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# Low-pressure, thermo-compressive lamination

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## Abstract

Lamination of green ceramic tapes is one of the most important technological processes in multilayer ceramic technology. Lamination affects the quality of all 3D structures (e.g., channels, chambers, membranes, etc.). Novel chemical methods of lamination reduce the deformation of 3D structures. However, these methods are useless in the fabrication of thin membranes and structures with thick-film electronic components or electric vias. Therefore, thermo-compressive lamination is still the best solution for the lamination of green ceramic tapes. Low-pressure thermo-compressive lamination with an insert material is presented in this paper. The influence of pressure and Low Temperature Cofired Ceramics (LTCC) material on the compressibility and shrinkage of LTCC, as well as the influence of the insert material on deflection and distortion of the membranes are presented and discussed in this paper.

*Keywords:* Lamination; Low pressure; LTCC; Joining; Bonding; Green tape; Insert; Ceramics; Low temperature; Thermo; Compressive

## 1. Introduction

Low-Temperature Cofired Ceramics (LTCC) technology has been developed over the past few decades.[1–3] The technique was first used in the fabrication of multilayer electronic substrates and passive components.[4] Since the early 1990s, LTCC has been used in the fabrication of sensors, actuators and microsystems.[5,6]

Tape machining of LTCC tapes,[7,8] thick-film deposition,[9] lamination (tape bonding in the green state)[10] and the sintering profile affect the parameters of sensors, actuators and microsystems. Therefore, all these processes must be very carefully optimized. The quality of 3D structures depends primarily on the lamination and on firing. The most popular lamination method is thermo-compression. The tapes are joined at elevated temperatures (70–90 °C)[11] using high pressure (5–20 MPa)[11] for a certain amount of time (3–10 min).[12] The lamination of green tapes is schematically presented in Fig. 1. Single tapes presented in Fig. 1a consist of ceramic particles, glass and organic material (mainly binder). Tapes during lamination are heated and pressed together. Hence, the organic material becomes plastic, and the tapes can be joined. Moreover, the applied pressure forces the ceramic particles and glass from all the tapes together. Therefore, if the pressure is high the lamination strength between the tapes is strong. During firing, the organic material is driven off, and the glass melts, wets and joins the ceramic particles together.

The final bonding strength between LTCC ceramic tapes depends on three main mechanisms[13]:

- joining of the melted resinous constituents (occurs during lamination),
- the mechanical joining of rough surface (occurs during lamination),
- viscous flow of the glass material (occurs during firing).

The simplest model of green LTCC is that of an elastic–plastic material. Therefore, good lamination can occur only if the lamination pressure is higher than the yield point of the organic material. Moreover, the yield point decreases if the temperature increases, and therefore, lower lamination pressures can be used if the laminated stack is heated more. A more complicated model should also include the influence of the organic material, the ceramic particles and the glass material contents. The organic material should enable proper mechanical properties of the green tape. However if there is too much organic material, the ceramic particles are too far apart, and there is no possibility for proper firing of such a structure.[14]

The organic material is driven off during the cofiring process. The melting of the glass plays a dominant role in the

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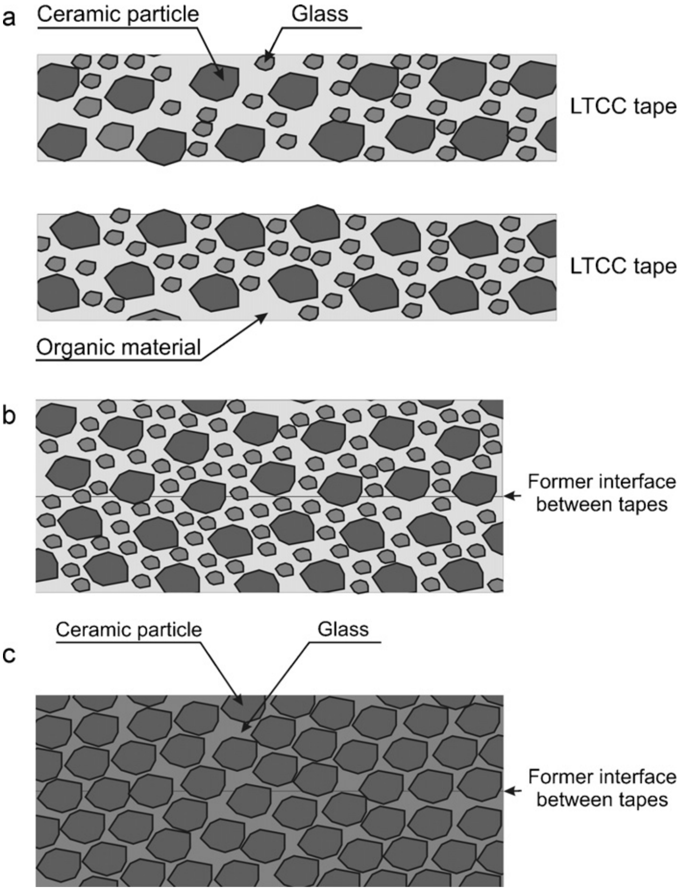

Fig. 1. Bonding of LTCC tapes: (a) before lamination, (b) after lamination, and (c) after firing.

viscous-flow mechanism among the constituents, and hence the glassy material used in LTCC tapes is very important for proper bonding and sintering of glass/ceramic composites.[13] The glass melts above the softening temperature and penetrates the void space between the ceramic particles. The viscous flow of the glass depends on the ceramic-powder particle size, glass and ceramic particle content, glass softening point and glass melting point.[13] The melted glass wets ceramic particles and enables to achieve strong final structure. Therefore, the glass material enables a lowering of the LTCC firing temperature below 900 °C. Structure warping, distortion and other deformations occur during sintering and are associated with viscous flow.[13] Therefore, in many cases, the structure after sintering is more deformed than after lamination. The deformation can also be multiplied by the shrinkage of the LTCC during firing. The penetration of the glass material between the ceramic glass can also bond tapes together. If during firing the tapes are joined together, then viscous flow occurs not only in a single tape but also between tapes. Therefore, ceramic particles from laminated tapes can be permanently bonded. Thermo-compression does not affect the properties of thick, passive films and enables the fabrication of structures that have more than 40 layers. However, high pressure and temperature can very easily damage thin membranes, wide channels and large chambers.

Therefore, chemical lamination, lamination with fugitive phases and lamination with inserts were proposed.[10] The first chemical bonding was reported by Roosen.[12,15–18] The tapes are joined with double-sided adhesive tape, which ensures temporary adhesion between the tapes before sintering. The tapes are permanently joined during a cofiring process. The method enables a reduction in the deformation of 3D structures and does not dissolve the tape surfaces. However, the ceramics should have relatively high porosity to ensure sufficiently high forces, which join the tapes during the firing. Moreover, structures with a higher number of tapes tend to crack. The method enables bonding of metallized tapes. However, the influence of the method on the reliability of thick–film components and electrical vias has not been reported. Upgraded versions of the deposition of a joining medium onto the tapes have been reported.[17,18] Other chemical methods, which have also been reported, are very similar. Temporary bonding between the tapes is achieved by applying some adhesive agent onto the tapes.[19–22] Then, the tapes are permanently joined during cofiring. The tapes can also be joined using solvents.[23–25] A diluted fluid applied onto the green tape dissolves both the joined surfaces and bonds the tapes together. However, this method affects the parameters and reliability of electrical vias and thick-film components.[25] The adhesive-based chemical methods might also affect the properties of vias and thick-film components. Therefore, alternative lamination methods have been developed. Thermo-compressive lamination with a fugitive phase is the best solution for the fabrication of closed chambers and channels. In this method, the tapes are bonded at high pressure and at elevated temperature, but the chambers and the channels are filled with a sacrificial material,[26–29] which protects the 3D structures from damage during lamination. The fugitive phase is intended to disappear during the sintering process. The sacrificial material can also protect the structure from deformation during the debinding process. However, the firing process must be extended to ensure proper burning out of the sacrificial material. The alternative is to replace the sacrificial material by an insert material.[10] Open 3D structures are filled with the insert material, which protects the structures from deformation during lamination. After the process, the insert is removed. Moreover, the insert is not present during firing, and therefore, a standard cofiring profile can be used.

