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Use of stereology to derive a new kinetic equation for mean curvature driven grain growth

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ABSTRACT

The idea that normal grain growth driven by surface tension may be described theoretically by the hypothesis that the local velocity of an element of grain boundary is proportional to its local mean curvature dates back more than half a century. von Neumann was the first to derive this relation and used it to predict the rate of evolution of a two dimensional cell structure. MacPherson and Srolovitz extended this development to describe growth in three dimensions; however, their result was couched in terms that did not facilitate tests of the theory. In this paper expected value theorems established in stereology are invoked to extend their result to provide a new equation that predicts the rate of change of volume of grains in a microstructure which, while preserving the rigor and generality of the result, expresses it in terms of quantities that can be measured in microstructures. This is illustrated with a set of measurements based upon the theory derived from a grain growth simulation that successfully tests its predictions. It is interesting that this result also exhibits an "*n*-6 rule" that is similar to, but not identical with, that contained in von Neumann's theory.

1. Introduction

In 1951 [1] von Neumann presented what has become a classical theory of coarsening of a two dimensional soap film network at a conference on grain growth as a written discussion to a paper by Cyril Stanley Smith [2]. This theory applies Gibbs' description of the thermodynamics of systems with curved interfaces [3] to derive the result that the local velocity of an element of soap film in this coarsening process is proportional to its local curvature. The theory assumes that the motion is controlled by the rate of diffusion of atoms of the gas filling the bubbles on both sides through the film. His theory also predicts the remarkable result that the rate of growth of the area of a given soap cell is completely determined by the number of corners it has, a topological property of the cell, and is independent of the details of the geometry of the cell boundaries. He showed that cells with less than six corners shrink while those with more than six corners grow. This classical result has become known as the "*n*-6 rule"; it has fascinated researchers for more than half a century.

As a consequence researchers in grain growth and related areas have sought to extend von Neumann's theory to three dimensional cell structures. Most successful of these developments is that by MacPherson and Srolovitz [4], which starts with the analogous three dimensional version of von Neumann's prediction, i.e., the local velocity of an element of surface is proportional to its local mean curvature, and derives an equation for the rate of change of the volume of each cell in the three dimensional system:

$$\frac{dV}{dt} = -2\pi\mathcal{M}\gamma\left(\mathcal{L}(D) - \frac{1}{6}\sum_{i=1}^{n} e_i(D)\right) \tag{1}$$

where $\mathcal{M}$ is a mobility, γ is the surface tension, $\mathcal{L}(D)$ is a "natural measure of the linear size" of the polyhedral cell, an abstruse geometric property carefully defined in their text, and e_i is the length of an edge of the polyhedron and the summation is carried over the set of edges. This rigorous three dimensional derivation provides the definitive solution to the quest for a three dimensional version of von Neumann's fascinating result.

This mathematically rigorous but abstract result does not provide a pathway for designing tests of the theory, either in the physical world or in simulations. To this end this paper presents a derivation of the three dimensional version of the von Neumann equation that yields a new final result (an extension of MacPherson and Srolovitz' equation) that is also without geometric assumptions and is expressed in terms of stereological parameters that can be evaluated in simulations or serial sectioning experiments. This new result has a tantalizing doff of the cap to von Neumann's two dimensional equation involving his

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topological parameter, the number of corners per grain. However it is demonstrated that other *metric* parameters are also involved in determining the rate of volume change of a grain, and the result is thus not topological as von Neumann showed in two dimensions. This presentation focuses on the new version of the theory, and an assessment of its predictions using our grain growth simulation.

2. Kinetic theory of grain growth

The classical *phenomenological* hypothesis in the theory of Mullins and others [5–7] of surface tension driven grain growth suggests that an element of grain boundary area moves toward its center of curvature with a local velocity, v, which is proportional to the local mean curvature of that element, H:

$$v = -kH \tag{2}$$

where

$$H = \frac{1}{2}\left(\frac{1}{r_1}+\frac{1}{r_2}\right) \tag{3}$$

Here r_1 and r_2 are the local principal radii of curvature of the surface element. The conventions chosen here are defined with respect to any isolated grain. Focus on an element of area of the faces of the grain. The local velocity associated with an element of surface is defined to be positive if the element moves in the outward direction thus increasing the volume of the grain; v is negative if directed inward, contributing a decrease in the volume of the grain. Similarly the sign of each local radius of curvatures is positive if its curvature vector points toward the inside and negative if outside. Convex surface elements result when both radii are positive (H is positive), saddle surface elements occur when they have opposite signs (H may be positive or negative, depending upon the values of r_1 and r_2), while concave elements are characterized by both curvatures being negative (H is negative). Accordingly, convex surface elements move inward (v is negative), concave elements move outward (v is positive), and saddle elements may move inward or outward depending upon the values of r_1 and r_2. Small grains are bounded by mostly convex surface elements and shrink and disappear; surface elements of very large grains are mostly concave and grow.

