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Study of deformation and porosity evolution of low temperature co-fired ceramic for embedded structures fabrication

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Abstract

The deformation behaviors of suspended low temperature co-fired ceramic (LTCC) laminates over a cavity and the evolution of open porosity of LTCC are studied for the fabrication of embedded structures in a multi-layer LTCC platform using carbon material. The effects of the type of LTCC materials (self-constrained and unconstrained LTCC), cavity width, laminate thickness, and lamination conditions on the deformation of the suspended LTCC laminate over a cavity are studied. For suspended three-layers and six-layers LTCC laminates over cavity width ranges from 10 to 25 mm, the self-constrained LTCC laminates were more dimensionally stable (sagged by less than $-120 \,\mu m$) after sintering as compared to the unconstrained LTCC. The evolution of open porosity and the distribution of open pores in the self-constrained LTCC with changes in sintering temperature and laminate thickness are also studied for process optimization.

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1. Introduction

Maintaining the structural dimensional stability in the fabrication of embedded structures in a multi-layer low temperature co-fired ceramic (LTCC) platform remains as a challenge. The suspended LTCC material over the embedded structures is susceptible to deformation during fabrication. During lamination of LTCC green tapes, the suspended green tapes over the embedded structures could deform or sag due to a high lamination pressure (10.3–20.7 MPa). The suspended laminated green tapes (or laminate) could also deform due to the body force induced by the softening of glass component in the LTCC material during sintering. 1–4

To minimize the deformation of suspended LTCC laminate, techniques have been developed to sustain the embedded structures during lamination and/or sintering.^{5–13} Techniques include the employment of temporary inserts,^{5,6,10} fugitive materials (such as carbon material,^{1,2,4,13} waxes¹¹ and polymeric materials^{11,12}), and lead bi-silicate glass² as supporting

medium and using adhesive materials^{7–9} for low pressure lamination. Among these techniques, fabrication of embedded structures using carbon material is one of the most popular techniques. ^{1,2,4,13}

In the fabrication of embedded structures in multi-layer LTCC platform, carbon material is used to support the structures during lamination and/or sintering. The carbon material is required to be completely removed from the embedded structures before the full densification of the LTCC. During sintering, glass component in the LTCC material starts to flow and thus the suspended portion of the embedded structures might deform. The open pores in LTCC material decreases with densification. Simultaneously, carbon material decomposes and diffuses away through the porous LTCC material at this temperature range. As a result, the carbon burnout process tends to compete with the elimination of open pores in LTCC. The intrinsic nature of this process, which involves phase change and deformation of the materials, is complicated. Thus, the understandings of material behaviors, such as carbon burnout, densification of LTCC, pore evolution in LTCC, and deformation of suspended LTCC, are important in optimizing the fabrication process for embedded structures in a multi-layer LTCC platform. Carbon burnout and densification of LTCC material were investigated in our previ-

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ous paper. ¹⁴ To further understand this fabrication process, our current investigation focuses on the deformation of suspended LTCC and open porosity evolution in LTCC during sintering. Hitherto, the deformation behavior of suspended LTCC material over a cavity, especially during sintering, have not been fully understood and investigated. Bau et al. ¹ studied the deformation or sagging of a single layer of unconstrained LTCC tape (DuPont 951-C2) suspended over a cavity. However, the sagging of the suspended LTCC tape was overestimated because the material flow of the tape was not constrained by lamination.

2. Methodology

2.1. LTCC and carbon materials

Commercially available self-constrained LTCC tape (HL 2000, Heraues, Germany), unconstrained LTCC tape (CT 800, Heraues, Germany), and carbon tape (Harmonics Inc., USA) were used. For the subsequent discussions, the HL 2000 and CT 800 tapes are referred as the self-constrained and unconstrained LTCC tapes, respectively. The self-constrained LTCC tape consists of three sub-layers. 15 The top and bottom sub-layers consist of glass ceramic composite and organic binder. The middle sublayer is a refractory ceramic layer that does not shrink during co-firing. Due to the rigidity of this refractory ceramic sub-layer, the shrinkages of the tape are constrained by this refractory ceramic sub-layer in the x- and y-directions. However, due to the porosity of this refractory sub-layer, the glass component in the top and bottom sub-layers melts and infiltrates mostly into this refractory ceramic sub-layer, contributing significantly to the shrinkage in the z-direction. The unconstrained LTCC tape is made of glass ceramic composite particles and organic binder. This material encounters shrinkage in three directions during sintering.