Isotactic, thermo-compressive lamination with inserts under very low pressures has not been widely discussed in the literature. Therefore, the influence of the lamination pressure in the range from 0.5 to 20 MPa and of the LTCC material on the bonding quality, LTCC tape compressibility and shrinkage of LTCC were analyzed. Moreover, the relative membrane deflections for various membrane dimensions and tape types were investigated. The membrane deflections of the structures laminated with and without the inserts were compared and discussed. The authors wish to show that viscous flow of glass during firing can be a dominant joining mechanism and can ensure good bonding quality.

## 2. Experiment

The experiment was carried out on four different commercially available LTCC tapes: HL800, HL2000, DP951 and

Table 1
The cofiring profiles used in the experiment.

| DP951 | Ceram Tape GC | HL2000 | HL800 |
|---|---|---|---|
| Ramp at 7.6 °C/min to 480 °C<br>Debinding at 480 °C for 60 min | Ramp at 2 °C/min to 180 °C<br>Debinding ramp at 0.6 °C/min to 450 °C<br>Debinding at 450 °C for 120 min | Ramp at 3 °C/min to 100 °C<br>Debinding ramp at 1 °C/min to 450 °C | Ramp at 6 °C/min to 300 °C<br>Debinding ramp at 2.5 °C/min to 470 °C |
| Ramp at 6.7 °C/min to 880 °C<br>Firing peak at 880 °C for 15 min | Ramp at 6 °C/min to 900 °C<br>Firing peak at 900 °C for 30 min | Ramp at 10 °C/min to 865 °C<br>Firing peak at 865 °C for 30 min | Ramp at 3 °C/min to 880 °C<br>Firing peak at 880 °C for 45 min |

Ceram Tape GC. The experiment consisted of four main parts (stages). In the first stage of the experiment, the influence of the thermo-compressive lamination pressures (0.5, 1, 2, 5, 10, 15, 20 MPa) on the lamination quality was investigated. The test structure used at this stage consisted of four, LTCC, square-sheet pieces each with dimensions of 25 mm × 25 mm. The shapes were cut using a hot knife. The tapes were stacked and laminated in an isotactic chamber using various pressures at 70 °C for 10 min. Then, the tapes were cofired at standard cofiring profiles recommended by the LTCC manufacturers. The cofiring profiles used in the experiment are presented in Table 1. The sintered structures were cut using a Nd-YAG laser, and the bonding quality was analyzed by scanning electron microscopy (SEM). This investigation enabled determination of the lowest pressure that can be used to join tapes together and achieve good bonding without delamination.

The test structure in the second stage of the experiment consisted of four, square membranes with various dimensions (with square sides of 2, 5, 7 and 10 mm after sintering). The tapes were cut using a Nd-YAG laser. The total thickness of the structures was approximately 1 mm in the green state. The test structures were laminated in an isostatic chamber using the optimal pressure (i.e., the lowest pressure that achieves good bonding quality, which was found for each tape in the previous experimental stage).

The influence of the lamination and firing processes on displacement and distortion of the membrane is schematically presented in Fig. 2. The tapes before isostatic lamination were not deformed (Fig. 2a, where $h_0$ is the distance between the bottom of the structure and membrane before lamination). Deformation that occurs during lamination (Fig. 2b, where $\Delta h_1$ is the change in dimension between the bottom of the structure and the membrane upon lamination, $\Delta x_1$ is the change in chamber dimensions in the x and y directions, and $p$ is the lamination pressure) are caused by two phenomena: the applied pressure in the membrane surface area and the plastic behavior of tapes. The first problem is especially severe when closed cavities or channels are fabricated. If the pressures applied on both sides of the membrane are not in equilibrium, the displacement of the membrane is directed from an area of higher pressure to one of lower pressure. Fabrication of open, 3D-multilayer structures is easier because in this case pressure equilibrium during lamination is always achieved. Additional covers for laminating the LTCC stack can solve the problem. The covers should exhibit a high Young's modulus as well as high flexural and compressive strengths. If such a material is added as top and bottom covers of the stack, these covers will protect the surface of the membrane from the applied pressure. The tape will only be compressed around the membrane. Such a solution is sometimes called semi-isostatic lamination because applied pressure on the top and bottom parts of the stack is applied as in uniaxial compression and along the $x$ and $y$ axes as in normal isostatic pressing. The method is also used to align single tapes together during lamination. The plastic behavior of the tapes, which is the second cause of deformation during lamination, is more complicated. Semi-isostatic lamination cannot inhibit the deformation of buried, 3D-multilayer structures. For such applications, it is necessary to use chemical bonding or thermo-compressive lamination with fugitive phases.[26–29] The deformation of open structures caused by the second phenomenon can be reduced by only using hard covers on the membrane side. However, isostatic lamination is carried out either in water or in oil, and hence the laminated stack must be closed in a vaporized bag to protect the laminates from the fluid. Such bags typically deform the edges of open 3D structures when pressure is applied. Therefore, on the one hand, such structures also can be laminated using chemical methods or thermo-compressive lamination with fugitive phases. On the other hand, a rubber-like insert material can be used to protect the 3D structure from the deformation that is caused by pressure. The insert material should exhibit highly elastic behavior, i.e., a rubber-like material (very low Young's moduli), and moreover, the insert material cannot exhibit plastic behavior. All these methods would protect the edges of 3D structures from deformation. The total deformation of the membrane caused by lamination is presented in Fig. 2c (where $h_1$ is the distance between the bottom of the structure and the membrane after lamination). The next distortion of the membrane occurs during firing. The direction and type of membrane deformation are not so easy to predict as in lamination. The membrane can bend or become wavy after firing. Therefore, the deformation presented in Fig. 2d is just one of the possibilities (where $\Delta h_2$ is the dimensional change between the bottom of the structure and the membrane that occurred during firing, $\Delta x_2$ is the change in the chamber dimension in the $x$ and $y$ directions during firing). The problem is caused by viscous flow of the glass during firing. Theoretically, there are two solutions to the problem: using fugitive phases or performing a second firing cycle with loading. The first solution can significantly improve the geometry of the structure. However, the fugitive phase must be completely driven off from the stack during firing. Hence, a significantly longer firing profile must be used (even three times longer than the standard one). The second solution gives good results for most simple