The coefficient k is a kinetic factor containing the grain boundary energy and mobility of the boundary element.

von Neumann *derived* his kinetic equation in his two dimensional example based upon the assumption that the boundary migration is controlled by the diffusion of gas atoms through the film with boundary conditions at the two interfaces bounding the film determined by local equilibrium with the curved surfaces. Since these boundary elements are curved, local equilibrium concentrations are determined in part by the local curvature of these surfaces, the capillary shift. This curvature dependence carries through in the evaluation of the diffusion fluxes in the liquid and thence to the velocity of the moving interfaces. This scenario does not describe the grain growth process. In grain growth, there is no film separating adjacent grains: only one interface; no capillarity shifted interface concentrations determining a rate of volume diffusion; indeed there are no volume diffusion fluxes that determine the local velocity of the boundary interfaces. The inclusion of the local curvature comes not from an assumption of a capillarity shifted local interfacial equilibrium, but from a comparison of the thermodynamics at a curved interface with that at an analogous flat one. In its application to grain growth Eq. (2) is not a derived kinetic equation; indeed it has the status of a *plausible phenomenological hypothesis*.

The rate of change of the volume of any given grain may be computed from the distribution of local velocities, v, over its faces by invoking the *kinematic equation* [8]:

$$\frac{dV}{dt} = \iint_S v dS = \iint_S -kHdS = -k\iint_S HdS = -kM_S \tag{4}$$

where the integration is carried over the areas S of the surfaces of the grain faces; M_S is called the *integral mean curvature of the faces*, and thus becomes the central geometric factor in the analysis:

$$M_S = \iint_S HdS \tag{5}$$

The kinetic factor, k, is assumed to be constant in this theory. Relaxation of this assumption can lead to generalizations of the theory, but is beyond the scope of this paper.

The remainder of the development focuses upon the evaluation of M_S in terms of measurable quantities.

The integral mean curvature M of a polyhedron, see Appendix A, such as a single grain, with curved edges and faces has contributions from both its faces (S) and edges (E) [9,10]:

$$M = M_S + M_E = M_S + \frac{1}{2}\int_L \chi dL \tag{6}$$

where L is length of edges on the polyhedron and χ is the local *dihedral angle*, i.e., angle between the surface normals to the elements of cell faces that meet to form an element of edge. From Eq. (5) the geometric parameter at the center of interest in this kinetic theory of grain growth is

$$M_S = M - \frac{1}{2}\int_L \chi dL$$

or

$$M_S = M - \frac{1}{2}\frac{\int_L \chi dL}{\int_L dL}\int_L dL = M - \frac{1}{2}\langle\chi\rangle L \tag{7}$$

where $\langle\chi\rangle$ is here defined to be the average dihedral angle along the edges of the polyhedron.

The kinetic equation for any given grain in the structure may be obtained by inserting Eq. (7) into Eq. (4):

$$\frac{dV}{dt} = -k\left(M - \frac{1}{2}\langle\chi\rangle L\right) \tag{8}$$

This equation applies separately and without any simplifying geometric assumptions to *every grain in the structure*. Comparison with MacPherson and Srolovitz, Eq. (1), shows that

$$M = 2\pi\mathcal{L}(D) \text{ and } \frac{1}{2}\langle\chi\rangle L = \frac{\pi}{3}\sum_{i=1}^{n} e_i(D) \tag{9}$$

in their notation.

3. Stereological relationships

The geometric properties in Eq. (8) can be evaluated conceptually and experimentally without assumption by applying two of the fundamental relationships of stereology [11]. The stereological equations for these parameters are generally formulated in terms of values *per unit volume of microstructure*. Thus M_V is the integral mean curvature of a set of polyhedral features *per unit volume*, M_{VS} is the integral mean curvature of their faces per unit volume, and L_V is the total length of their edges per unit volume.

The pertinent stereological equations are [12–14]:

$$M_V = 2\pi N_A \tag{10}$$

and

$$L_V = 2P_A \tag{11}$$