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Sintering force behind the viscous sintering of two particles

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ABSTRACT

The mechanical principles of viscous sintering were analyzed by finite element simulation of a simple model: the coalescence of two identical spheres. The sintering force in the non-equilibrium process of viscous sintering was defined as the difference between the average pressure on the contact area and the surface tension along its circumference. The average strain rate on the contact plane was proportional to the sintering force, so that the sintering force was the thermodynamic driving force for both neck growth and shrinkage. Conversely, a theoretical method was proposed to elicit the sintering force from neck growth curves. The simulation also shows that the tensor–virial equation, which is an alternative method to describe the overall anisotropic deformation of aggregates of particles, is valid for viscous sintering.

1. Introduction

When two liquid drops touch, they coalesce into a single bigger drop driven by surface tension to decrease the total surface area [1,2]. The physical principle of this particle coalescence is common to viscous sintering of glasses and polymers at temperatures higher than their glass transition. While the microstructural evolution during sintering of a huge number of particles is very complicated, the sintering of two identical spherical particles is a simple model for understanding sintering phenomena as proposed by Frenkel [3]. A circular contact area is formed at the point of contact of spheres. As its radius increases with time, the length of the particle pair decreases. The neck growth and the shrinkage are essential characteristics of sintering. The purpose of the present paper is to reveal that the sintering force is the thermodynamic driving force for both neck growth and shrinkage in viscous sintering. Conversely, the sintering force can be elicited by the observation of neck growth rate.

The sintering force is originally defined for equilibrium states, where the mechanical force just balances the surface tension forces so that the porous materials do not shrink [4]. It is determined rigorously for a row of particles [5,6], a constrained particle pair [7], and periodic porous structures under the constraint of fixed volume [8–12]. The sintering force in equilibrium states is independent of the details of sintering kinetics, but, depends on only geometry and the ratio of grain boundary energy to surface energy. In equilibrium states, the sintering force is defined from energy, curvature, and mechanical force, and all three methods give the identical value [11]. On the other hand, in non-equilibrium processes, the sintering force is directly related to the sintering kinetics, e.g., the relative velocity between two crystalline particles is proportional to the sintering force in the sintering by coupled grain boundary diffusion and surface diffusion [13,14]. The macroscopic shrinkage of a powder compact is described by bulk viscosity and sintering stress in the continuum model of sintering [15–19]. The analysis of sintering force between two particles is a basis for bridging the microscopic and the macroscopic models, because the sintering stress arises from sintering forces acting among a huge number of particles in the initial stage of sintering.

For the viscous sintering of two glass spheres, most of works have focused mainly on neck growth and shrinkage. Frenkel's approximate model on neck growth was corrected by Eshelby [20] and has been referred to as the Frenkel–Eshelby model. However, this model overestimates the neck growth rate, and does not match to the results of numerical simulations by finite element method (FEM) [21,22], boundary element method (BEM) [23], and boundary integral method [24,25]. A modified Frenkel model was later

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proposed to approximate the neck growth in all stages of sintering [26]. The numerical simulations agreed well with experimental results [27]. Numerical simulations have been further extended to the sintering of aggregates of many particles [28–30]. Moreover two dimensional problems were solved for sintering of cylinders [31–33]. But, only Jagota and Dawson [21] analyzed the sintering force in viscous sintering by using the energy method. We found that the sintering force defined by a mechanical force is appropriate to describe the shape evolution of a single ellipsoidal particle [34]. In the present paper we show that the sintering force drives the local flow field near the contact plane in non-equilibrium process of viscous sintering of two spherical particles. The neck growth and the shrinkage are related to this local flow field. We discuss also the tensor–virial equation which describes the overall deformation of aggregates of particles in viscous sintering.

