SIMULATION OF SINTERING OF LAYERED STRUCTURES

E. Olevsky[*], V. Tikare[**], T. Garino[**], and M. Braginsky[**]

[*]Department of Mechanical Engineering, San Diego State University, 5500 Campanile Dr., San Diego, CA 92182-1323
[**]Sandia National Laboratories, Albuquerque, NM 87185, USA

Abstract - An integrated approach, combining the continuum theory of sintering and Potts model based mesostructure evolution analysis, is used to solve the problem of bi-layered structure sintering. Two types of bi-layered structures are considered: layers of the same material with different initial porosity, and layers of two different materials. The effective sintering stress for the bi-layer powder sintering is derived, both at the meso- and the macroscopic levels. Macroscopic shape distortions and spatial distributions of porosity are determined as functions of the dimensionless specific time of sintering. The effect of the thickness of the layers on shrinkage, warpage, and pore-grain structure is studied. Ceramic ZnO powders are employed as a model experimental system to assess the model predictions..
KEYWORDS: SINTERING, CONTINUUM MODELING, MESOSCALE SIMULATION

I. INTRODUCTION

The present work is an attempt to link two different length scales of material description: meso and macro, enabling a new continuum-structural modeling approach. Material constitutive parameters, in particular, the effective sintering stress and the bulk modulus, are determined by a mesoscale Monte-Carlo simulation of the sintering of a 2-D powder compact. These parameters are calculated as functions of one structural characteristic - porosity.

The parameters obtained from the mesoscale Monte Carlo simulations are substituted into a finite-element code based on the continuum model of sintering. Here we demonstrate the application of this code to simulate sintering of bi-layered ceramic composites.

II. DETERMINATION OF CONSTITUTIVE PARAMETERS

The continuum theory of sintering [1] is used to simulate the mechanical response of a nonlinear-viscous porous body subjected to sintering and an external load by a rheological (constitutive) equation that relates the components of stress tensor σ_{ij} and strain rate tensor $\dot{\varepsilon}_{ij}$:

$$\sigma_{ij} = \frac{\sigma(W)}{W}\left[\varphi\dot{\varepsilon}_{ij} + \left(\psi - \frac{1}{3}\varphi\right)\dot{e}\delta_{ij}\right] + P_L\delta_{ij} \quad (1)$$

where W is the "equivalent strain rate", and $\sigma(W)$ is the "equivalent stress"; φ and ψ are the shear and bulk viscosity moduli, which depend on porosity θ as formalized in [2], $\varphi = (1-\theta)^2$, $\psi = \frac{2}{3}\frac{(1-\theta)^3}{\theta}$); δ_{ij} is a Kronecker symbol ($\delta_{ij} = 1$ if i=j and $\delta_{ij} = 0$ if $i \neq j$); $\dot{e}$ is the first invariant of the strain rate tensor, i.e. sum of tensor diagonal components: $\dot{e} = \dot{\varepsilon}_{11} + \dot{\varepsilon}_{22} + \dot{\varepsilon}_{33}$. The porosity θ is defined as $1 - \frac{\rho}{\rho_T}$, where ρ is density, ρ_T is full theoretical density and ρ/ρ_T is relative density. Physically, $\dot{e}$ represents the volume change rate of a porous body.

Equivalent strain rate W is related to current porosity by the invariants of the strain rate tensor:

$$W = \frac{1}{\sqrt{1-\theta}}\sqrt{\varphi\dot{\gamma}^2 + \psi\dot{e}^2} \quad (2)$$

$\dot{\gamma}$ is the second invariant of the strain rate tensor deviator:

$$\dot{\gamma} = \left[\left(\dot{\varepsilon}_{ij} - \frac{1}{3}\dot{e}\delta_{ij}\right)\left(\dot{\varepsilon}_{ij} - \frac{1}{3}\dot{e}\delta_{ij}\right)\right]^{1/2} \quad (3)$$

Physically, $\dot{\gamma}$ represents the shape change rate of a porous body. It can be expressed in terms of the main elongation rates: $\dot{\varepsilon}_1, \dot{\varepsilon}_2, \dot{\varepsilon}_3$:

$$\dot{\gamma} = \frac{1}{\sqrt{3}} \sqrt{(\dot{\varepsilon}_1 - \dot{\varepsilon}_2)^2 + (\dot{\varepsilon}_2 - \dot{\varepsilon}_3)^2 + (\dot{\varepsilon}_3 - \dot{\varepsilon}_1)^2} \quad (4)$$

$\sigma(W)$ determines the constitutive behavior of a porous material. If $\sigma(W)$ is described by the linear relationship: $\sigma(W) = 2\eta_0 W$, where η_0 is the shear viscosity of a fully-dense material, one obtains the equation describing the behavior of a linear-viscous porous body:

$$\sigma_{ij} = 2\eta_o \left[\varphi \dot{\varepsilon}_{ij} + \left(\psi - \frac{1}{3}\varphi \right) \dot{e} \delta_{ij} \right] + P_L \delta_{ij} \quad (5)$$

P_L is an effective sintering stress.