2.2. Deformation of suspended LTCC laminates

2.2.1. Deformation of suspended LTCC laminate during lamination

The deformation behavior of suspended self-constrained LTCC (HL 2000) laminates during lamination but before sintering was studied. Channel of 1 mm in width, 10 mm in length, and 0.125 mm in depth was individually formed on four LTCC tapes. Subsequently, these four tapes were stacked and laminated with another three layers of LTCC tapes to form a LTCC laminate with an open channel of 0.53 mm in depth. The LTCC laminate with open channel were then laminated again with a three-layers LTCC laminate on the open side of the channel to form an embedded channel; see Fig. 1(a). During lamination, the LTCC laminate containing the embedded channel was sandwiched with silicone rubber and glass plates. To study the effect of lamination pressure on the sagging of a suspended LTCC laminate over an embedded channel, three different lamination pressures were explored, namely 0.1, 0.4, and 2.1 MPa. The maximum deformation or deflection of the suspended LTCC laminate after lamination was measured using a stylus pro-

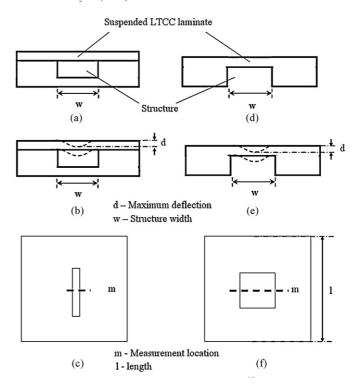


Fig. 1. (a) Cross-section of suspended LTCC laminates over open channel with dimensions of $1.0\,\mathrm{mm}\times10\,\mathrm{mm}\times0.50\,\mathrm{mm}$, (b) maximum deformation of suspended LTCC laminate after lamination, (c) top view of measurement location of maximum deformation of surface profile, (d) cross-section of suspended LTCC laminates over open cavity with depth of 0.53 mm and area of either $10\,\mathrm{mm}\times10\,\mathrm{mm}$, $15\,\mathrm{mm}\times15\,\mathrm{mm}$, $20\,\mathrm{mm}\times20\,\mathrm{mm}$, or $25\,\mathrm{mm}\times25\,\mathrm{mm}$, respectively, (e) maximum deformation of suspended LTCC laminate after sintering, and (f) top view of measurement location of maximum deformation of surface profile.

filometer (Form Talysurf Series 2, Taylor–Hobson); see Fig. 1(b) and (c).

2.2.2. Deformation of suspended LTCC laminate during sintering

Deformation behaviors of two types of suspended LTCC materials during sintering were studied and compared. These materials are self-constrained LTCC (HL 2000) and unconstrained LTCC (CT 800). Suspended LTCC materials over embedded structures could deform after lamination and/or sintering. To identify the contribution to deformation by sintering alone, and the complication of introducing deformation during the lamination stage, a suspended LTCC laminate over an open cavity was used instead; see Fig. 1(d).

Deformation of a suspended LTCC laminate consisting of a single layer of LTCC tape over a cavity was not studied. This is because suspended single layer LTCC was too weak to obtain stable surface profile before sintering for subsequent analysis. Instead, suspended LTCC laminates made of three and six layers of tapes over a cavity width of 10, 15, 20, and 25 mm were studied, respectively.

To fabricate a suspended LTCC laminate, cavity consisting of four layers of LTCC tapes (with tape area of 75 mm \times 75 mm) was obtained by punching each layer individually. These four

-350

-550

LTCC tapes with cavity were laminated with either three or six layers of LTCC tapes to form either a three-layers or six-layers LTCC laminate containing an open cavity with depth of $0.53 \,\mathrm{mm}$. The cavity area is either $10 \,\mathrm{mm} \times 10 \,\mathrm{mm}$, $15 \text{ mm} \times 15 \text{ mm}, 20 \text{ mm} \times 20 \text{ mm}, \text{ or } 25 \text{ mm} \times 25 \text{ mm}.$ The isostatic lamination pressure and temperature were 10.3 MPa and 75 °C, respectively. During lamination, the LTCC laminate (with open cavity facing upwards) was placed on a glass plate and vacuum sealed with three to four layers of plastic bags. These LTCC laminate was then turned upside down to form a suspended LTCC laminate as shown in Fig. 1(d). Surface profile of the suspended LTCC laminate after lamination was measured using a stylus profilometer (Form Talysurf Series 2, Taylor–Hobson).

To further understand the effect of lamination condition on the deformation of the self-constrained LTCC material, suspended three-layers self-constrained LTCC laminate with area of $75 \,\mathrm{mm} \times 10 \,\mathrm{mm}$ and cavity width of $10 \,\mathrm{mm}$ was used. This laminate was similar to the 75 mm \times 75 mm laminate as shown in Fig. 1(e) and (f) except that the length of the laminate was reduced to 10 mm to further simplify the structure. This simplified structure was laminated under the above-mentioned lamination condition and an alternative lamination condition. In the alternative lamination condition, the LTCC laminate was sandwiched between a plastic plate with rubber sheet on top of the open side of the cavity and a glass plate at the bottom of the laminate.