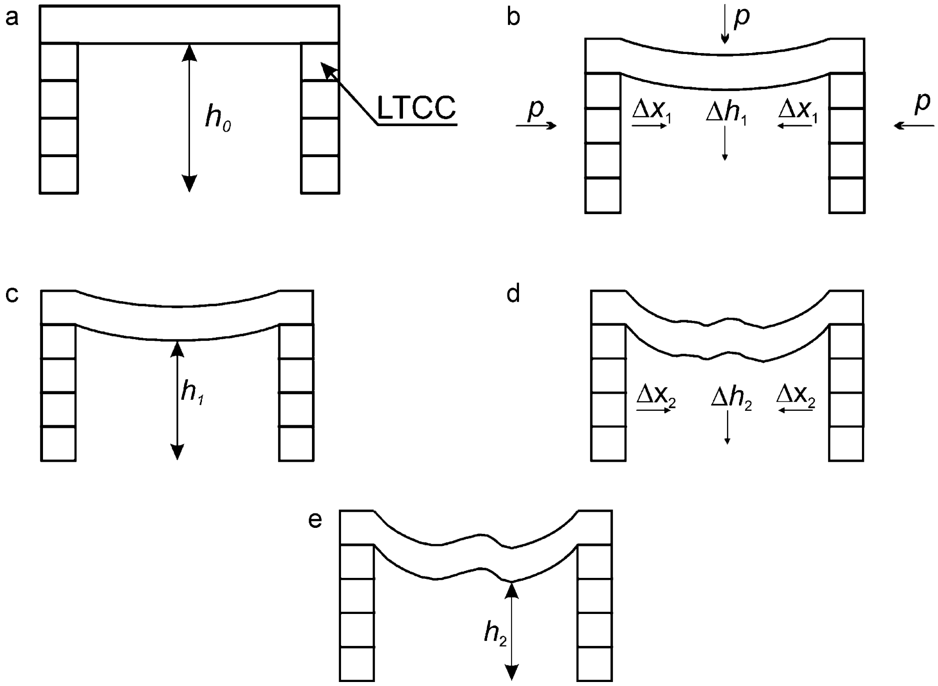

Fig. 2. Schematic diagram of deflection, structure: (a) before lamination, (b) during lamination, (c) after lamination, (d) during firing, and (e) after firing.

3D structures. The samples after firing are once more fired at a slight lower temperature of approximately 50 °C and are loaded using a piece of alumina, for example. If the glass softening temperature is reached, the sample became softer and warping can be reduced. However, in many LTCC materials in which a crystallized glass is used after firing, little glass residue is left. Hence, such a glass does not flow when it is fired again.[13] The total distortion of the membrane is presented in Fig. 2e (where $h_2$ is the distance between the bottom of the structure and the membrane after firing).

Each structure was laminated with a rubber-like die insert material. The lamination with the insert is schematically presented in Fig. 3. The LTCC structure was covered by a PCB FR-4[30] (Young's modulus of 24 GPa, Poisson's ratio of 0.3 and density of 1850 kg/m$^3$ [30]) from the top and the bottom, and the machined cavity was filled with the insert material, PDMS[31] (Young's modulus of 360–870 kPa, Poisson's ratio of 0.5 and density of 0.97 kg/m$^3$). The insert material protects the 3D structure from deformation during thermo-compressive lamination. After the bonding, the insert was removed, and the structure was cofired with the standard cofiring profile (the same as presented in Table 1). The sintered structures were cut using a Nd-YAG laser, and the lamination quality and membrane deflections (only the smallest membranes) were analyzed in structure cross-sections by SEM.

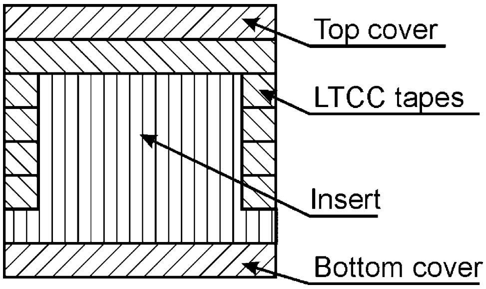

Fig. 3. Schematic cross-section of lamination using insert.

The influence of the lamination pressure and LTCC material on the LTCC compressibility and shrinkage were investigated in the third part of the experiment. The average relative compressibility and average shrinkage were calculated based on measurements of 12 samples. The test structures were the same as in the first stage of the experiment. However, the influence of only four different pressures (0.5 or 2 MPa; 6 MPa; 13.5 MPa and 20 MPa) on the lamination quality was investigated, and moreover, the fired modules were not laser-cut at all. The relative compressibility in the green state was calculated using Eq. (1) (where $z_{\mathrm{ref}}$ is the thickness of the test structure laminated at 20 MPa and $z$ is the thickness of the test structure laminated at various pressures). Therefore, if the relative compressibility is higher than 100%, the compression is smaller in comparison to the relative value. The values of shrinkage along the $x$ and $y$ axes and along the $z$ axis were calculated using Eqs. (2) and (3), respectively, (where $z_0$ is the thickness of the test structure after lamination, $z_1$ is the thickness of the test structure after firing, $x_0$ are the dimensions of the test structure along the $x$ and $y$ axes after lamination, $x_1$ is the dimension of the test structure along the $x$ and $y$ axes after firing).

$$\beta_{rel} = \frac{z}{z_{\mathrm{ref}}} 100\% \tag{1}$$

$$\frac{\Delta z}{z_0} = \frac{z_0 - z_1}{z_0} 100\% \tag{2}$$

$$\frac{\Delta x}{x_0} = \frac{x_0 - x_1}{x_0} 100\% \tag{3}$$

In the fourth part of the experiment, the influences of pressure, membrane dimensions and LTCC material on the average membrane displacement and displacement repeatability were analyzed. The test structures were fabricated in the same way as presented in the second part of the experiment. However, the thicknesses of the membranes were approximately the same for all samples and were approximately equal to 290, 280 and 260 μm for the Ceram Tape, HL2000 (two layers) and DP951, respectively. The membrane thicknesses were calculating based on the third part of the experiment (measuring the compressibility). The average deflection was calculated based on measurements of 12 samples. The deflections after lamination and sintering were calculated as $h - h_1$ and $h - h_2$, respectively, (Fig. 2).

Moreover, the variability coefficient was calculated for all of the calculated values. The variability coefficient is given by Eq. (4) (where $\sigma$ is the standard deviation and $\bar{y}$ is the average value).

$$V = \frac{\sigma}{\bar{y}} 100\% \tag{4}$$

## 3. Results and discussion

The samples from the first part of the experiment after firing were laser cut and cross-sections were fabricated. The bonding quality was investigated based on cross-sectional observations. The sample bonding quality was first investigated by optical microscopy. The examination confirmed a good quality of lamination for all the analyzed tapes and pressures, and no delaminations were noticed in this stage. Therefore, only the structures joined at the three lowest pressures (0.5, 1 and 2 MPa) were analyzed using SEM. The influence of the lamination pressure on the bonding quality of samples based on Ceram Tape GC, DP951, HL2000 and HL800 at various pressures, is presented in Figs. 4 and 5 (samples after firing). The former interface between tapes is marked with a white arrow in all of the figures. Contamination after laser cut is also described in the figures. A single sheet of the zero-shrinkage tape consists of three sub-layers: the top, bottom and inner layers (marked in Fig. 5b and c). The top and bottom layers are plastic and enable lamination of the tapes. The inner layer consists of a different powder composition in comparison to the other two layers, and this inner layer does not become denser during firing. Therefore, the inner layer significantly reduces the tape shrinkage along the *x* and *y* axes. The former interfaces between the Ceram Tape GC tapes after lamination at the two lowest examined pressures are still visible in the cross-sections. A pressure equal to or higher than 2 MPa ensures a good bonding quality. The bonding quality between D951 (Fig. 5a), HL2000 (Fig. 5b) and HL800 (Fig. 5c) is good for the lowest examined pressure (0.5 MPa). The highest porosity was observed for the HL2000 tapes. The porosity is more visible because of the higher magnification in Fig. 5b in comparison to Figs. 4 and 5a. However, the magnification in Fig. 5c is approximately the same as in Fig. 5b. The influence of the lamination pressure on the green-tape density and shrinkage of the fired samples is discussed in more detail in the third part of the experiment. The test-structure cross-sections in the green state were not investigated because the lamination strengths before firing were too low, and the structures were damaged during the fabrication of cross-sections. Therefore, a great influence of viscous flow of the glass during firing on the bonding strength between tapes can be assumed. It must be borne in mind that the difference between the lowest pressures that ensure good bonding quality and the differences in porosity can be affected by different tape compositions and firing profiles. There is a great probability that every tape consists of a different binder and glass. The glass transition temperature of a binder affects its yield point. The yield point is very important to achieve a proper lamination quality of tapes in the green state. The lamination pressure must be higher than yield point of the tape to ensure plastic deformation. The deformation ensures compression between the joined materials. Therefore, if a binder with a relatively high glass transition temperature were used in Ceram Tape GC, the lowest lamination pressure must be higher to ensure a proper bonding quality in comparison to the three other tapes. The amount of binder also affects the lamination