In the constitutive relationship (1), three main parameters P_L, φ, and ψ should be determined as functions of porosity.

Effective sintering stress

In the framework of the continuum theory of sintering, the effective Laplace pressure (sintering stress) P_L is defined:

$$P_L = \frac{\partial F}{\partial \vartheta} \quad (6)$$

where F is surface free energy, and ϑ is a porous volume subjected to sintering.

Effective sintering stress is the result of the collective action of local capillary stresses in a porous material. The
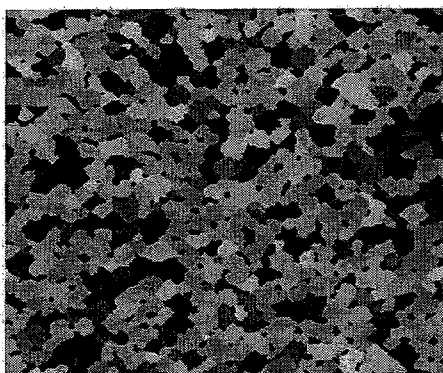

Fig. 1 Microstructure
Colored features are grains, black features are pores

relationship between the effective and local Laplace pressure depends on the procedure of averaging the local capillary stresses over a mesoscopic porous volume.

We determine the effective sintering stress using an original approach based upon mesoscale simulations of sintering in a 2-D
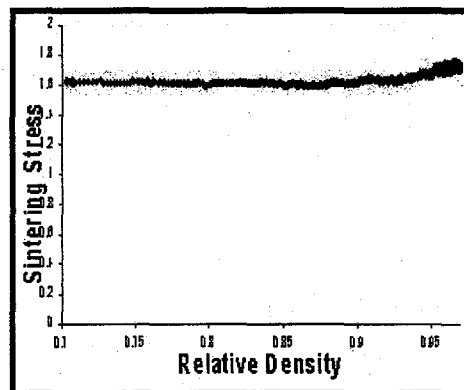

Fig. 2 Dependence of the effective sintering stress on porosity as a result of the mesoscale simulations

porous microstructure. At each step of the microstructure evolution the shrinkage and the free energy of the grain-pore surface, shown in Fig. 1, are calculated. The effective sintering stress as a function of relative density is determined using relationship (6) and is plotted in. Fig. 2. Applying regression analysis, the following analytical expression for the effective sintering stress as a function of porosity is obtained:

$$\overline{P}_L = 1.7(1 - \theta)^{0.26} \quad (7)$$

Effective normalized bulk modulus.

Using the kinetics of shrinkage and the dependence of the effective sintering stress on porosity obtained from mesoscale simulations, one can determine the effective bulk modulus. The corresponding regression analysis of the numerical data enables the following analytical expression for ψ:

$$\psi = \frac{2}{3} \frac{(1-\theta)^{2.23}}{\theta^{1.12}} \quad (8)$$

III. SOLUTION OF THE PROBLEM OF SINTERING OF BI-LAYERED CERAMIC COMPOSITES

When incorporated in a finite-element code, the continuum theory of sintering allows the solution of one of the most important problems of co-firing multilayer ceramic composites - it can predict warpage caused by differential sintering.

In the case of linear-viscous properties of the porous body skeleton, the finite-element approximation of equation (5) is immediately reduced to the solution of a

set of linear equations describing the nodal velocities. Using the conventional finite-element analysis designations, one can represent this set in the form:

$$\left[\int_{\vartheta}[B]^T[D][B]d\vartheta\right]\{V_n\} = -\int_{\vartheta}[B]^T P_L\{1\}d\vartheta \quad (9)$$

where [B] is the matrix correlating the strain rates with the nodal velocities V_n; [D] is the matrix correlating the stresses with the strain rates ("matrix of viscosities"); ϑ is a macroscopic porous volume under investigation; g is the gravity acceleration. The right-hand part of the latter equation represents the nodal forces. If material properties are uniformly distributed in the volume ϑ, the nodal forces associated with the sintering stress P_L (the first term in the right-hand part of Eq. (9)) will be zero everywhere except at the nodes which belong to the external boundary of ϑ. The
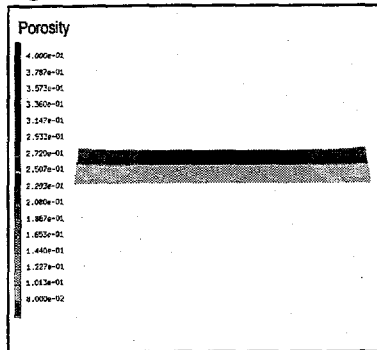

Fig. 3 Sintering of a bi-porous structure on a rigid substrate. Initial porosity is 40% in top layer and 20% in bottom

multiplier $\left[\int_{\vartheta}[B]^T[D][B]d\vartheta\right]$ in the left-hand part corresponds to the coefficients in the set of linear equations (9) of the unknown nodal velocities V_n.