2. Mechanics of viscous sintering

The sintering of glasses and polymers occurs by viscous flow driven by surface tension. The stress in a viscous fluid is expressed by

$$\sigma_{ij} = -p\delta_{ij} + \mu\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right) \tag{1}$$

where u_i is the velocity, μ is the viscosity, $p = -\sigma_{ii}/3$ is the pressure. We consider a very small particle with high viscosity, then, the deformation of the particle is described by Stokes equation [35].

$$\frac{\partial p}{\partial x_i} = \mu\frac{\partial^2 u_i}{\partial x_j \partial x_j}. \tag{2}$$

The summation convention for repeated indices is applied throughout this paper. The mass conservation in incompressible flow is expressed as

$$\frac{\partial u_i}{\partial x_i} = 0. \tag{3}$$

The boundary condition on the surface is

$$-pn_i + \mu\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right)n_j = \gamma_s \kappa n_i \tag{4}$$

where γ_s is the surface energy, n_i is the unit (outward) normal to the surface, and $\kappa = \mathrm{div}\,\mathbf{n}$ is the curvature. The curvature is defined that it is negative for a spherical particle.

Imagine two identical particles touch as shown in Fig. 1, and cut it into two parts V and V' at the contact area A. The total force acting on the part V, that is, the integral of forces $F_i = \partial\sigma_{ij}/\partial x_j$ distributed over V, is a sum of the traction on the surface S of the part V and the traction spread over the contact area A.

$$\int_V F_i \mathrm{d}V = \int_V \frac{\partial \sigma_{ij}}{\partial x_j}\mathrm{d}V = \int_S \sigma_{ij} n_j \mathrm{d}S + \int_A \sigma_{ij} n_j \mathrm{d}S \tag{5}$$

Here, we used the divergence theorem of Gauss. We assume the surface energy is constant. From the boundary condition of the surface, Eq. (4), and the application of Stokes theorem (see Appendix B in Ref. [14]), the first term on the right hand side of Eq. (5) is given as the line integral along the contour C of the cross-section A

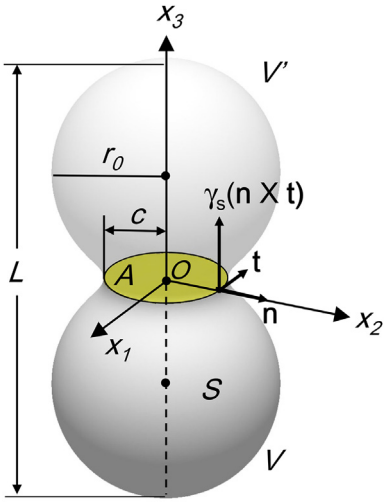

Fig. 1. Geometry of the two particle model in three dimensions. The particle on the upper side is translucent to show the contact area.

$$\int_S \sigma_{ij} n_j \mathrm{d}S = \int_S \gamma_s \kappa n_i \mathrm{d}S = \int_C \gamma_s (\mathbf{n}\times\mathbf{t})_i \mathrm{d}r \tag{6}$$

where $\mathbf{n}$ is the unit normal vector to the surface, and $\mathbf{t}$ is the unit tangent vector along the contour C. Substituting Eq. (1), the second term on the right hand side of Eq. (5) is expressed as

$$\int_A \sigma_{ij} n_j \mathrm{d}S = -\int_A p n_i \mathrm{d}S + \mu\int_A \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right)n_j \mathrm{d}S \tag{7}$$

In the sintering of two identical spheres, the total force acting on the part V is zero. Substituting Eqs. (6) and (7) into Eq. (5), the axial component is given as

$$\gamma_s l - \bar{p}A + 2\mu\int_A \frac{\partial u_3}{\partial x_3}\mathrm{d}S = 0 \tag{8}$$

where $A = \pi c^2$ is the cross sectional area, c is the contact radius (the neck radius), $l = 2\pi c$ is the circumference, and $\bar{p}$ is the average pressure on the contact plane

$$\bar{p} = \frac{1}{A}\int_A p \mathrm{d}S \tag{9}$$

The average strain rate on the contact area is defined as

$$\dot{E}_{ij} = \frac{1}{2A}\int_A \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right)\mathrm{d}S \tag{10}$$

From Eq. (8), the axial strain rate, which is related to the shrinkage, is expressed as

$$\dot{E}_{33} = -\frac{F^s}{2\mu A} \tag{11}$$

Here, the sintering force F^s is defined as the difference between the average pressure on the contact plane and the surface tension $\gamma_s l$ along the circumference

$$F^s = -\bar{p}A + \gamma_s l \tag{12}$$