The suspended LTCC laminate was then co-fired in a box furnace. The co-firing profile was 1 °C/min from 30 to 445 °C for organic binder removal from the LTCC laminate and 8 °C/min from 445 to 875 °C and held at 875 °C for 10 min for sintering the LTCC laminate.

To study the resistance to deformation of the suspended LTCC laminates (due to the softening of LTCC material) during sintering, the maximum deflections of the suspended LTCC laminates after sintering were measured again using the stylus profilometer; see Fig. 1(e) and (f).

2.3. Evolution of open porosity in LTCC during sintering

Mercury instrusion porosimeter (Autopore IV 9500, Micromeritics) was used to measure the open porosity. LTCC laminates made of three layers of tapes (with size of $8 \text{ mm} \times 8 \text{ mm}$) were first partially sintered in a box furnace prior to porosity measurement. The LTCC laminates were heated from 30 to 500 °C at 1 °C/min and from 500 °C to a desired sintering temperature at 8 °C/min. The desired sintering temperature was 500, 650, 700, 750, 800, 850, 900, and 950 °C, respectively. The LTCC laminates were subsequently cooled down to room temperature. Open porosity of the LTCC laminates were then measured. Scanning electron microscope (SEM) was used to study the microstructures of the LTCC. Self-constrained six-layers laminates were cross-sectioned and analyzed after sintered at 500, 650, 700, 750, 800, and 850 °C, respectively.

Two cases were used to show the interaction of carbon burnout with self-constrained LTCC based embedded structures. Self-constrained LTCC substrates containing $20 \,\mathrm{mm} \times 20 \,\mathrm{mm} \times 0.20 \,\mathrm{mm}$ embedded cavity were fabricated

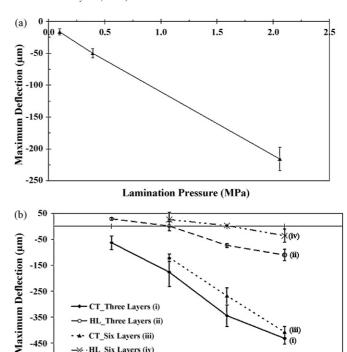


Fig. 2. Maximum deformation of suspended LTCC laminate (a) after lamination at 0.1, 0.4, and 2.1 MPa and (b) after sintering for (i) suspended three-layers unconstrained LTCC laminate, (ii) suspended three-layers selfconstrained LTCC laminate, (iii) suspended six-layers unconstrained LTCC laminate, and (iv) suspended six-layers self-constrained LTCC laminate over cavity widths of 10, 15, 20, and 25 mm after sintering.

Cavity Width (mm)

25

30

using carbon tape as supporting medium. The substrates were co-fired using the normal co-firing profile in the first case. For the second case, the substrates were co-fired with an additional step of holding the heating temperature at 650 °C for 120 min.

3. Results and discussion

3.1. Deformation of suspended LTCC laminate

HL_Three Layers (ii)

CT_Six Layers (iii)

→ ·HL_Six Layers (iv)

10

The maximum deformation was estimated based on the measurement of the surface profile of a suspended LTCC laminate before lamination and after sintering. Depending on the cavity width, the deformation of suspended LTCC laminate could either sag (deform downward as shown in Fig. 1(b) and (e)) or swell (deform upward). In Fig. 2(a) and (b), the sagging and swelling are expressed in negative and positive values, respectively.

3.1.1. Deformation of suspended LTCC laminate during lamination

Even for lamination at a relatively low pressure (10 times lesser than the conventional lamination pressure of 10.3–20.7 MPa), the suspended three-layers self-constrained LTCC laminate sagged in the 1 mm wide channel; see Figs. 2(a) and 3. Since the suspended LTCC laminate was in the green state during lamination, the material underwent plastic

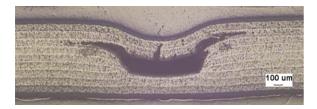


Fig. 3. Cross-section of sagged suspended LTCC laminate over cavity width of 1 mm after lamination at 2.1 MPa.

deformation and flowed into the channel when subjected to the lamination pressure. The magnitude of sagging increased almost linearly from -16 to $-215\,\mu m$ as the lamination pressure increased from 0.1 to 2.1 MPa; see Fig. 2(a). In addition to sagging, crack and delamination were observed on the embedded channel even laminated at these relatively low lamination pressures (see Fig. 3). These results indicate that unless low pressure lamination $^{7-9}$ is employed, supporting medium $^{1-6,10-13}$ has to be used to sustain the embedded structures in a multi-layer LTCC laminate under the conventionally high pressure lamination.

