Camber evolution and stress development during cofiring of dielectric and ferrite bilayer laminates

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Abstract

Camber evolution and stress development of low-temperature cofirable dielectric and ferrite laminates were investigated as a function of the heating rate and the thickness ratio of two layers. Isotropic uniaxial viscosities of individual layers were determined through a vertical sintering technique by in-situ measuring shrinkage curves via an optical dilatometer. Sintering mismatch stresses as a result of the densification rate difference were calculated by using the as-measured curvature of bilayers. A good agreement was achieved in the evolution laws of experimentally measured and theoretically predicted cambers and stresses. However, quantitatively analyzed data indicated obvious differences between experimental results and theoretical predictions, especially where high heating rates and small thickness ratios of dielectric to ferrite layers were adopted. This discrepancy was ascribed to the porosity gradients and preferred pore orientations in the cofired layers as a result of the existing mismatched stresses.
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1. Introduction

The low-temperature co-fired ceramics (LTCC) technology has been used for the integration of passive components and the package of electronic devices and circuits [1–4]. Nowadays, the market for LTCC devices is growing fast due to their excellent electrical and mechanical properties, high reliability and stability as well as the ability of fabricating components with complex 3D structures [5–10]. Recently, LTCC laminates of dielectric and ferrite layers have attracted much attention since applications in filters, inducers and oscillators can be achieved by integrating magnetic functions into the LTCC modules [11–15].

During manufacturing such devices, the co-firing process is one of the most crucial steps. Undesirable co-firing defects, such as cracks, de-bonding and poor densification are usually observed in laminates as a consequence of sintering incompatibility between different materials [16–18]. A few progresses have been made in understanding the cofiring behavior of laminates and the effect of sintering mismatch stress on the camber evolution [1,11,18]. Cai et al. investigated the sintering behavior of an asymmetric laminate consisting of Al_2O_3 and ZrO_2 layers and simultaneously proposed a viscous model [17,19]. Kanters et al. proposed another theory through studying the sintering behavior of thin layers constrained by a thick substrate [20]. Jean et al. studied a series of cofiring cases, concluding factors of affecting the camber evolution and sintering mismatch stress development [2,11]. It has been demonstrated that the camber evolution should be affected not only by intrinsic viscous properties of the laminate layers but also by other factors like geometry and gravity of the laminate, and heating conditions [3,21–24].

In order to study the camber evolution in multilayer laminates, the viscous sintering parameters of individual layer have to be determined. Various experimental methods have been developed to determine the uniaxial viscosity [25–27]. Typically, the sinter forging is one of the most utilized techniques for bulk materials [28–30]. The uniaxial viscosity and viscous Poisson's ratio have been rigorously determined

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for polycrystalline and LTCC materials through a discontinuous sinter-forging process, in which an isotropic microstructure can be maintained during the whole sintering cycle [31–33]. As proposed by Lee et al., the uniaxial viscosity of ceramic layers can be determined by a beam bending method, where an obvious deflection was induced by their own weights [25]. Recently, a vertical sintering technique has been proposed by Cologna et al., through which the uniaxial viscosity of thin layers can be determined by hanging tapes during the sintering process [34]. It has been demonstrated that isotropic microstructure can be maintained in the vertically sintered tapes throughout the sintering process [35].

In this work, bilayer laminates composed of dielectric and ferrite layers were fabricated and different thickness ratios can be achieved by adjusting the number of individual layers. The camber evolution was recorded through a lab-made optical dilatometer and the laminate with a thickness ratio of 1:1 was also studied at different heating rates. Uniaxial viscosities of dielectric and ferrite layers were determined by the vertical sintering technique. According to a continuum sintering theory, theoretical predictions on the camber evolution were conducted and then compared with the experimental results. The sintering mismatch stresses in laminates were predicted through the theoretical model and the microstructure was also characterized. In addition, reasons for the differences between the theoretical and experimental results were discussed in detail.

