201 Internal Stress Diagrams of Sintering Stress versus Viscosity for Graded Multilayers

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Abstract: The internal stress of graded powder compacts during sintering is analyzed by using plate theory with shrinkage and warpage taken into consideration. Diagrams giving the in-plane stress on the surface of the compacts, produced by mismatch and bending, are presented as a function of sintering stress and viscosity. The diagrams show the contours of tension and compression as well as the direction of warpage for given multilayered substrate. When the viscosity of the top layer is smaller than that of the substrate, the shrinkage constrained by the substrate significantly influences the internal stress. In the opposite case, the bending stress dominates the stress condition. The tension/compression boundaries in the diagrams change according to the number and the thickness of the graded layers as well as the sintering stress and the viscosity.

Key Words: Functionally Graded Materials, Powder Metallurgy, Stress Analysis

1. INTRODUCTION

Functionally graded materials (FGMs) for high temperature applications have been developed to eliminate the thermal stress in metal/ceramic laminates [1, 2]. When FGMs are fabricated by powder processing, the internal stress generated during sintering must be also suppressed because it often produces cracks in sintered bodies. Cracking may occur even under the low tensile stress when the particle contacts are small, that is, at the early stage of sintering. For the optimal design of the FGMs, it is important to examine the conditions to produce tensile stresses in the graded powder compacts during sintering.

Many researchers have been analyzed the mismatch stress in constrained densification in sintering, especially, for the thin film on a rigid substrate [3-7] or symmetrical three layers [8-10]. With respect to the graded powder compacts, a finite element method for sintering has been applied to predict the distortion as well as the internal stress [11-15]. In a finite element analysis of warping of FGMs during sintering, the tensile stress appears on the top layer, as a result of a combination of the bending stress and the mismatch stress, which may cause cracks on the surface [16].

The finite element method is a powerful tool for design of products in various shapes, but the analysis by the plate theory may be also useful to realize the nature of graded multilayers. In this study, surface cracking at the early stage of sintering is focused and how the sintering properties and the graded structures affect the internal stress on the top surface is analyzed by using the plate theory based on viscoelastic/elastic analogy [12]. To display the analytical results, diagrams giving the in-plane stress are presented as a function of sintering stress and viscosity.

2. ANALITICAL METHOD

2.1 Constitutive model

As an example of a constitutive model for sintering, the following equation, proposed in the previous study [17] is considered;

$$\dot{\varepsilon}_{ij} = \frac{1}{2\eta} \frac{1}{\rho^{2n-1}} \left\{ \sigma'_{ij} + \delta_{ij} \frac{2}{9f^2} (\sigma_m + \sigma_s) \right\} \quad (1)$$

where $\dot{\varepsilon}_{ij}$ is the strain rate ($i, j = x, y, z$), σ'_{ij} is the deviatoric stress, η is the viscosity, ρ is the relative density, n is a constant, f is a function of the relative density representing the degree of influence of the hydrostatic stress component and σ_s is the sintering stress. According to the

viscoplastic/elastic analogy, Eq. (1) can be rewritten as

$$\begin{aligned}\dot{\epsilon}_{ij} &= \frac{1+\nu}{E} \sigma'_{ij} + \delta_{ij} \frac{1-2\nu}{E} (\sigma_m + \sigma_s) \\ E &= 3\eta\rho^{2n-1} \frac{9f^2}{9f^2+1} \\ \nu &= \frac{9f^2-2}{2(9f^2+1)}\end{aligned}\quad (2)$$

where E is the modulus of longitudinal viscosity and ν is Poisson's ratio, which is taken to be 0.2 in the present study. Free sintering strain rate ($\sigma'_{ij} = \sigma_m = 0$) is given by

$$\dot{\epsilon}_s = \frac{1-2\nu}{E} \sigma_s \quad (3)$$

In this paper, compression is taken to be positive.

2.2 Plate theory

The in-plane normal strain rate can be expressed as [12]

$$\dot{\epsilon}_x = \dot{\epsilon}_0 + \kappa z - \dot{\epsilon}_s \quad (4)$$

where $\dot{\epsilon}_x$ is the in-plane strain rate of the middle plane ($z=0$), κ is the evolving curvature of the plane. Substituting Eqs. (2) and (3) into Eq. (4) under the conditions of plain stress ($\sigma_z = \tau_{yz} = \tau_{zx} = 0$) and isotropy of material gives

$$\begin{aligned}\sigma_x &= \frac{E}{1-\nu} \dot{\epsilon}_x \\ &= \frac{E}{1-\nu} \left\{ \dot{\epsilon}_0 - (1-2\nu) \frac{\sigma_s}{E} \right\} + \frac{Ez}{1-\nu} \kappa\end{aligned}\quad (5)$$