The effective sintering stress and the effective normalized bulk modulus given by equations (7) and (8) are used. The effective normalized shear modulus from the Skorohod model [2] is used:

$$\varphi = (1-\theta)^2 \quad (10)$$

After the solution of set (9), the field of strain rates is calculated. The new values of relative densities are calculated using the continuity equation for each finite element:

$$\frac{\dot{\theta}}{1-\theta} = \dot{e} \quad (11)$$

This finite-element algorithm is implemented for the solution of three problems of sintering of bi-layer structures.
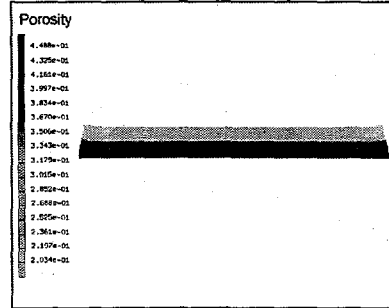

Fig. 4 Sintering of a bi-layered structure on a rigid substrate. Initial porosity in both layers is 40%, sintering stress ratio is 1:50

The first two problems consider the sintering of bi-layers on a rigid substrate. The first is sintering of a bi-porous structure (initial porosities are 40 % for the top layer and 20% for the bottom layer) with the same chemical composition or equivalently, the same sintering stress and bulk modulus for a given porosity. The second problem is sintering of a bi-layer structure with layers of the same initial porosity (40%) and different shrinkage rates (sintering stress ratio is 1:50). One can think of these bilayers as having different surface energies resulting in different sintering stress. The results of the simulations are shown in Figs. 3 and 4, respectively. In both cases the densification front propagates starting from the top peripheral area that has the highest density. The bottom peripheral zone has the highest porosity due to the imposed constrain from the rigid substrate. In both cases a porous core is formed in the top layer. The simulations indicate that difference in chemical composition of adjacent layers resulted in the development of higher stresses and larger density gradients than those for difference in porosity.

The third problem that was considered is free sintering of a bi-layer structure with the uniform initial distribution of porosity (40%) and with different shrinkage rates for the two layers. The ratio of the effective sintering stresses is assumed

to be 1:50. Fig. 5 shows a 3-D image of the sintered cylindrical bi-layer specimen with its porosity spatial distribution. Due to the difference in shrinkage rates, the top layer contracts faster, causing the pronounced bending of the specimen.
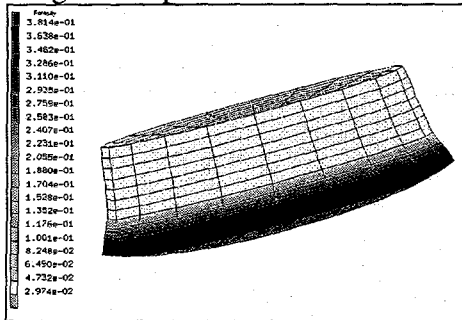
Fig. 5 Free sintering of a bi-layered structure. Initial porosity for both layers are 40%, sintering stress ratio is 1:50

To experimentally validate the predicted results, sintering of a bilayered ZnO powder specimen was investigated. Solid phase shear viscosity data of Aldrich 0.2 nm ZnO was measured under cyclic loading.

The following relationship was obtained for the dependence of the viscosity on the temperature:

$$\eta_0(T) = \left[51.7\left(\frac{T}{750}\right)^2 - 106.6\frac{T}{750} + 56.4\right]10^{10}\,\text{Pa·s} \quad (12)$$

and the surface tension of the ZnO powder was assumed to be:

$$\alpha = 1.27\,J/m^2 \quad (13)$$

These same parameters were used to calculate the sintering deformation in the continuum simulations. The results are shown in Fig.6.

A bi-layered ZnO with one layer of 47% initial density and the other of 57% was sintered.. Final dimensions were 6.7 mm(long) x 2.15 mm (thick) x 3.89 mm (deep). The results of the experiment are shown in Fig.6. One can see that the modeling results agree satisfactorily with the experimental data on the sample distortion.

IV. CONCLUSIONS

- The success of modeling at the macroscopic level depends on the accurate determination of constitutive parameters of porous structure. Mesoscale simulations can be used to determine material constitutive parameters.
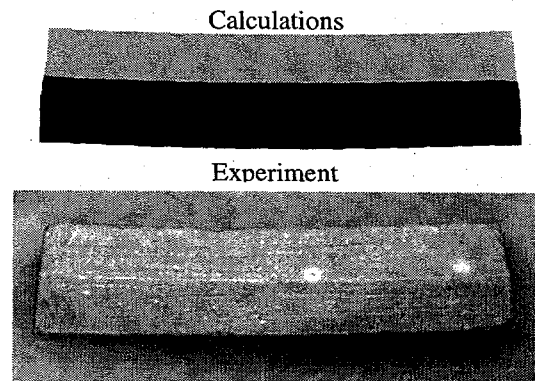
Fig.6 Comparison of the calculation and experimental data on sintering of a biporous ZnO powder specimen

- The continuum theory of sintering can be used to predict dimensional changes of bilayer system for both free and constrained sintering.
- The simulation results agree satisfactorily with the experimental results of ZnO bilayer sintering.

ACKNOWLEDGMENT

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