quite acceptable when dealing with, e.g. quasicrystals. One may find possible applications of the modeling offered in the areas of biophysics and physical metallurgy.

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1. Introduction

Since the appearence of pioneering works of Fortuin and Kasteleyn [1–4], followed thoroughly by more recent studies [5,6], it is accepted that the Ising and percolation systems have very much in common. Moreover, this statement may even be extended when dealing with others, say, Ising-like systems, e.g. the Potts models, or its practical realization in biophysics, named the Pink model of clustering [7–9].

In this short paper, we are going to show that two kinetic processes, being apparently different at a first look, i.e. the formation of polycrystalline microstructures

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(represented by an inhomogeneous diffusion type model in the space of crystallites sizes [10–16]) and a random walk realized in a continnum (Swiss cheese) percolation matrix near criticality (a position space), seem to be practically equivalent, because of possessing very similar kinetic characteristics. Thus, in Section 2, a presentation of the reasoning, supporting the above stated observation, is given. In Section 3, in turn, some extension of scaling relations characterizing the same process of polycrystaline microstructure formation, quite important in physical metallurgy is presented, in terms of its non-Euclidean geometrical characteristics. Section 4 contains closing remarks.

2. On some equivalence between the formation of polycrystals and a kinetic random walk on the continuum Swiss cheese-like percolation matrix

In a series of papers (see, [10–13] and references therein), we regarded the formation of grains containing (i.e., space filling) systems in a d-dimensional space, where an analytical model of (inhomogeneous) diffusion was used and where the process was grain boundary curvature driven, rather, than with an expected ability to obey the 1st Fick's law. (Therefore, we called sometimes the process to be of *explicitly* non-Fickian character, abbreviated here as the NF-process.) In another study (see, [14–16] and references therein) doing the same, but for some, e.g. recrystallizing assemblies (which do not obey the condition of space filling), we used a diffusion-type model, with an explict requirement of the 1st Fick's law to be fulfilled, contrary to the NF-case mentioned [10–13]. It explored a well-known continuity equation of the form

$$\frac{\partial}{\partial t} f(v,t) = -\frac{\partial}{\partial v} J(v,t), \quad v \in [0,\infty)\,, \tag{1}$$

where $f(v,t)$ is a distribution of crystallites of volume v (at time t) in the system of total hypervolume $V^{(d)}$, and the local flux $J(v,t)$ is straightforwardly obtainable from the 1st Fick's law. It was asssumd that the diffusion function involved in the flux is a power function of v, cf. [10–16], though an inverse power dependence upon t, like $\sim (const.+t)^{-h}$ (h – a dimensionless competition [17,18] dynamic parameter; cf. [10–13] for details) was considered as well. (We will, however, take $h=0$ in our present study [14–16], which means, that the growth process passes smoothly and within a moderate range of temperature values through a set of thermodynamic quasiequilibria; see [19–21], too. We called sometimes that process [14–16] to be of purely Fickian character, abbreviated here as the F-process.) Surprisingly, solving both the above introduced systems for the same initial (delta-Dirac) and θ-Dirichlet boundary conditions (it is usually a standard procedure of evaluating the first statistical moments of $f(v,t)$, but one may look into [10–16] for necessary details), one gets the same asymptotic result for the average radius of the crystallite, r_{av}, namely (for $t \gg 1$) [10–16]

$$r_{av}(t) \sim t^{1/d+1}\,, \tag{2}$$

where d stands for the dimension of the space ($d=1,2,3,\ldots$), irrespective of whether the system is space filling (grain boundary curvature driven) or not. (Note that the

standard diffusional scaling form is recovered exclusively when $d = 1$.) It was, however, not a physical paradox, since we knew that the systems of first type (NF-systems) do conserve their $V^{(d)}$ (e.g., in the normal grain–growth or bubbles containing processes [22,13]) while the other ones (F-systems), like certain gelling systems containing microcrystalline regions, do not; see examples listed in [14–16].

Another observation that comes out from these studies would be that the quantity r_{av} can be interpreted as an averaged distance travelled by the boundary of an "averaged" grain from its center of mass (inertia). The process of grain boundary wandering in a random fashion is thought to be realized in the environment of other (neigboring, etc.) grains that stand for randomly distributed obstacles perturbing the walk of the boundary. This scenario resembles very much a random walk of a tracer on the percolation Swiss cheese-like continuum space, both for $d = 2, 3$.

It can be learned from classical literature sources [23–25] that for $t \gg 1$

$$r_{rw}(t) \sim t^{k_p}\,, \tag{3}$$

where $k_p = \nu/2\nu + \mu - \beta$, and ν, μ, β represent the standard critical exponents of the percolation system at criticality, when the incipient infinite cluster (IIC) is formed for the first time. If the cluster is finite, but really the largest taken from the population of finite clusters (a FL-cluster), the numerator in k_p (see also the right-hand side of (3) and specification of k_p thereafter), must be replaced by $\nu - (\beta/2)$. Certainly, r_{rw} stands for the mean distance travelled by the walker on the percolation space [23–25].