3.1.2. Deformation of suspended LTCC laminate during sintering

The maximum deformations of the surface profiles of suspended LTCC laminates made of three and six layers of self-constrained and unconstrained LTCC tapes over the cavity widths of 10, 15, 20, and 25 mm were measured before and after sintering; see Fig. 1(d)–(f). Before sintering, the surface profiles of the suspended LTCC laminates were reasonably flat. The deformations of the surface profiles of these suspended LTCC laminates after lamination were less than 25 μm over a 10 mm wide cavity and less than 50 μm over a 25 mm wide cavity. The effects of the type of LTCC material, cavity width, thickness of suspended LTCC laminate, and lamination condition on the deformation of the suspended LTCC laminate are discussed as follows.

(1) The effect of type of LTCC material on deformation of suspended laminate.

After sintering, the suspended three-layers selfconstrained LTCC laminates over cavity width ranged from 10 to 25 mm sagged at least three times less than that of the suspended three-layers unconstrained LTCC laminates (comparing curves (ii) and (iv) with (i) and (iii) in Fig. 2(b)). LTCC materials are glass-based materials that experience viscous phase sintering. During viscous phase sintering, the glass component melted into its liquid phase. The liquid phase induces capillary forces between the particles and enhances the densification of the materials.¹⁶ However, the liquid phase could also reduce the rigidity of the solid skeleton consisting of ceramic particles and might cause material collapse under their own body forces. ¹⁷ The sagging of the unconstrained LTCC (CT 800) is most likely due to this viscous deformation of glass component during the viscous phase sintering. In contrast, self-constrained LTCC (HL 2000) consists of three sub-layers. The middle refractory ceramic sub-layer is sandwiched by the top and bottom glass-based sub-layers. ¹⁵ Thus, the relatively low magnitude of sagging for this material (as compared to the unconstrained LTCC) was likely attributed to the middle refractory ceramic sub-layer that possesses relatively high mechanical strength and rigidity.

(2) The effect of cavity width on the deformation of suspended laminate.

After sintering, the three-layers self-constrained and unconstrained suspended LTCC laminates deformed with the maximum sagging increased with an increase in the cavity width; see curves (i) and (ii) in Fig. 2(b). For both selfconstrained and unconstrained LTCC materials, three-layers suspended laminates over a cavity width of 10 mm sagged less than $-70 \,\mu\text{m}$; see curves (i) and (ii) in Fig. 2(b). As cavity width increased from 10 to 25 mm, the suspended self-constrained laminates experienced less deformation as compared to the unconstrained LTCC. The maximum deformations of the suspended three-layers self-constrained LTCC laminates increased by more than three times from $30 \text{ to } -110 \,\mu\text{m}$ as cavity width increased from 10 to 25 mm. In contrast, the maximum deformations of suspended threelayers unconstrained LTCC laminates increased by seven times (from -65 to $-430 \mu m$) as the cavity width increased from 10 to 25 mm. As cavity width increased, the volume of suspended LTCC laminate increased, and thus the volume of suspended glass component increased. Higher body force was induced due to this relatively higher volume of suspended glass component during sintering. As a result, sagging of the suspended LTCC laminate increased as this induced body force increased.

(3) The effect of thickness of suspended laminate on its deformation.

To study the effect of the number of tape layers on sagging during sintering, suspended six-layers LTCC laminates over cavity width of 15, 20, 25 mm were also studied; see curves (iii) and (iv) in Fig. 2(b). The suspended six-layers selfconstrained and unconstrained LTCC laminates experienced less deformation than that of the three layers; comparing curve (ii) with (iv) and (i) with (iii) in Fig. 2(b). Overall, for both self-constrained and unconstrained LTCCs, the ratio of maximum deformations between the suspended threelayers and six-layers LTCC laminates ranged from 1 to 3 times. This result indicates that for suspended LTCC laminate consisting of more number of layers, the more rigid skeleton made of ceramic particles will withstand better the body forces induced from the viscous flow of glass component during sintering. As a result, the deformations of thicker suspended LTCC laminates were reduced after sintering.

(4) The effect of lamination condition on deformation of suspended laminate.