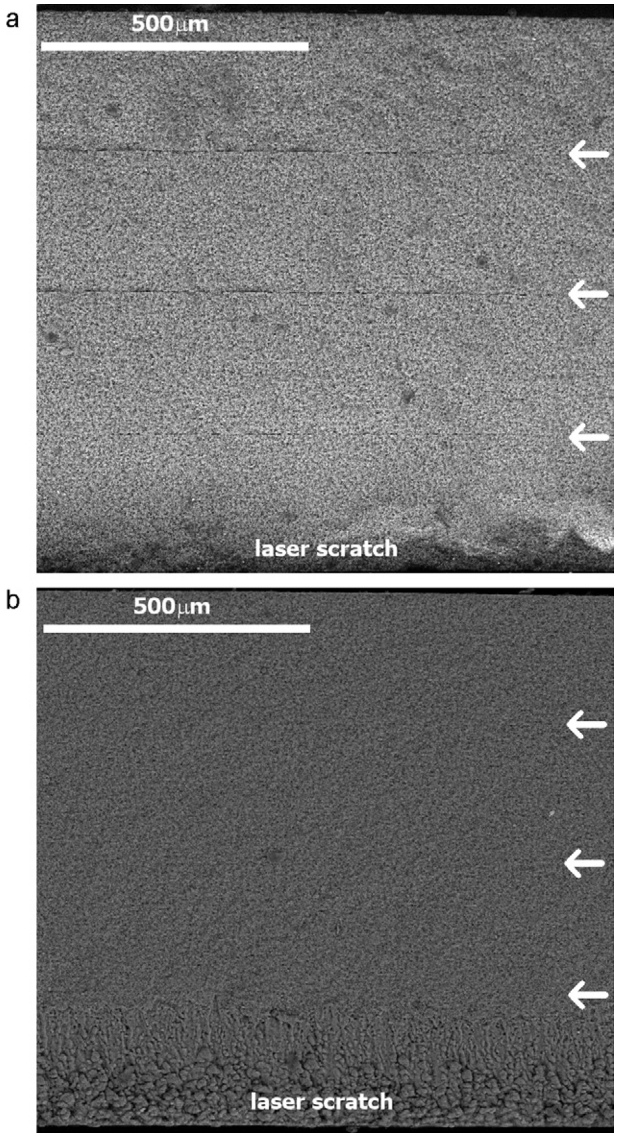

Fig. 4. Laminated Ceram Tape GC tapes after firing, lamination pressures: (a) 1 MPa and (b) 2 MPa.

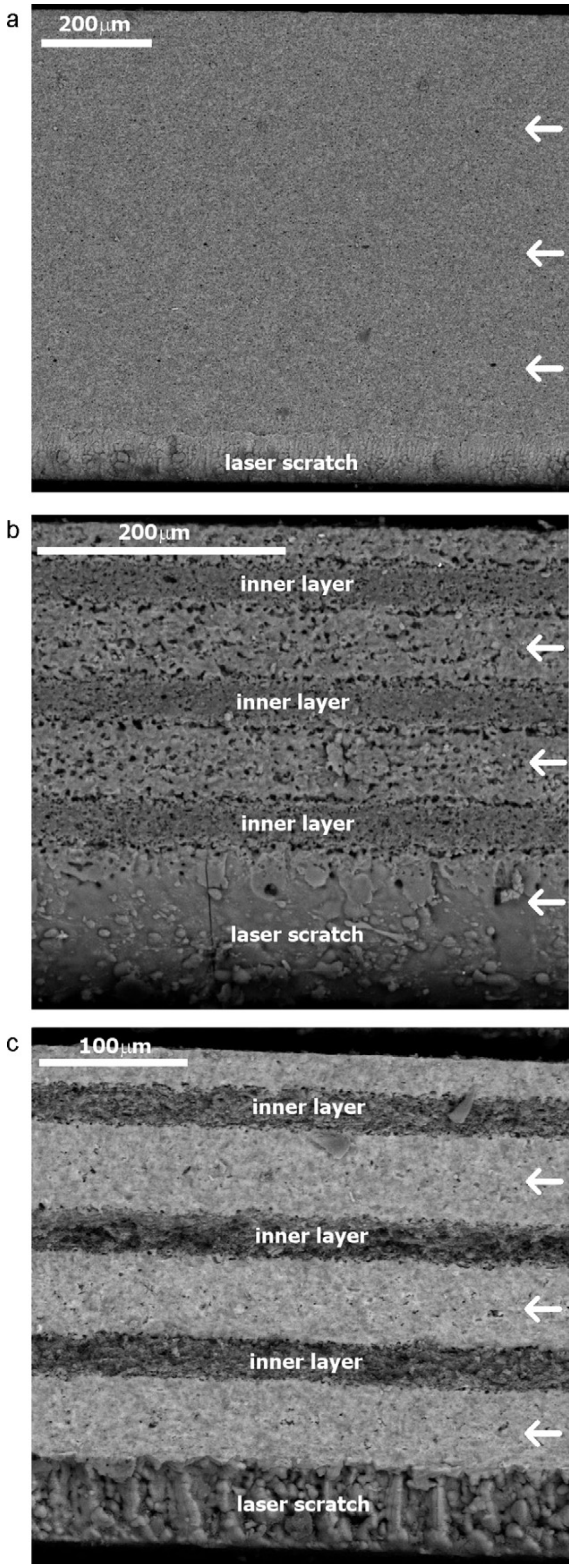

Fig. 5. Bonding quality of tapes laminated at 0.5 MPa after firing: (a) DP951 tapes, (b) HL2000 tapes, and (c) HL800 tapes.

quality and porosity. If a tape has excessive organic material, the bonding quality in the green state can be good. However, during firing of tapes, excessive organic material must be driven off, and the porosity of the final structure can increase. Moreover, gasification of an organic material can affect the bonding quality between tapes. If the tape is lacking in organic material, the lamination in the green state can be too weak to ensure proper joining after firing. The melting temperature of the glass also affects the bonding quality and structure porosity after firing. The porosity observed for the HL2000 structure can also be higher because of insufficient firing. On the one hand, the excessive sintering of LTCC can affect the occurrence of gas within the material.[13] On the other hand, the organic material would not be completely driven off if the firing profile were too short. Because of the different material contents of the tapes, the firing profiles vary. During firing, all of the organic materials must be released, and the glass must be melted to ensure proper bonding quality and low porosity. Therefore, a proper firing profile must be used for each LTCC tape. In all of the experiments presented herein, the standard profiles recommended by the tape suppliers (Table 1) were used to ensure good debinding and sintering. Based on the experimental results, a pressure equal to 0.5 MPa was chosen to laminate the HL800, HL2000 and DP951 test samples. A pressure equal to 2 MPa was chosen to laminate the Ceram Tape GC test samples in the subsequent parts of the experiment.