2. Theoretical background

2.1. Cofiring of bilayer laminates

It is clear that the difference in sintering strain rates between film layers would result in sintering mismatch stress in the laminate. In a bilayer laminate, the layer densifying faster or at lower temperatures will be under a tensile stress, which can be partially relaxed as the laminate bends. In the present case, LTCC film layers in laminates can be considered to be linear and viscous. The normalized curvature rate $\dot{\kappa}$, indicating the camber evolution, can be obtained from the following formula [17,19]:

$$\dot{\kappa} = \frac{d((h_1 + h_2)/r)}{dt} = \frac{6(m+1)^2 mn}{m^4 n^2 + 2mn(2m^2 + 3m + 2) + 1} \cdot \Delta\dot{\epsilon} \quad (1)$$

where h_1 and h_2 are the film thickness of the dielectric and ferrite layer, respectively, r is the radius of curvature, $m = h_1/h_2$ is the thickness ratio, n is the viscosity ratio between layers and $\Delta\dot{\epsilon}$ is the mismatched strain rate between different film layers. By assuming a uniform strain rate within individual films, the mismatched strain rate can be calculated. For the present LTCC film layers, the viscosity ratio could be determined as follows:

$$n = \frac{E_{p1}}{1 - \nu_1} \cdot \frac{1 - \nu_2}{E_{p2}} \quad (2)$$

where E_{p1} and E_{p2} are the uniaxial viscosity of dielectric and

ferrite layers, ν_1 and ν_2 are the viscous Poisson's ratio of dielectric and ferrite layers, respectively. The viscous Poisson's ratio can be roughly predicted as a function of relative density using the following equation [36,37]:

$$\nu = 0.5\sqrt{\rho/(3 - 2\rho)} \quad (3)$$

According to the Cai's model, the sintering mismatch stress development in bilayers can be predicted through the curvature rate, provided the relevant mechanical properties of both layers are known. In current case, the average sintering mismatch stress, which is an average biaxial stress in the dielectric layer, can be calculated using the equation [17,19]:

$$\sigma = \left[\frac{m^4 + mn}{m^4 + 2mn(2m^2 + 3m + 2) + n^2} \right]\dot{\sigma} \quad (4)$$

where $\dot{\sigma}$ is the nominal viscous mismatch stress in the dielectric layer and it can be calculated through the following two formulas:

$$\dot{\sigma} = \left[\frac{m^4 n^2 + 2mn(2m^2 + 3m + 2) + 1}{6(m+1)^2 mn} \right]E_p\dot{\kappa} \quad (5)$$

$$\dot{\sigma} = \frac{E_p \cdot \Delta\dot{\epsilon}}{1 - \nu_p} \quad (6)$$

where $\dot{\kappa}$ is the curvature rate of the bilayer laminate and $\Delta\dot{\epsilon}$ is the mismatched strain rate between two film layers in the laminate, E_p and ν_p are the uniaxial viscosity and viscous Poisson's ratio, respectively.

2.2. Determination of the uniaxial viscosity through a vertical sintering technique

Owing to the extremely large ratio of in-plane dimension to the thickness dimension, most of loading methods for the bulk sample could not be directly adopted in determining the uniaxial viscosity of thick films. Recently, a vertical sintering technique proved to be a valid method for the determination of the uniaxial viscosity. By considering the relationship between the sintering strain rate and the gravity of itself, the uniaxial viscosity of the LTCC film could be determined using the following equation [34]:

$$E_p = \frac{\rho g L}{2(\dot{\epsilon}_v - \dot{\epsilon}_f)} \quad (7)$$

where $\dot{\epsilon}_v$ and $\dot{\epsilon}_f$ are the true strain rates in the vertical direction of vertically and freely sintered LTCC films, respectively, g is the gravity constant, L is the length in the vertical direction, and ρ is the instantaneous density of the sintered film. By assuming an isotropic shrinkage behavior, the relative density of freely sintered sample could be calculated from the true strain data using the following equation:

$$\rho = \rho_0 \cdot exp(-3\varepsilon_f) \quad (8)$$

In order to make the density values reliable for vertically sintered films, the density should be calculated based on the following equation:

$$\rho = \rho_0 \cdot exp(-2\varepsilon_h - \varepsilon_v) \quad (9)$$