

Mechanics of viscous sintering on the micro- and macro-scale

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Abstract

The macroscopic quantities in a continuum model of viscous sintering are determined from analysis of the dynamic evolution of pore structures on the microscopic scale that is governed by the principle of fluid mechanics. A single ellipsoidal pore shrinks to be more anisotropic due to local viscosity, even though the surface tension acts to make it more spherical. The constitutive equation is derived by defining macroscopic quantities as volume averages over a volume element which contains many pores. The presence of dispersed pores generates a bulk stress that affects the macroscopic bulk viscosity and the shear viscosity. The hydrostatic component of macroscopic sintering stress is simply calculated from the total pore volume and the total pore area without knowing their surface curvature. The sintering stress, and then the shrinkage rate, are predicted rigorously from the distribution function of pore size.

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1. Introduction

The sintering of glass, amorphous preform, gel and polymer takes place at elevated temperatures due to viscous flow driven by applied stress and the surface tension according to the principles of fluid mechanics [1,2]. The theories of viscous sintering proposed by Frenkel [3], Mackenzie and Shuttleworth [4] and Scherer [5,6] use a thermodynamic assumption which states that the heat released by viscous dissipation is equal to the total work done by surface tension to reduce the surface area.

The macroscopic constitutive equation of viscous sintering for an isotropic system is expressed as [7–10]:

$$\dot{E}_{ij} = \frac{\Sigma'_{ij}}{2G} + \delta_{ij} \frac{(\Sigma_m - \Sigma^s)}{3K}, \quad (1)$$

where \dot{E}_{ij} is the macroscopic strain rate, Σ'_{ij} and Σ_m are deviatoric and hydrostatic components of macroscopic stress, Σ^s is the sintering stress, and G and K are the shear viscosity and the bulk viscosity, respectively. Note that the

sign convention used for sintering stress, or sintering pressure [6], is opposite to that used for stress, since pressure is positive if it is compressive. These macroscopic quantities depend not only on the relative density, but also on the structures on a particle scale, e.g. the shape and structure of pores [11] and the distribution function of pore size [12,13]. On the other hand, the microstructural evolution in viscous sintering is predicted by solving the Stokes equation numerically [14–17], e.g. by using the finite-element method [18]. It is important to study the relationship between the macroscopic properties and the microscopic structures in terms of fluid mechanics.

The purpose of the present paper is to derive the macroscopic constitutive equation of viscous sintering directly from the principles of fluid mechanics. In Section 2 the dynamics of a single pore is analyzed by an integral equation including the surface energy tensor. The densification is driven by the hydrostatic component of sintering stress, and the deviatoric components are the driving forces for the spheroidization of pores. The deformation of an ellipsoidal pore in shear flow is solved by using Eshelby's method [19,20]. In Section 3, Batchelor's [21] formulation of a macroscopic stress system in suspension is applied to

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viscous materials containing many pores of different sizes and shapes. A complete formulation is presented here in order to make this paper self-contained upon first reading. The presence of pores generates a bulk stress, which affects the bulk viscosity and the shear viscosity. Numerical simulations of viscous sintering in microscopic and macroscopic scales have been performed in Section 4. The effects of pore size distribution on macroscopic sintering stress and densification rate are investigated theoretically, and compared with experiments.

The present analysis provides a method to calculate the macroscopic sintering stress of a volume element containing many pores of any shape and size from the total pore volume and the total pore area without knowing their surface curvatures. Since high-resolution X-ray microtomography makes it possible to visualize the real three-dimensional microstructural evolution in sintering [22–24], our method will be useful to estimate the sintering stress from the knowledge of microstructure.