The samples from the second part of the experiment after firing were laser cut, and cross-sections were fabricated. The lamination quality was investigated based on cross-sectional observations. SEM images of the smallest membranes (square sides equal to 2 mm) for the HL800, HL2000 and DP951 samples laminated with an insert material are presented in Fig. 6a–c, respectively. The structure based on Ceram Tape GC was not examined in this part of the experiment because of the higher lamination pressure that was necessary to join these tapes together and the significantly thicker membrane (the thickness was approximately equal to 335 µm). The former interface between the tapes is marked with a white arrow in all of the figures. Contamination after laser cut is also described in the figures. Once more, dark strips that can be observed in the HL800 and HL2000 tapes were caused by the zero-shrinkage tape composition, which is described in more detail in the last paragraph. The lamination quality is quite good for the DP951 and HL800 structures. However, a higher porosity near the former interface between the layers can be observed for the HL2000 structure. In this case, the structure without thick-film electronic components, this bonding can be assumed to be adequate. The higher porosity is most likely caused by low lamination pressure, which was described precisely in the last paragraph. The membrane deflection is very low for all of the depicted structures. However, the HL800 sample deformed at the bottom part of the structure. The structure in the green state did not deform, and the defect occurred after sintering. However, the phenomenon can be only observed if the structures were previously milled or laser cut. Therefore, the defect also occurred for membranes and was not observed for samples from the first part of the experiment. Typically, such deformation can be reduced using, for example, a longer firing profile or by firing the structure once more with

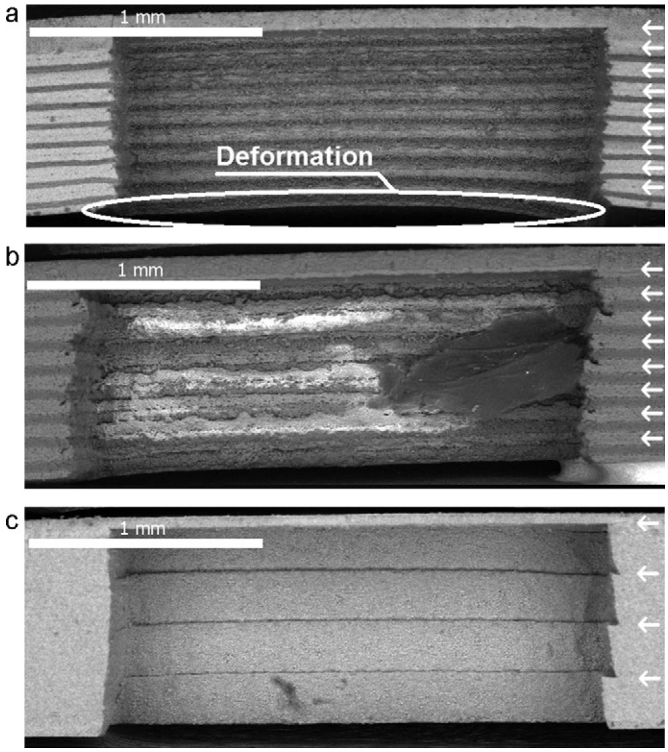

Fig. 6. Membranes after firing, tape: (a) HL800,[32] (b) HL2000, and (c) DP951 (membrane thickness of $40\,\mu\text{m}$).

a load, which is explained in more detail in the introduction. However, neither of the solutions works in this case. This finding can be explained by crystallization of the glass in the LTCC during firing. Therefore, during the second firing, there was no glass that could flow. Additional experiments should be carried out to prove this hypothesis. However, according to the knowledge of the authors, HL800 is no longer commercially available. Because of deformation of 3D structures fabricated using HL800 and a lack of material in the third and fourth experimental stages, the tapes were not analyzed.

In comparison to solvent-based chemical lamination, thermo-compressive bonding with an insert material enables the lamination of very thin membranes (down to $40\,\mu\text{m}$). The thinnest tapes that were successfully bonded using solvent-based chemical lamination were more than $100\,\mu\text{m}$ thick. Thinner tapes were completely dissolved by the solvents. Moreover, lamination with an insert is less complicated in comparison to the solvent bonding, and there is no need to look for new solvents to fulfill the requirements of different tape compositions.

The influence of the lamination pressure and LTCC material on LTCC compressibility and shrinkage were investigated in the third part of the experiment and are presented in Fig. 7. The average relative compressibility and average shrinkage were calculated based on measurements of 12 samples. The influence of the lamination pressure on the relative compressibility of green tapes and on the variability coefficient of the relative compressibility is presented in Fig. 7a and b, respectively. The relative compressibility was calculated based on Eq. (1). Therefore, if the relative compressibility is higher than 100%, the compression is smaller in comparison to the relative value. The tape consisted of organic material, ceramic powder and glass. However, the mixture also consisted of some air bubbles. Hence, during lamination, the material can be pressed, the air bubbles are driven off, and the material density increases. The compressibility decreases (relative compressibility increases) if the lamination pressure decreases for all of the examined samples, which is consistent with the theory. The highest impact of pressure on compressibility can be observed for Ceram Tape GC. The difference in compressibility between the HL2000 and DP951 structures is rather small. The Ceram Tape GC relative compressibility is higher for a lower pressure in comparison to the DP951 and HL2000 structures. This phenomenon is caused by tape composition and tape thickness. The Ceram Tape GC most likely contains a binder with a higher glass transition temperature, and therefore the organic material is not plastic enough for lower pressures. Hence, the compressibility is significantly smaller for these samples. The compressibility of thicker tapes is also higher according to the theory, which is caused by trapping of more air bubbles in the tape during tape casting. This characteristic can affect the compressibility of Ceram Tape GC tapes. A higher compressibility for 13.5 MPa for the HL2000-based structures was most likely caused by a measuring error, which was approximately 5% of the measured value. The variability coefficient of the relative compressibility did not depend on the pressure for DP951 and Ceram Tape GC samples and did not exceed 0.5%. The variability coefficient was expected to increase if pressure decreased, but the change for DP951 and Ceram Tape GC samples could be so small that it was not possible to measure. The variability coefficient increased if the pressure decreased for the HL2000-based structures, which is consistent with the theory. This exception is apparent for the lowest pressures used for lamination of the HL2000 tapes. The variability coefficient in this case was approximately equal to zero. This finding was a coincidence due to the small number of samples that were tested (12). Pressure has a greater effect on the variability coefficient of the relative compressibility of HL2000 tapes in comparison to the DP951 and Ceram Tape GC samples. However, even for the HL2000 samples, compressibility does not exceed 1.5%. The higher variability coefficient observed for the HL2000 structures was most likely caused by a more complicated structure (zero-shrinkage material) or by the higher porosity of the HL2000 samples in comparison to the others.