As shown in Fig. 4, the three-layers unconstrained LTCC laminate sagged with increased magnitude as the cavity width increased from 10 to 25 mm. Swelling was solely observed on the suspended self-constrained LTCC laminate (see Fig. 5). The suspended three-layers self-constrained LTCC laminate over a 10 mm wide cavity swelled with

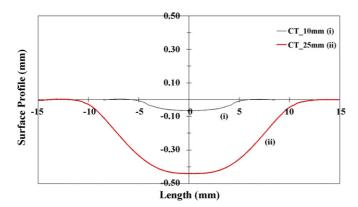


Fig. 4. Surface profile of suspended three-layers unconstrained LTCC laminate over cavity of 10 mm in width and 25 mm in width after sintering over and above the initial deformation of the surface profile.

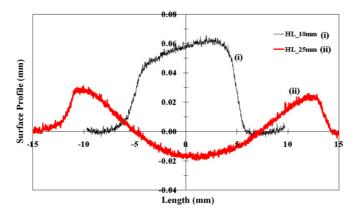


Fig. 5. Surface profile of suspended three-layers self-constrained LTCC laminate over cavity of 10 mm in width and 25 mm in width after sintering over and above the initial deformation of the surface profile.

average magnitude of less than 30 µm; see curve (ii) in Fig. 2. Further investigation was conducted to understand the swelling observed on the suspended self-constrained LTCC laminate. As described in Section 2.2.2, suspended

three-layers self-constrained LTCC laminates with area of $75 \,\mathrm{mm} \times 10 \,\mathrm{mm}$ and cavity width of $10 \,\mathrm{mm}$ were laminated under two different lamination conditions separately. Before vacuum sealed with plastic bags, these laminates were either (i) placed on a glass plate with open cavity facing upwards or (ii) sandwiched between a plastic plate with rubber sheet on top of the open side of the cavity and a glass plate at the bottom of the laminate. The trend of swelling (similar to the trend shown in Fig. 5(a)) was observed for the suspended three-layers self-constrained laminate that underwent lamination condition (i). The swelling of the suspended laminate ranged from 48 to 62 µm. However, the swelling of the suspended three-layers self-constrained laminate was significantly minimized after laminated by employing lamination condition (ii). The maximum deflection of the suspended laminate was measured to be -8 to 3 µm. These observations imply that the swelling of the suspended self-constrained LTCC laminate was most likely due to the non-uniformity of lamination stress (especially at the corners of the open cavity) during lamination. Using plastic plate with rubber sheet in lamination condition (ii) could have induced more uniform lamination stress over the open cavity on the laminates as compared to solely using few layers of plastic bags in lamination condition (i). As swelling did not occur on the suspended unconstrained LTCC laminates (as shown in Fig. 4(a) and (b)), it could be deduced that the refractory ceramic layer in the self-constrained material was sensitive to the lamination condition.

Overall, our experimental results indicated that the sagging due to sintering is relatively less critical as compared to the sagging due to the high lamination pressure for the fabrication of relatively small-embedded structures (with dimensions of less than 10 mm) for both self-constrained and unconstrained LTCCs. This also implies that maintaining supporting medium, such as carbon,² for the embedded structure during sintering is not critical for the fabrication of relatively small (less than

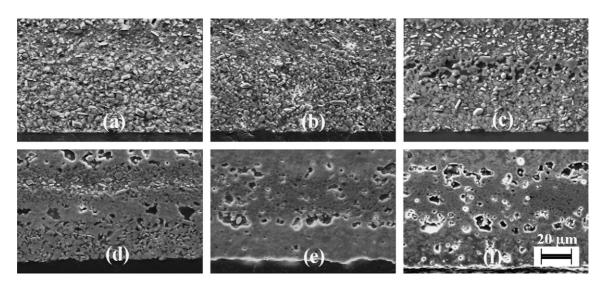


Fig. 6. Microstructure evolution of self-constrained LTCC laminate (SEM-SE micrographs at $1000\times$) with maximum heating temperature at (a) $500\,^{\circ}$ C, (b) $650\,^{\circ}$ C, (c) $700\,^{\circ}$ C, (d) $750\,^{\circ}$ C, (e) $800\,^{\circ}$ C and (f) $850\,^{\circ}$ C.

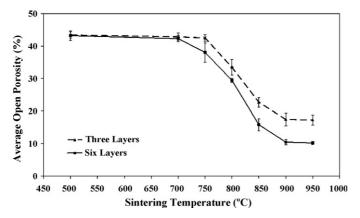


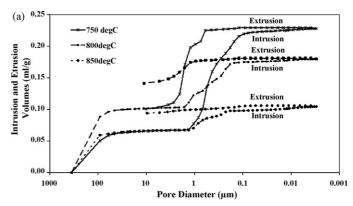
Fig. 7. Change of average open porosity versus sintering temperatures (at 500, 700, 750, 800, 850 and 900 $^{\circ}$ C) for three-layers and six-layers composite ceramic laminates.