Equilibrium equations of the stress and the moment can be written as

$$\dot{\epsilon}_0 \int \frac{E}{1-\nu} dz + \kappa \int \frac{Ez}{1-\nu} dz - \int \sigma_s \frac{1-2\nu}{1-\nu} dz = 0 \quad (6)$$

$$\dot{\epsilon}_0 \int \frac{Ez}{1-\nu} dz + \kappa \int \frac{Ez^2}{1-\nu} dz - \int \sigma_s \frac{1-2\nu}{1-\nu} z dz = 0 \quad (7)$$

Consider L layers with each thickness of h as shown in Fig. 1. Since the l th layer is divided by m_l elements, the thickness of each layer is given by

Fig. 1 Modeling of multilayers

(a) With different sintering stresses.

(b) With different viscosities.

Fig. 2 Stress distributions in multilayers.

$$H = m_1 h \quad (8)$$

and the total thickness of the multilayer is

$$T = \sum_1^L H_1 = h \sum_1^L m_1 = 2Nh \quad (9)$$

where $2N$ is the total number of the elements. Solving Eqs. (6) and (7) gives

$$\dot{\epsilon}_0 = -\frac{\kappa h}{2} \frac{\Delta \bar{E}}{\bar{E}} + \frac{\bar{\sigma}_s}{\bar{E}} \quad (10)$$

$$\dot{\kappa} = -\frac{1}{h} \frac{6(\Delta \bar{E} \cdot \bar{\sigma}_s - \Delta \bar{\sigma}_s \cdot \bar{E})}{4\bar{e} \cdot \bar{E} - 3\Delta \bar{E}^2} \quad (11)$$

$$\Delta \bar{E} = \sum_k^N \{(p_{uk} E_{uk} - p_{wk} E_{wk})(2k-1)\}$$

$$\bar{E} = \sum_k^N (p_{uk} E_{uk} + p_{wk} E_{wk})$$

$$\bar{e} = \sum_k^N \{(p_{uk} E_{uk} + p_{wk} E_{wk})(3k^2 - 3k + 1)\}$$

$$\bar{\sigma}_s = \sum_k^N (q_{uk} \sigma_{suk} + q_{wk} \sigma_{swk})$$

$$\Delta \bar{\sigma}_s = \sum_k^N \{(q_{uk} \sigma_{suk} - q_{wk} \sigma_{swk})(2k-1)\}$$

$$p = \frac{1}{1-\nu}, \quad q = \frac{1-2\nu}{1-\nu}$$

where subscripts u, w represent upper and lower side of the multilayer from the middle plane, respectively.

3. RESULTS

3.1 Internal stress distribution

The internal stress consists of the mismatch stress and the bending stress, that is, the first and the second term of the right hand side of Eq. (5), respectively. Figure 2 shows the stress distributions with the two terms for some three layers as examples. When the sintering stress of the top layer ($k=1$, $z/T=0.16 \sim 0.5$) is small, the correspondent mismatch stress is compressive (+) as shown in Fig. 2(a), because the top layer shrinks along with the substrate. The top layer, however, becomes convex side of warping and the total on the surface ($z/T=0.5$) is tensile (-) due to the bending stress. Even when only the viscosities are different among the layers,

Fig. 3 Stress on top surface

Table 1 Substrate of multilayers

Substrate	l	E/E_0	σ_s/σ_0	$\dot{\epsilon}_s/\dot{\epsilon}_0$
Sb1	2	3	3	1
	3	1	1	1
Sb2	2	3	2.5	0.83
	3	1	1	1
Sb3	2	2	3	1.5
	3	1	1	1

mismatch stress generates and the bending stress also arises with different inclines, as demonstrated in Fig. 2(b).

3.2 Effects of Sintering Properties on Internal Stress

The stress on the surface of the top layer varies with the combinations of the sintering stresses and the viscosities. Consider the replacement of the top layer ($l=1$) when the multilayered substrate is fixed. Three samples of 2-layer substrate are listed in Table 1 with their sintering properties, where $1/E_0$, $1/\sigma_0$ and $1/\dot{\epsilon}_0$ are normalization factors. The bottom layers ($l=3$) in the three substrates are set to be the same. Figure 3 displays the change in the internal stress on the top surface, σ_{xT} , with the viscosity and the sintering stress of the top layer, E_1 and σ_{s1} , respectively. The change in curvature, that is, direction of warping, is also indicated in the same figure.