Tape shrinkage along the $x$ and $y$ axes and along the $z$ axis are presented in Fig. 7c and e, respectively. The shrinkage variability coefficients along the $x$ and $y$ axes and along the $z$ axis are shown in Fig. 7d and f, respectively. The variability coefficient varies for different pressures for each tape. However, no clear trend can be observed. Lack of a clear trend is caused by the small value of the coefficient below 1.6 and 1% for the $z$ axis and the $x$ and $y$ axes, respectively. Therefore, it can be inferred that the average value of the measurements is quite stable for all the pressures, and the deviation does not depend on the lamination pressure. Hence, shrinkage can be predicted with very good accuracy. The average shrinkage should depend on compressibility of the tape and should therefore be affected by the lamination pressure. If the compressibility is small, there is still a relatively large amount of air bubbles trapped in the green, laminated stack.

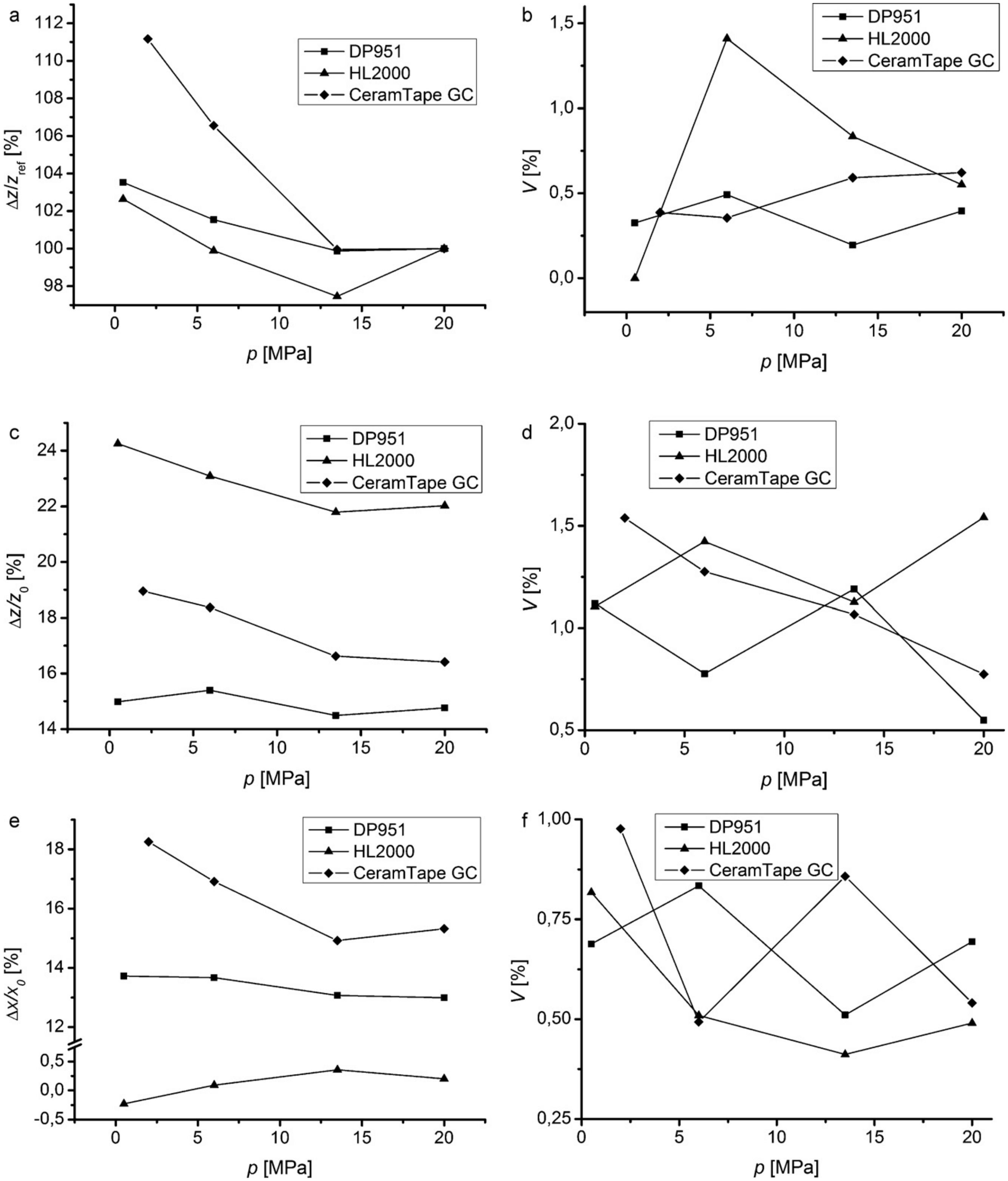

Fig. 7. Relative compressibility and shrinkage: (a) average relative compressibility, (b) variability coefficient of compressibility, (c) shrinkage along the *z* axis, (d) variability coefficient of shrinkage along the *z* axis, (e) shrinkage along the *x* and *y* axes, and (f) variability coefficient of shrinkage along the *x* and *y* axes.

During firing, the glass melts and can fill the voids where the air bubbles were previously. Hence, shrinkage can be higher in comparison to structures laminated using higher pressures. Such a tendency can be observed for shrinkage along the *x* and *y* axes for CeraTape GC samples (Fig. 7c) and for shrinkages along the *z* axis for HL2000 and Ceram Tape GC (Fig. 7e). These curves are consistent with the theory described above. The shrinkages along the *x* and *y* axes for HL2000 and shrinkages along all of the axes for the DP951 structures does not depend on the lamination pressure. The explanation for HL2000 is that this tape is a zero-shrinkage tape, and therefore, the shrinkage along the *x* and *y* axes is almost zero. The stability of shrinkage for DP951 is affected by a small relative compressibility. The glass transition temperature of the DP951 binder is relatively low, and hence, tape could be properly laminated at very low pressures. As a result, the compressibility was not affected by the pressure and therefore, shrinkage was neither.

In the fourth part of the experiment, the influences of pressure, membrane dimensions and LTCC material on the average membrane displacement and displacement repeatability after lamination and sintering were analyzed. The average deflection was calculated based on measurements of 11 samples. The membrane deflection after lamination (green state) vs. the membrane square size and the standard deviation of membrane deflection after lamination (green state) vs. membrane square size for various tapes laminated with and without inserts are presented in Fig. 8a and b, respectively. Proper lamination can be achieved if both the temperature and pressure are high enough to enable plastic flow of the bonded LTCC materials (above the yield point of LTCC). Plastic flow of the material causes deformation of the membranes and walls of the 3D structures as is schematically presented in Fig. 2. The membrane and wall deformation increase if the bonding temperature or pressure increases. Therefore, if the temperature and pressure are decreased, collapse of the 3D structure can be diminished. However, deformation will be always present because of plastic flow, which must occur during thermo-compressive lamination. Therefore, the application of a rubber-like filler that fills the open chamber during the lamination significantly reduces the collapsing effect of 3D structures. As seen in Fig. 8a, the deflection of HL2000 membranes is slightly higher in comparison to other two LTCC materials. However, it must be borne in mind that the HL2000 membranes are slightly thinner, and therefore, the deflection is higher. Moreover, the tendency of membrane deflection is consistent with theory and is higher if the membrane thickness is smaller. Hence, the lowest and highest deflections were measured for Ceram Tape (membrane thickness approximately equal to 290 μm) and HL2000 (membrane thickness approximately equal to 260 μm), respectively. The deflection of membranes laminated using fillers was approximately the same for all materials. Moreover, fillers significantly reduced variations in the membranes thicknesses, which was caused by the protecting properties of the filler during lamination. Moreover, neither influence of the membrane thickness nor LTCC