 $10\,\text{mm})$ embedded structures. For fabrication of relatively large (ranges from 15 to 25 mm) embedded structures, supporting medium is required during sintering for structures made of unconstrained LTCC material since suspended unconstrained three-layers and six-layers laminates sagged with magnitudes of -100 to $-430~\mu\text{m}$ after sintering. In contrast, self-constrained LTCC material is more dimensionally stable during sintering. The observed saggings after sintering for suspended three-layers and six-layers self-constrained laminates were less than $-120~\mu\text{m}$. These results indicated that supporting medium is not necessary during sintering for the fabrication of relatively large embedded structures (with width ranges 15–25 mm) made of self-constrained LTCC material. The fabrication of large embedded cavity is practically useful in realizing embedded air cavity for stacked patch antennas 18,19 and pressure sensor. 13

3.2. Evolution of open porosity of self-constrained LTCC during sintering

Figs. 6 and 7 show the evolution of microstructure and average open porosity of self-constrained LTCC material during sintering. Fig. 8 shows the cumulative intrusion and extrusion volumes of three-layers and six-layers self-constrained LTCC laminates. During porosity measurement, mercury was pressurized and intruded into the open pores of a sample. As such, the open pore volume was obtained based on the intrusion volume of the mercury according to Washburn equation. The extrusion volume was measured when the mercury was extracted out from the sample. Based on the measured intrusion volume, the pore size distribution of the LTCC material was obtained in Fig. 9.

As shown in Fig. 6(a) and (b), the evolution of microstructure was hardly observed for self-constrained LTCC sintered at 500 °C and 650 °C. The composite glass ceramic powders remained as a porous skeleton after debinding and before sintering. During this temperature range (before 700 °C), the average open porosity in the self-constrained LTCC laminates did not evolve significantly. The open porosities of the self-constrained three-layers and six-layers LTCC laminates, which sintered at 500 and 700 °C, were 42–43 % (see Fig. 7).



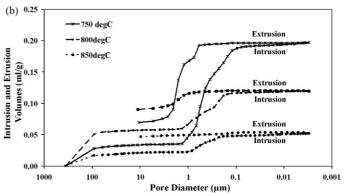
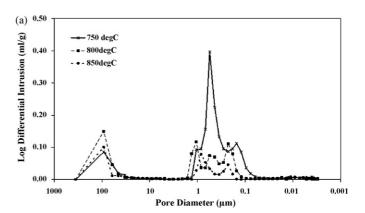


Fig. 8. Cumulative intrusion and extrusion volumes versus pore diameter for (a) three-layers LTCC laminates and (b) six-layers LTCC laminates sintered at 750, 800, and $850\,^{\circ}$ C.



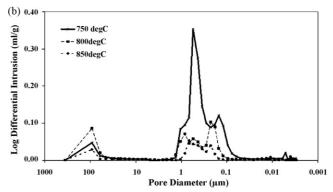


Fig. 9. Logarithmic volume distribution function versus pore diameter for (a) three-layers LTCC laminates and (b) six-layers LTCC laminates sintered at 750, 800, and $850\,^{\circ}$ C.

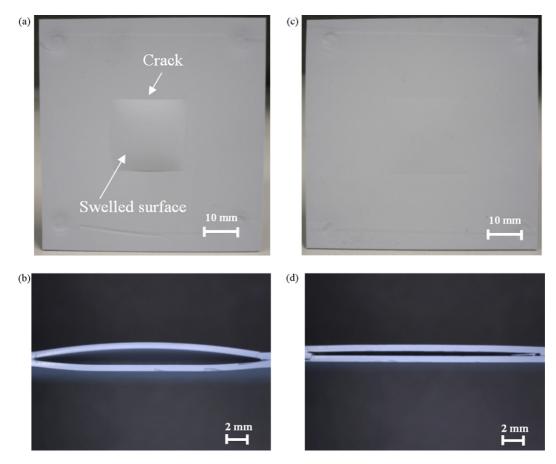


Fig. 10. Top and cross-section views on embedded cavity fabricated with carbon tape using (a and b) standard co-firing profile and (c and d) co-firing profile with additional isothermal heating.

Obvious densification was observed for self-constrained LTCC material sintered at above 700 °C. The glass component from the glass ceramic composite sub-layers melted and infiltrated into the refractory ceramic sub-layers in the self-constrained LTCC material (see Fig. 6(c)–(f)). The open pores in the LTCC material began to be eliminated as the material densified. Open porosity of the three-layers and six-layers LTCC laminates decreased to 17 and 10 %, respectively, after sintered at 900 °C (see Fig. 7). The evolution of open porosity reached a plateau after 900 °C.