At $E_1/E_0=2$ for $\sigma_{s1}/\sigma_0=2$ on Sb1, the layers do not warp ($\lambda=0$) and $\sigma_{xT}=0$, because all layers have the same sintering rate. If E_1/E_0 is larger than 2, however, σ_{xT} is tensile due to the bending stress. When E_1/E_0 is smaller, σ_{xT} also becomes tensile because the shrinkage in the top layer is constrained by the substrate. On the other hand, compressive stress regions appear in $0.5 < E_1/E_0 < 2$ for $\sigma_{s1}/\sigma_0=0.5$, and in $2 < E_1/E_0 < 3.5$ for $\sigma_{s1}/\sigma_0=3.5$. In the case of Sb2 and Sb3, stress curves of σ_{xT} are shifted to the direction of plus or minus depending on the intrinsic warpage of the graded substrate itself.

There is a solution of the viscosity, E_c , to give the same σ_{xT} for any value of σ_{s1}/σ_0 , where E_c depends only on the viscosities of the substrate. For three layers with equal thickness, as an

Fig. 4 Diagram of stress

example, solving the equation that the coefficient of σ_{s1} in Eq. (5) equals zero with $z=\pi/2$ gives

$$E_c = \frac{E_2^2 + 14E_2E_3 + E_3^2}{5E_2 + 11E_3} \quad (12)$$

$E_c/E_0=2$ for Sb1 and Sb2, and $E_c/E_0=1.58$ for Sb3 are obtained from Eq. (12). Although the internal stress does not generate at E_c for Sb1, σ_{xT} at E_c is nonzero for Sb2 and Sb3.

3.3 Diagrams of Sintering Stress versus Viscosity

To examine the possibility of the selection or the improvement of the sintering properties of the top layer, a diagram showing the condition to produce tensile stress may be effective. Figure 4 demonstrates the schematic stress diagram for Sb1. The vertical and the horizontal axis of the diagram are the sintering stress and the viscosity of the top layer, respectively, where the slope in the figure is proportional to the free sintering strain rate.

The combinations of the sintering properties to produce no bending appear as a line. Above the line, the layers warp to the side of the top layer, that is, the concave side. Reversely, the top layer becomes convex side in the region under the line. Another boundary, E_c , is also put in the diagrams. The right region from E_c is significantly affected by the bending stress, because the stress distribution becomes steep due to large viscosity. In this case, the region of a tensile stress appears under the line of no bending. Since the constraint by the substrate is predominant in the left region

from E_c , a tensile stress generates when the sintering rate of the top layer is larger than that of the substrate.

Consider controlling the sintering properties by grain size, for example. If it is assumed that the viscosity is proportional to a cubic of grain size d , and the sintering stress varies inversely with the grain size [18], the relationship between E and σ_s is given by

$$\sigma_s = C / E^{1/3} \quad (13)$$

For some values of C , Eq. (13) is plotted in Fig. 4. From the diagrams, the initial grain size to eliminate tensile stress on the surface can be found.

4. APPLICATIONS

Figure 5 is the actual diagram of Sb1, Sb2 and Sb3, respectively. The sintering properties of the layers in the substrate are also interpolated in the diagram for comparison with those of the top layer. When the sintering rates of the layers in the substrate are not uniform, the tension/compression boundaries change. The two tension regions, which appear for Sb1, expand and join in the diagrams for Sb2, but shrink for Sb3. Thus, σ_{xT} at E_c is tensile and compressive for Sb2 and Sb3, respectively.

When the number of the layers is large, the domination by the substrate becomes stronger. Figure 6 (a) shows the stress diagrams for 5-layers with equal thickness ($H_{1-5}=0.2T$). E_c shifts to a large value, and the tension region overwhelms the diagram. To overcome the domination by the substrate, gaining the thickness of the top layer ($H_1=0.4T$) is effective as shown in Fig. 6(b). E_c shifts to the left hand side of the diagrams, and the compressive region is restored.

5. CONCLUDING REMARKS

For the design of multilayers to escape surface cracking on a top brittle layer, effects of the sintering properties and the graded structures on the internal stress were analyzed by using the plate theory. To display the analytical results, diagrams giving the tension/compression regions were presented as a function of sintering stress and viscosity. Selection of the sintering properties for the top layer with no tension on the

Fig. 5 Stress diagrams

Fig. 6 Stress diagrams for 5-layers

surface was indicated for given multilayered substrates. To complete the material design for sintering, distortions of the sintering layers should be finally checked through the whole process by using finite element analysis [13].

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