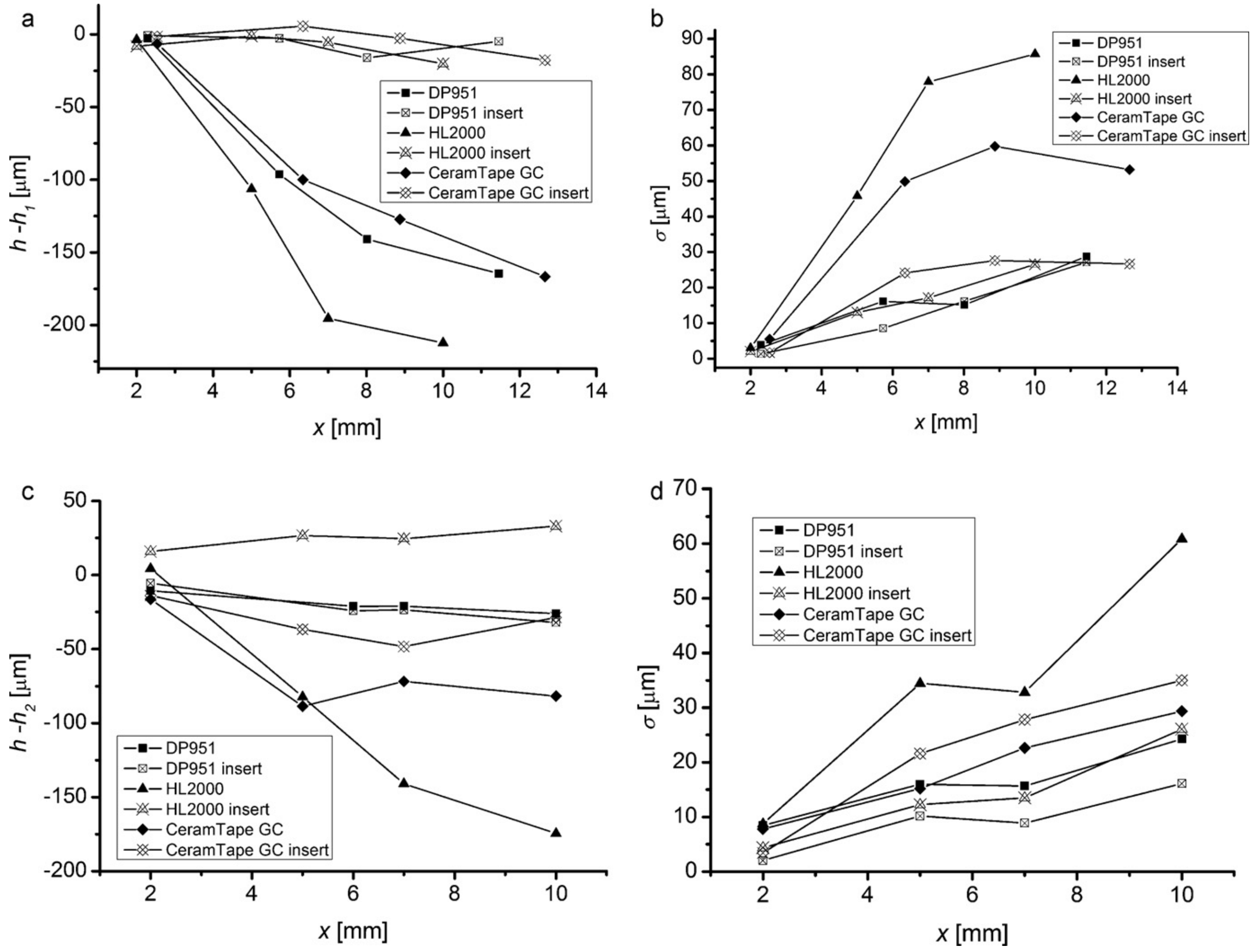

Fig. 8. Membrane deflection: (a) deflection after lamination, (b) standard deviation of deflection lamination, (c) deflection after firing, and (d) standard deviation of deflection after firing.

material on deflection was observed in this case. However, as seen in Fig. 8b, the LTCC composition significantly affected the standard deviation of membranes. The lowest standard deviation was found for the DP951 structures, and moreover, the standard deviation for this tape is approximately the same for structures laminated with and without fillers. The HL2000 membranes are the thinnest, and therefore, the highest deviation was observed for these samples. However, the thicker Ceram Tape GC membranes exhibit a higher standard deviation of membrane deflection in comparison to the DP951-based membranes, which was most likely caused by the different material composition. Hence, the material composition might affect the standard deviation of membrane deflection more than the tape thickness.

The structures after measurements were fired in thermal cycles presented in Table 1. The membranes were inverted, and membranes faced down during firing. Therefore, planarization of membranes during firing was possible. A decrease in deflection is affected by the softening of glass during firing. If the glass is soft, the force of gravity is sufficient to decrease the membrane deflection slightly. Membrane deflection and the standard deviation of membrane deflection after firing (LTCC in sintered state) are presented in Fig. 8c and d, respectively. The significance of firing is very apparent in both figures. The firing process enables permanent bonding of tapes because of viscous flow of the melted glass during firing. However, viscous flow of glass also affects membrane deflection and the standard deviation of deflection. During lamination of the tapes, some residual stresses can be generated in a plastic material. During the relaxation of stresses during firing, deformation of the membrane occurs. This deformation significantly depends on the LTCC composition and residual stress. This phenomenon is clearly visible in Fig. 8c. The deflection of the DP951-based membranes was approximately the same for structures laminated with and without rubber-like inserts. The standard deviation of deflection for the DP951 structures laminated with inserts is just slightly lower in comparison to the DP951 structures thermo-compressed without a filler. Hence, the protective role of filler is not so apparent after firing in this case. The deflection of membranes laminated using fillers increased after firing, which was most likely due to relaxation of residual stress during this process. The deformation of samples laminated without a filler decreased after firing process. The deformation most likely decreased because of softening of the glass and flattening effect caused by gravity, which was described above. The standard deviation decreased for samples laminated without a filler but increased for samples with a filler. This difference is caused by the same phenomenon that was described for fired membranes deflections.

## 4. Conclusions

A very low pressure, approximately equal to 0.5 MPa, can be used to laminate green LTCC tapes. However, good bonding quality is achieved after sintering.

Very thin tapes (even 40 μm) can be laminated using thermo-compressive bonding with an insert rather than by solvent lamination. However, the thinnest tapes are very soft, and therefore handling the tapes is very difficult, and the relative deflection is the highest. Lamination with an insert is less complicated in comparison to solvent bonding, and there is no need to look for new solvents for different tape compositions.

The highest impact of pressure on compressibility can be observed for Ceram Tape GC. The difference in compressibility between the HL2000 and DP951 structures was rather small. The Ceram Tape GC relative compressibility was higher for a lower pressure in comparison to the DP951 and HL2000 structures. This phenomenon was caused by the tape composition and tape thickness. The Ceram Tape GC most likely contained a binder with a higher glass transition temperature, and therefore the organic material was not plastic enough for the lower pressures. Hence, the compressibility was significantly smaller for these samples. The compressibility of thicker tapes as also higher, in accordance to theory, and was caused by trapping of higher amount of air bubbles in the tape during tape casting.