2. Dynamics of a single pore in viscous sintering

2.1. Stokes equation

We consider the shrinkage and deformation of a single pore in an incompressible viscous material. The stress in the matrix is expressed by:

$$\sigma_{ij} = -p\delta_{ij} + \mu \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right), \quad (2)$$

where u_i is the velocity, μ is the viscosity and $p = -\sigma_{ii}/3$ is the pressure. When the Reynolds number of the relative motion of the viscous matrix near one pore is small compared to unity, the Stokes equation and mass conservation are [1]:

$$\frac{\partial p}{\partial x_i} = \mu \frac{\partial^2 u_i}{\partial x_k \partial x_k} \quad (3)$$

$$\frac{\partial u_i}{\partial x_i} = 0. \quad (4)$$

The summation convention for repeated indices is applied throughout this paper. The boundary condition on the pore surface is:

$$-pn_i + \mu \left(\frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i} \right) n_k = \gamma_s \kappa n_i, \quad (5)$$

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

We will assume that the surface velocity of the pore can be separated into two parts: the contribution u_i^s driven by the surface tension, and the contribution u_i^{flow} induced by the macroscopic shear flow \dot{E}_{ij} :

$$u_i = u_i^s + u_i^{\text{flow}}. \quad (6)$$

2.2. Spheroidization of a pore bubble

The deformation of a single pore due to the surface tension (Fig. 1) is divided into the spheroidization and the shrinkage. The spheroidization, where the pore volume is kept constant, is analyzed by considering a bubble filled with an inviscid incompressible fluid in Fig. 1b.

We consider a volume integral of the stress over a region that includes the bubble. The contribution to the volume integral from the portion of the interfacial surface A_n lying within the integration volume V_n is (see Appendix in Ref. [21]):

$$2S_{ij}^{\text{energy}} = \int_{A_n} \gamma_s (\delta_{ij} - n_i n_j) dA, \quad (7)$$

where S_{ij}^{energy} is the surface energy tensor [25], the trace of which is the total surface energy $\Gamma_n = S_{ii}^{\text{energy}} = \gamma_s A_n$ of the bubble. With V_n^+ denoting the volume of a bubble bounded by the closed surface A_n^+ on the outer side of the interface layer, and V_n^- and A_n^- the corresponding quantities for the inner side of the same interface layer, we have:

$$\int_{V_n^+} \sigma_{ij} dV - \int_{V_n^-} \sigma_{ij} dV = \int_{A_n} \gamma_s (\delta_{ij} - n_i n_j) dA. \quad (8)$$

Substituting Eq. (2) into Eq. (8), we have:

$$\begin{aligned} - \int_{V_n^+} p \delta_{ij} dV + \mu \int_{V_n^+} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) dV + \int_{V_n^-} p^* \delta_{ij} dV \\ = \int_{A_n} \gamma_s (\delta_{ij} - n_i n_j) dA, \end{aligned} \quad (9)$$

where p^* is the pressure inside the bubble.

Here we consider an ellipsoidal pore, because Eshelby [19,20] found that the assumption of an ellipsoidal shape greatly simplifies the problem. The general ellipsoid is versatile enough to cover a wide variety of particular cases, e.g. cylinder-like pore, spherical pore and flat crack-like pore. In an ellipsoidal pore, we assume the strain rate is independent of position, consistent with Eshelby's analysis. The strain rate $\dot{\epsilon}_{ij}$ and the velocity are related by:

$$\dot{\epsilon}_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right). \quad (10)$$

We have:

$$-\delta_{ij} p V_n + 2\mu \dot{\epsilon}_{ij} V_n + \delta_{ij} p^* V_n = \int_{A_n} \gamma_s (\delta_{ij} - n_i n_j) dA. \quad (11)$$

Since the bubble volume is kept constant, the strain rate tensor $\dot{\epsilon}_{ij}$ is traceless. The pressure inside the bubble must balance with the hydrostatic component of the surface energy tensor and the pressure in the matrix:

$$p^* = p + \frac{2\gamma_s A_n}{3V_n}. \quad (12)$$

We define the sintering stress of a pore of arbitrary shape as:

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