Overall, the open pore volumes decreased with an increase in thickness of laminate (see Fig. 7). For those LTCC laminates sintered at 750–900 °C, the open porosity in three-layers LTCC laminates were 10–40 % higher than that of in six-layers LTCC laminates (see Fig. 7). This phenomenon was probably due to the increased frequency of overlapping of open pores with glass ceramic composite material (which created more close pores) as the thickness of laminate increased.

The characteristic of open porosity (i.e. the pore structure, the pore size distribution, etc.) in the self-constrained LTCC laminate did not change with an increase in thickness of laminate (see Figs. 8 and 9). Hysteresis was observed in which not all the mercury was expelled or extruded from the sample during testing (see Fig. 8(a) and (b)). The entrapment of

the mercury might be due to the possible existent of ink-bottle pores with narrow neck²¹ or porosity network²² in the self-constrained LTCC laminates. These results qualitatively indicate that the self-constrained LTCC material was tortuous during sintering.

The pore size distribution curve indicates that the open pores in the LTCC laminates distributed within two distinct categories during densification (see Fig. 9(a) and (b)). The diameters of relatively large pores ranged from 30 to 300 μm . For the relatively small pores, their diameter ranged from 0.1 to 1.0 μm . As the LTCC laminates were sintered at 750–850 °C, relatively large volume of pores with diameter of 0.6 μm decreased. Comparison of Fig. 6 with Fig. 9(b) indicates that the relatively large pores could be formed between the interface of the glass ceramic composite sub-layer and the refractory ceramic sub-layer or on the outer surface of the laminate.

Two cases were studied to observe the effect of heating profile on the carbon burnout for the fabrication of embedded structure in a multi-layer LTCC platform. LTCC substrate containing $20 \text{ mm} \times 20 \text{ mm} \times 0.2 \text{ mm}$ embedded cavity was fabricated using carbon tape as supporting medium. The substrate was co-fired using standard heating profile and modified heating profile for the first and second case respectively. Cracks and severe swelling were observed on the substrate for the first case (see Fig. 10(a)). The swelling of the suspended LTCC

laminate could be as high as 900 μm . For the second case, the maximum swelling for the suspended LTCC laminate was reduced to 110 μm and no crack was observed (see Fig. 10(b)). The swelling or deformation of the suspended LTCC laminate was mainly attributed to the pressure buildup of the decomposed carbon in the embedded cavity. These observations imply that process optimization, especially the control of heating profile for carbon burnout, is still critical even when the existence of open pores was observed for the fully sintered self-constrained LTCC material.

The results on the deformation of suspended self-constrained LTCC laminate over an embedded structure (as discussed in Section 3.1) imply that the increase in thickness of the suspended self-constrained LTCC laminate could provide a relatively more stable structure during sintering. However, it could decrease the open porosity in the self-constrained LTCC laminate as well, which in turn increase the resistance for the diffusion of decomposed carbon. These factors (i.e. the stability or deformation of the suspended LTCC laminate and the evolution of open porosity) are important parameters to be considered in modifying the co-firing profile in order to allow the completion of carbon burnout without inducing defect on the embedded structures in a multi-layer LTCC platform.

4. Conclusions

The deformation behavior of suspended self-constrained and unconstrained LTCC laminates during lamination and sintering and the evolution of open porosity in self-constrained LTCC, which are critical for embedded structure fabrication, were investigated. The results indicate that the deformation due to sintering is relatively less critical as compared to the deformation caused by high lamination pressure for the fabrication of relatively small embedded structures (with dimensions of less than 10 mm) for both self-constrained and unconstrained LTCCs. Therefore, maintaining carbon material to support the structure during sintering is not critical for these relatively small structures. The suspended three-layers self-constrained LTCC laminates over cavity width ranged from 10 to 25 mm sagged (with magnitude of less than $-120 \,\mu\text{m}$) at least three times less than that of the suspended three-layers unconstrained LTCC laminates. This result implies that the self-constrained LTCC material is more feasible for the fabrication of embedded structures of relatively large dimensions (ranges from 15 to 25 mm). The deformation of the suspended self-constrained LTCC laminates during sintering could be reduced with thicker suspended laminates. However, the decrease in open porosity for carbon burnout and diffusion with an increase in laminate thickness has to be considered. The understandings of these material behaviors provide a guideline for optimizing the fabrication of embedded structures in multi-layer LTCC platform using carbon material.