During firing, the glass melts and can fill voids previously occupied by air bubbles. Hence, shrinkage can be higher in comparison to structures laminated using higher pressures. Such a tendency can be observed for shrinkage along the *x* and *y* axes for CeraTape GC samples and for shrinkages along the z axis in HL2000 and Ceram Tape GC. The shrinkages along the *x* and *y* axes for HL2000 and shrinkages along all of the axes for the DP951 structures did not depend on the lamination pressure. The explanation for HL2000 is that this tape is a zero-shrinkage tape, and therefore, the shrinkage along the *x* and *y* axes was almost zero. The stability of shrinkage for DP951 was affected by a small relative compressibility. The glass transition temperature of the DP951 binder was relatively low, and hence, the tape could be properly laminated at very low pressures. As a result, the compressibility was not affected by the pressure and therefore, shrinkage was neither.

The membrane and wall deformation increased if either the lamination temperature or pressure increased. Therefore, if the temperature and pressure were decreased, collapse of the 3D structure could be mitigated. However, deformation will be always present because of plastic flow, which must occur during thermo-compressive lamination. Therefore, the application of a rubber-like filler, which fills the open chamber during lamination significantly reduces the collapsing effect of 3D structures.

The firing process enables permanent bonding of the tapes because of viscous flow of the melted glass during firing. However, viscous flow of glass also affects membrane deflection and the standard deviation of deflection. During lamination of the tapes, some residual stresses can be generated in a plastic material. During the stress relaxation during firing, deformation of the membrane occurs. This deformation significantly depends on the LTCC composition and residual stress.

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## References

1. Gongora-Rubio MR, Espinoza-Vallejos P, Sola-Laguna L, Santiago-Aviles JJ. Overview of low temperature co-fired ceramics tape technology for meso-system technology (MsST). *Sensors and Actuators A* 2001;**89**:222–41.
2. Golonka L. Technology and applications of low temperature cofired ceramic (LTCC) based sensors and microsystems. *Bulletin of the Polish Academy of Sciences* 2006;**54**:223–33.
3. Thelemann T, Thust H, Hintz M. Using LTCC for microsystems. *Microelectronics International* 2002;**3**:19–23.
4. Gupta T. *Handbook of thick- and thin-film hybrid microelectronics*. New Jersey: Wiley; 2003.
5. Peterson KA, Knudson RT, Garcia EJ, Patel KD, Okandan M, HO CK, et al. LTCC in microelectronics, microsystems, and sensors. In: *Proc. 15th international conference MIXDES*. 2008. p. 23–7.
6. Malecha K, Pijanowska D, Golonka LJ, Torbicz W. LTCC microreactor for urea determination in biological fluids. *Sensors and Actuators B* 2009;**141**:301–8.
7. Malecha K, Jurków D, Stiernstedt J, Golonka L. Laser patterning of coloured green ceramic tapes. In: *Proc. 34th IMAPS 2010 conference Poland chapter*. 2010.
8. Kita J, Dziedzic A, Golonka LJ, Zawada T. Laser treatment of LTCC 3D structures and elements. *Microelectronics International* 2002;**19**:14–8.
9. Pitt KEG. *An introduction to thick film component technology*. Luton: Mackintosh Publications Limited; 1981.
10. Khoong LE, Tan YM, Lam YC. Overview on fabrication of three-dimensional structures in multi-layer ceramic substrate. *Journal of the European Ceramic Society* 2010;**30**:1973–87.
11. Shan X, Maw HP, Lu CW. Solvent-assisted low pressure room temperature lamination of low temperature cofirable ceramic green tapes for formation of embedded micro channels. *Microsystems Technology* 2010;**16**: 1501–6.
12. Roosen A. New lamination technique to join ceramic green tapes for the manufacturing of multilayer devices. *Journal of European Ceramic Society* 2001;**21**:1993–6.
13. Imanaka Y. *Multilayered low temperature cofired ceramics (LTCC) technology*. New York: Springer; 2005, ISBN 0-387-23130-7.
14. Cahn RW, Haasen P, Kramer EJ. *Materials science and technology: processing of ceramics v. 17: a comprehensive treatment (materials science and technology: a comprehensive treatment)*. Weinheim: Verlag Chemie GmbH; 1996.
15. Piwonski MA, Roosen A. Low pressure lamination of ceramic green tapes by gluing at room temperature. *Journal of the European Ceramic Society* 1999;**19**:263–70.
16. Roosen A, Schindler K. Cold low pressure lamination of ceramic green tape. In: *Proc. 1st international conference and exhibition on ceramic interconnect and ceramic microsystem technologies*. 2005. p. 397–403.
17. Roosen A, Schulte T, Sibert M, Zollner S. Ceramic green body, method of manufacturing a green body of this type and method of manufacturing a ceramic body using the green body. United State Patent 20040011453; 2004.
18. Roosen A, Schroder A, Zollner S. Method for joining ceramic green bodies using transfer tape and conversion of bonded green body into a ceramic body. United State Patent 7318874; 2008.
19. da Rocha ZM, Ibañez Garcia N, Oliveira NA, Matos J, Rosário D, Gongora-Rubio MR. Low temperature and pressure lamination of LTCC tapes for meso-systems. In: *Proc. of IMAPS conference and exhibition on ceramic interconnect technology*. 2004.
20. Gurauskis J, Sanchez-Herencia AJ, Baudin C. Joining green ceramic tapes made from water-based slurries by applying low pressures at ambient temperature. *Journal of the European Ceramic Society* 2005;**25**:3402–11.
21. Sanchez-Herencia AJ, Gurauskis J, Baudin C. Processing of $Al_2O_3$/Y-TZP laminates from water-based cast tapes. *Composites B* 2006;**37**:499–508.
22. Burdon J W, Huang R, Wilcox D, Naclerio NJ. Method for fabrication a multilayered structure and the structures formed by the method. United States Patent 6592696; 2003.
23. Suppakarn N, Ishida H, Cawley JD. Roles of poly (propylene glycol) during solvent-based lamination of ceramic green tapes. *Journal of the American Ceramic Society* 2001;**84**:289–94.
24. Jurków D, Roguszczak H, Golonka LJ. Cold chemical lamination of ceramic green tapes. *Journal of the European Ceramic Society* 2009;**29**:703–9.
25. Jurków D, Golonka L. Cold chemical lamination—new bonding technique of LTCC green tapes. *International Journal of Applied Ceramic Technology* 2010;**7**(6):814–20.
26. Birol H, Maeder T, Ryser P. Application of graphite-based sacrificial layers for fabrication of LTCC (low temperature co-fired ceramics) membranes and micro-channels. *Journal of Micromechanics and Microengineering* 2007;**17**:50–60.
27. Peterson KA, Patel KD, Ho CK, Rohde SB, Nordquist CD, Walker CA, et al. Novel microsystem applications with new techniques in low temperature co-fired ceramics. *International Journal of Applied Ceramic Technology* 2005;**2**(5):345–63.
28. Malecha K, Golonka LJ. Microchannel fabrication process in LTCC ceramics. *Microelectronics Reliability* 2008;**48**:866–71.
29. Malecha K, Jurków D, Golonka L. Comparison of solvent and sacrificial volume material based lamination processes of Low Temperature Co-fired Ceramics (LTCC) Tapes. *Journal of Micromechanics and Microengineering* 2009;**9**:1–10.
30. Datasheet FR-4 Glass/Epoxy http://www.plasticsintl.com/datasheets/Phenolic_G10_FR4.pdf [accessed 28.10.11].
31. Web material data PDMS, http://www.mit.edu/∼6.777/matprops/pdms.htm [accessed 26.07.11].
32. M. Zawisza 2010 Pressure sensor *Engineer thesis* (Wroclaw University of Technology, Poland).