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Tensor virial equation of evolving surfaces in sintering of aggregates of particles by diffusion

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Abstract

The moment of inertia tensor is a quantity that characterizes the morphology of aggregates of particles. The deviatoric components indicate the anisotropy of the aggregate, and its compactness is described by the isotropic component, i.e. the second moment of inertia, which is related to the radius of gyration. The equation of motion of the moment of inertia tensor is proposed for the sintering and coalescence of crystalline particles by bulk diffusion and surface diffusion. Simulations of the evolution of aggregates of particles (linear chains, rings and branched chains) show that the aggregates become more compact and more isotropic structures, driven by the surface energy tensor or the surface force density. The tensor virial equation for diffusion is applicable also to evolution of pores, precipitates and inclusions embedded in a surrounding matrix.

Keywords: Sintering; Simulation; Micromechanical modeling

1. Introduction

The tensor virial equation is the equation of motion of the moment of inertia tensor of an aggregate of particles [1]. It is obtained by first multiplying the force by position and then integrating the result over the volume of the system. While the usual scalar virial theorem applies to isotropic systems, the diagonal terms have application to anisotropic systems. The tensor virial equation is of interest in a wide variety of problems involving anisotropic isolated systems [2], for example, an aggregate of particles evolving under the action of surface tension. Chandrasekhar [3] and later Rosenkilde [4] introduced the surface energy tensor that plays an important role in the dynamics of the moment of inertia tensor.

The tensor virial method was applied to the spheroidization of a single particle in viscous sintering where inertia forces are negligible [5]. A non-spherical amorphous/glass particle relaxes to its equilibrium shape by viscous flow driven by capillarity. In this system the tensor virial equation gives a relation between the volume integral of the velocity gradient tensor (strain rate) and the surface energy tensor. The equation shows that the deformation of an isolated ellipsoidal particle is driven by the deviatoric component of the surface energy tensor in viscous sintering.

In the chemical synthesis of powders, coagulation and sintering result in the aggregation of primary particles. The morphology evolves from fractal-like open structures to compact structures by viscous sintering of amorphous multi-particle aggregates [6–8]. Both the surface area and the second moment of inertia of aggregates reduce in the morphological evolution [9,10].

The sintering of an aggregate of crystalline particles takes place by diffusion at elevated temperatures. It is well known that the shrinkage is a result of the relative motion of particles caused by grain boundary diffusion [11,12]. The thermodynamic driving force for shrinkage is the sintering force [13,14], and the sintering stress [15] that is expressed as a function of the surface energy tensor [16]. At the same

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time, each particle changes its shape by bulk diffusion [17,18], surface diffusion [19–21] and evaporation–condensation [22].

Neck formation between primary particles converts the agglomerates to aggregates. Sintering and coalescence make aggregates become more compact structures, even in the absence of grain boundary diffusion [14,23]. When the surface energy is isotropic, the bulk diffusion and surface diffusion make aggregates of crystalline particles become more isotropic structures. While the densification of bulk material is measured by the bulk density, the compactness of a multi-particle aggregate is defined by the second moment of inertia (radius of gyration) [10,24,25]. The anisotropy of the aggregate is evaluated by the deviatoric components of the moment of inertia tensor.

The objective of this paper is to show the equation of motion of the moment of inertia tensor for microstructural evolution. The structure of the paper is as follows. In Section 2, we consider tensor virial equations for bulk diffusion and surface diffusion. The anisotropic deformation by bulk diffusion is expressed as a response to the deviatoric components of the surface energy tensor. For sintering by surface diffusion, the thermodynamic driving force is identified in the equation of motion of the moment of inertia. Brakke's Surface Evolver program [26] is used as a tool to simulate the evolution of aggregates of particles (linear chains, rings and branched chains) by surface diffusion in Section 3. Although we treat only isolated aggregates in the present paper, the tensor virial equation for diffusion is applicable also to the morphological evolution of pores, cavities, precipitates and inclusions embedded in a surrounding matrix. The moment of inertia tensor will be a useful quantity to characterize the microstructural evolution (size, shape, anisotropy and orientation), because it is experimentally observable through the developments of three-dimensional imaging techniques: atom probe tomography [27], electron microscopy tomography [28], focused ion beam tomography [29] and X-ray tomography [30].

2. Dynamics of evolving interface

2.1. Bulk diffusion

Microstructural development involves evolving interfaces and surfaces. When matter is transferred to/from the bulk, the normal velocity v_n of the surface is proportional to the flux j_n normal to the surface:

$$v_n = j_n \Omega \tag{1}$$

where Ω is the atomic volume. The diffusive flux in the bulk is proportional to the gradient of a chemical potential μ:

$$\mathbf{j} = -\frac{D_L}{kT\Omega}\nabla\mu \tag{2}$$

where D_L is the diffusion coefficient in the bulk, k is the Boltzmann constant and T is the absolute temperature. Mass conservation in the bulk gives

$$\nabla^2\mu = 0 \tag{3}$$

Herring [17,31] has shown that the chemical potential just below the surface is

$$\mu - \mu_0 = -\gamma_s \kappa \Omega \tag{4}$$

where μ_0 is the chemical potential under a flat surface and γ_s is the surface energy. The curvature κ, the sum of the principal curvatures (note this is twice the traditional definition of mean curvature), is expressed as the divergence of the unit (outward) normal vector n_i to the surface:

$$\kappa = -\partial n_k/\partial x_k \tag{5}$$

The summation convention for repeated indices is applied throughout this paper. The curvature is defined positive when the center of curvature is outside of the particle: it is negative $-2/r_0$ for a spherical particle with radius r_0.

We consider a body of uniform density with volume V enclosed by a surface A. The body may be a single isolated particle or an aggregate of particles. The moment of inertia tensor of the body about the position of the center of mass is defined by

$$I_{ij} = \int_V x_i x_j dV \tag{6}$$

Thus,

$$\frac{d}{dt}I_{ij} = \int_V (x_i v_j + v_i x_j) dV \tag{7}$$

where velocity is $v_i = dx_i/dt$. We regard the velocity field inside the particle as

$$v_i = j_i \Omega \tag{8}$$

From Eqs. (8) and (2), we obtain

$$\int_V x_j v_i dV = -\frac{D_L}{kT}\int_V x_j \frac{\partial \mu}{\partial x_i} dV \tag{9}$$

After integrating by parts, and using the divergence theorem, we have

$$\int_V x_j \frac{\partial \mu}{\partial x_i} dV = \int_A x_j \mu n_i dA - \delta_{ij}\int_V \mu dV \tag{10}$$

By substituting the boundary condition, Eq. (4), into Eq. (10), we obtain

$$\int_V x_j \frac{\partial \mu}{\partial x_i} dV = -\Omega\int_A \gamma_s \kappa n_i dA + \mu_0 \int_A x_j n_i dA - \delta_{ij} \times \int_V \mu dV \tag{11}$$

From the identity

$$\int_A x_j n_i dA = \int_V \frac{\partial x_j}{\partial x_i} dV = V\delta_{ij} \tag{12}$$

the tensor virial equation for bulk diffusion takes the form

$$\frac{1}{V}\frac{d}{dt}I_{ij} = -\frac{2D_L\Omega}{kT}\left(\frac{2S_{ij}}{V} - \delta_{ij}\bar{p}\right) \tag{13}$$