It can be anticipated as another evidence that the polycrystalline and percolation systems are dynamically equivalent. It is indeed so, since asymptotic relations (2) and (3) describe almost the same temporal behavior, since the exponent $k_p \approx 1/d + 1$. It is summarized by the following

Polycrystal NF, F	Continuum percolation IIC/FL	
$2d$: $r_{av} \sim t^{1/3}$	$r_{rw} \sim t^{0.3302}$	$r_{rw} \sim t^{0.3483}$
$3d$: $r_{av} \sim t^{1/4}$	$r_{rw} \sim t^{0.2414}$	$r_{rw} \sim t^{0.3103}$

where all the symbols used can be found in the text written in this section, and the data for continuum percolation have been taken from [23–25]. Moreover, note that for an FL-object, which mimics a space available for a random walk realization of a test particle, the listed exponents differ more markedly from the values of $\frac{1}{3}$ or $\frac{1}{4}$, for a $2d$ or $3d$ case, respectively. It is simply due to the finiteness of the system under consideration.

3. An analysis of the kinetics of stochastic grain growth: some extension

A couple of years ago an interesting study on the grain growth [26–29] kinetics was published [22], in which both, a crucial role of deterministic as well as stochastic

character of the process was emphasized. In the former, the most explored mechanism proposed for the driving force appears to be a capillary force, connected *via* the Gibbs–Thompson relationship, with a magnitude of the grain boundary curvature [10–13,30,31]. In the latter, in turn, a random walk (RW) concept is usually offered [19], which directly leads to the presumption that there exists a certain analogy between the RW (typically, approximated by the standard diffusion equation for a joint probability of finding a "walker" at a certain time-position point) of a particle in real space and the motion of grains in the space of their sizes [14–16,30,31].

The deterministic mechanism for the growing process mentioned always predicts

$$R(t) \sim t^{1/2}\,, \tag{4}$$

for the long times limit, which is also very characteristic of the so-called (normal) diffusion-controlled processes of any kind. Note that $R(t)$ is, very often, referred to as the linear grain size (or radius) taken at time t.

The stochastic character of the process was considered quite naturally by the authors of [22]. Namely, they really performed a RW realization for some stochastically changing ensemble of grains constituting a three-dimensional polycrystal, and they found out that the mean increase Δv in the volume of a certain number of growing crystallites reads [22]

$$\Delta v \propto t^{1/2}\,, \tag{5}$$

i.e., qualitatively the same relation as that written above as (4) has been obtained. There appeared, however, an important qualitative difference, since volume elements are simply related, by a cubic relationship (but, in the Euclidean space, exclusively), to the linear characteristics such as e.g., the mean radius r_{av} of the crystallite. Thus, being in close agreement with this elementary geometrical knowledge and utilizing (5), one is able to provide the asymptotic scaling formula valid for the "stochastic side" of the process under study, namely

$$r_{av}(t) \sim t^{1/6}\,, \tag{6}$$

which was also estimated by Chen and confirmed specifically by Monte Carlo simulations of Potts-like systems [9], mostly in some correspondence to metallurgical phenomena manifesting strong physi- and chemisorption as well as other retardation effects [10–13,26–31]. Another example may invoke polymeric quasicrystalline assemblies, in which the macromolecular adsorption is very much suspected to play a crucial role in inhibiting the growth. Thus, some experimental biophysical microdomain systems composed of multilamellar lipid vesicles, characterized by small fractional dimensionalities, can probably stand for well-chosen candidates, too [32–34].

Let us point out two useful but rather rarely mentioned extensions of the approach proposed above (and thoroughly presented in [22]). Both of them concern with some 'refinement' of its stochastic part.

Thus, the first extension that can be taken into account is just a simple geometrical observation that it would be useful to consider not only Euclidean objects (crystallites)

but also some non-Euclidean, say fractal (see [26–29]), quasicrystalline domains, or simply the evolution of domain systems in a fractal space of dimension d_f; see, also [10–13]. It immediately enforces to replace (6) by a more realistic asymptotic scaling rule

$$r_{av}(t) \sim t^{1/2d_f}\,, \tag{7}$$

where $0 \leqslant d_f \leqslant d$ (d_f is a non-integer), so that, e.g. the limit of the Euclidean dimension $d = 3$ is readily available, and for such (7) must be fully identified with (6).

The second extension towards our "refinement procedure" is probably more subtle [19]. Namely, it makes use of a more established knowledge on the RW process. That knowledge teaches us, in general, that there are three types of RW realizations, performed with the fractal dimension, d_w, of a random path along which the walker travels across the available space [23–25]. When the walk is realized in a certain accelerated way, $d_w < 2$, so as it would be systematically "kicked" (say, enforced to be in hurry) by the physical environment, in which the action takes place. Such a walk is said to be superdiffusive or just a "busy" RW. If, in turn, the walk is steadily caused to rest (e.g., by a chemical reaction), and its realization is anomalously slow, the walk, with $d_w > 2$, is called to be subdiffusive or just a "lazy" RW. (The walk "in between", considered in [22], is typically named the normal or "neutral" RW).