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References

- Bau, H., Ananthasuresh, S., Santiago-Aviles, J. J., Zhong, J., Kim, M., Yi, M. et al., Ceramic tape based meso systems technology. In Proceedings of the ASME International Mechanical Engineering Congress and Exposition, 1988, pp. 491–498.
- Epsinoza-Vallejos, P., Zhong, J.-H., Gongora-Rubio, M. R., Sola-laguna, L. and Santiago-Aviles, J. J., Meso(intermediate)-scale electromechanical systems for the measurement and control of sagging in LTCC structures. In *Materials Research Society Symposium Proceedings*, vol. 518, 1998, pp. 3–79.
- Gongora-Rubio, M. R., Espinoza-Vallejos, P., Sola-Laguna, L. and Santiago-Aviles, J. J., Overview of low temperature co-fired ceramics tape technology for meso-system technology (MsST). Sensors and Actuators A, 2001, 89, 222–241.
- Birol, H., Maeder, T. and Ryser, P., Processing of graphite-based sacrificial layer for microfabrication of low temperature co-fired ceramics (LTCC). Sensors and Actuators A, 2006, 130–131, 560–567.
- Bauer, R., Luniak, M., Rebenklau, L., Wolter, K.-J. and Sauer, W., Realization of LTCC-multilayer with special cavity applications. In *Proceedings of the 30th International Symposium on Microelectronics, ISHM'97*, 1997, pp. 659–664.
- Miehls, D. J., Martin, F. J., Pond, R. G. and Fleischner, P. S., Method of fabricating a multilayer electrical circuit. US Patent No. 5,249,355, October 5, 1993.
- Roosen, A., New lamination technique to join ceramic green tapes for the manufacturing of multilayer devices. *Journal of the European Ceramic Society*, 2001, 21, 1993–1996.
- Jones, W. K., Liu, Y. and Gao, M., Micro heat pipes in low temperature cofired ceramic (LTCC) substrates. *IEEE Transactions on Components and Packaging Technologies*, 2003, 26(March (1)), 110–115.
- Burdon, J. W., Huang, R.-F., Wilcox, D. and Naclerio, N. J., Method for fabricating a multilayered structure and the structures formed by the method. US Patent No. 6,592,696, July 15, 2003.
- Cawley, J. D., Heuer, A. H. and Newman, W. S., Method of constructing three dimensional bodies from lamination. US Patent No. 5,777,833, July 14, 1998.
- Trickett, E. A. and Assmus, R. C., Ceramic monolithic structure having an internal cavity contained therein and a method of preparing the same. US Patent No. 4,806,295, 1989.
- Alexander, J. H., Method of making ceramic article with cavity using LTCC tape, US Patent No. 5,601,673, 1997.
- Peterson, K. A., Patel, K. D., Ho, C. K., Rohde, S. B., Nordquist, C. D., Walker, C. A. et al., Novel microsystem applications with new techniques in low-temperature co-fired ceramics. *International Journal of Applied Ceramic Technology*, 2005, 2, 345–363.
- Khoong, L. E., Tan, Y. M. and Lam, Y. C., Carbon burnout and densification of self-constrained LTCC for fabrication of embedded structures in a multi-layer platform. *Journal of the European Ceramic Society*, 2009, 29, 457–463.
- Rabe, T., Schiller, W. A., Hochheimer, T., Modes, C. and Kipka, A., Zero shrinkage of LTCC by self-constrained sintering. *International Journal of Applied Ceramic Technology*, 2005, 2, 374

 –382.
- Mohanram, A., Messing, G. L. and Green, D. J., Densification and sintering viscosity of low temperature co-fired ceramics. *Journal of the American Ceramic Society*, 2005, 88, 2681–2689.
- Liu, J., Lal, A. and German, R. M., Densification and shape retention in supersolidus liquid phase sintering. *Acta Materialia*, 1999, 47, 4615–4626.
- Panther, A., Petosa, A., Stubbs, M. G. and Kautio, K., A wideband array of stacked patch antennas using embedded air cavities in LTCC. IEEE Microwave and Wireless Components Letters, 2005, 15, 916–918.

- 19. Komulainen, M., Mahonen, J., Tick, T., Berg, M., Jantunen, H., Henry, M. *et al.*, Embedded air cavity backed microstrip antenna on an LTCC substrate. *Journal of the European Ceramic Society*, 2007, **27**, 2881–2885.
- 20. Lowell, S., Shields, J. E., Thomas, M. A. and Thommes, M., *Characterization of Porous Solids and Powders: Surface Area, Pore Size and Density*. Kluwer Academic Publishers, 2004.
- Moscou, L. and Lub, S., Practical use of mercury porosimetry in the study of porous solids. *Powder Technology*, 1981, 29, 45–52.
- Paik, U., Kang, K.-M., Jung, Y.-G. and Kim, J., Binder removal and microstructure with burnout conditions in BaTiO₃ based Ni-MLCCs. Ceramics International, 2003, 29, 939–946.