In this context, Eq. (5) is to be modified as

$$\Delta v \propto t^{1/d_w}\,, \tag{8}$$

so that (7) has to be rewritten as

$$r_{av}(t) \sim t^{1/d_w d_f} \tag{9}$$

which for the standard case ($d_w = 2$) coincides very well with (7), and for $d_f = d = 3$ also with (6).

One can recall many physical grounds for which (9) is a useful approximation. They are juxtaposed e.g., in [26–29], i.e. in a metallurgical context. Let us mention here, for example, Zener (due to pinning) or Mullins (due to thermal grooves) drag forces providing some rationale for introducing RW with $d_w > 2$, because the drag forces are serious kinetic obstacles for the polycrystalline growth. (Also, by noticing that there exists, in the limit of $q \to 0$, a possibility of mapping the q-state Potts model [5–9], i.e. a very useful tool for reproducing by means of the Monte Carlo simulation technique polycrystalline microstructures [10–13], onto a percolation microstructure [1–6,23–25] at criticality, one may hope for a proper use of (8) at least for having $d_w = 2 + \varepsilon$, i.e., $d_w > 2$, where $\varepsilon > 0$, and typically of fractional value, can be completely determined by the critical percolation exponents [23–25,32–35].) Or, the presence of high-angle grain boundaries, or even small grains (fine-grained structure), both with large curvatures, which in turn ensures a vigorous realization of the growing process.

As was proposed by Furukawa and others [14–16], who studied systems of droplets (clusters) in quenched but unstable states, one may eventually carry the whole analysis to get some interpolating scaling formula (see also [10–13]), which is now not

$r_{av}(t) \sim t^{1/3}$ [8], i.e., in a Lifshitz–Slyozov (LS) form, mostly characteristic of the Ostwald ripening [14] but, by combining (4) and (8), in the long times limit, one provides

$$r_{av}(t) \sim t^{\frac{(1/2)+(1/d_w d_f)}{2}} \tag{10}$$

which for $d_w = 2$ (standard RW) and $d_f = d = 3$ recovers just the LS form.

Let us note that result (10) is readily capable of reproducing a huge spectrum of the temporal behaviours characteristic of the normal as well as a-normal (supernormal or abnormal) grain growth as well as recrystallization processes in pure metals (Al) or alloy systems ($\alpha - Fe$ or $\gamma - Fe$, for example) [26–29]. Invoking the long-living analogy between metallurgical and biophysical [10–13,32–35] systems, it can also serve to elucidate some asymptotic temporal behaviour in micelles or vesicles; cf. [19–21], and references therein. To summarize the efforts of this section, let us state explicitly that we have proposed a self-consistent and simple picture of the polycrystalline structure formation, realized very much in a spirit of earlier deterministic-stochastic studies [22,30,31], but also extended naturally to a variety of anomalous (random walk) and fractality-dependent cases of its realizations [19–21,10,12]. The extensions proposed may serve to elucidate many anomalous kinetic data, frequently reported in the literature [10–13,32–35].

4. Closing remarks

The presented study exemplifies another way of how to support a formal analogy between the polycrystalline as well as percolation systems. In Section 2, it was done by noticing that a kinetic (percolation) exponent, k_p, is of a value being quite comparable to $1/d + 1$ (d – dimension of the space), and some additional possibilities of how to approach this value more closely have also been proposed in [10–13] ($h \neq 0$). A qualitative rationale that stays behind says that a walk of a random tracer on the continuum percolation matrix can be thought of to be, on an average, (nearly) equivalent to a random motion of the boundary of a crystallite. In Section 3, in turn, certain possibilities of extending well-known kinetic characteristics of a polycrystalline (in particular, grains containing) system) to non-Euclidean, let us say fractal cases, have been pointed out, mostly based on [10–13,22,30–35]. In particular, note that when looking at the exponent in (10), one might just redefine the fractal dimension of a "grain walk" in the space of grain (crystallite) sizes as

$$d_w^g = d d_w \,, \tag{11}$$

where d_w^g could presumably be understood as the RW dimension of a path of a randomly walking particle, of fractal dimension d_w, when the particle jumps over the grain boundaries, assumming for the moment that the grain boundary network were artificially frozen, but with a general rule (restriction) superimposed, namely, that the jumps over low-curvature boundaries are energetically favourable. Although the concepts

presented here seem to be still waiting for further more formal exploration, it is worth noticing that contrary to the material presented mostly in [1–6], the analogy between polycrystals and (weighted) percolation is explored here on a dynamic level, where certain asymptotic temporal characteristics (Eqs. (2)–(10)) have to be discussed. (For a recent minireview on the percolation as a phase transition problem one is strongly encouraged to consult [35].)

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