

## Cold chemical lamination of ceramic green tapes

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### Abstract

The cold chemical lamination (CCL) is a new technique of bonding ceramic green tapes into one 3D structure. Instead of a standard thermo-compression method, new solvent-based lamination is presented. A film of a special chemical agent is put on the green tape surface. The solvent melts the surface. Then the tapes are stacked. The bonding of the green tapes is made at a room temperature. The new method is used for joining green tapes of the low temperature co-fired ceramics (LTCC). A quality of the bonding depends on the solvent type. The cold chemical lamination is examined on two types of the LTCC tapes: DuPont 943 and DuPont 951. Six types of the solvents are analyzed in the paper. The bonding quality and geometry of the test structures are examined. The lamination quality is investigated by the scanning electron microscope.

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*Keywords:* Joining; Pressing; Lamination; LTCC

### 1. Introduction

The low temperature co-fired ceramic (LTCC) technique is well known for last two decades. The market of the LTCC devices is still growing. It is caused by the advantages of the LTCC. It combines advantages of a thick film and a high temperature co-fired ceramic (HTCC) techniques. The LTCC technological process consists of: tape casting, shapes cutting in the green tape ceramic, screen printing, lamination and cofiring. All these steps are important to achieve good quality of the final structure. However, lamination of the low temperature co-fired ceramic is one of the most important technological steps during a manufacturing of sensors,<sup>1–4</sup> channels,<sup>2,4,5</sup> microfluidic systems,<sup>6</sup> chambers, reactors,<sup>2,7</sup> cooling systems,<sup>4,5</sup> micropumps,<sup>4</sup> microwave filters,<sup>8</sup> miniaturize antennas<sup>8</sup> and others 3D devices made in the LTCC technique. A standard thermo-compression lamination is the most common method of bonding green tapes. The bonding is created at temperature 50–80 °C at high pressure above 5 MPa, for 2–15 min. During this process the LTCC tapes are getting soft and are joining together. The method has several advantages. Many layers can be bonded in one package. The bonding is strong. However, there is one important problem. High pres-

sure and temperature cause deformation of a manufacturing structure.<sup>5</sup> The problem can be reduced by using fugitive phase (sacrificial material) intended to disappear during a cofiring process.<sup>9,10</sup> The first alternative method of bonding LTCC tapes was shown by Roosen.<sup>11–13</sup> He called it cold low pressure lamination (CLPL). It is based on temporary gluing step. The LTCC green tapes are bonded together by two side adhesive tape. During the burning process the adhesive tape is melting and diffusing into the LTCC tapes. The method has many advantages: the bond is made at room temperature at low pressure, the chambers and channels are not deformed during the process. However, it has also disadvantages. The close chambers cannot be done. The laminated stack might crack during the burning process. It is common for structures consists above 10 tapes with close chamber. The effect might be caused by increasing of pressure in the chamber during firing. The method can be used for making open chambers and channels.

Another adhesive-method was presented by Rocha<sup>14</sup>. The layers are bonded by adhesive liquid. There are used several different types of such substations (e.g. natural honey). The LTCC tapes are covered by the organic liquid film. Then they are stacked together. The liquid glues the layers. The temporary gluing process is realized at very low pressure at room temperature. The bonding achieve by the method is good. The chambers are not deformed. The non-metallized, metallized tapes and chambers are available in the technique. The technique can be an alternative for thermo-compression.

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Cold chemical lamination (CCL) is presented in the paper. It is solvent-base method of lamination. The LTCC tape surface is covered by a film of a solvent. The tape surface is melted by the liquid. Then the tapes are stacked and compressed by pressure below 0.5 MPa. The bonding is created immediately. Since then the layers cannot be separated any more. The bonding between the layers is based on a diffusion process. The method has many advantages. The bonding is strong. No others materials are used during lamination, the solvent is vaporized completely. The big close chambers can be fabricated in the method. However, it has also disadvantage. The film of the solvent must be printed very precisely. Too thick film causes deformation of the structure. Too thin layer do not make strong bonding. The lamination quality is analyzed for six different solvents and for two types of the LTCC tapes (DuPont 951 and DuPont 943). The results are presented in the paper. The bonding quality and chamber geometry are analyzed. The bonding quality is investigated by the scanning electron microscope.

## 2. Experimental

Commercial available DuPont 951 and DuPont 943 LTCC green tapes are used. Six solvents: DuPont thinner 4553, DuPont thinner 9450, DuPont thinner 8250, DuPont thinner 4036, plasticizers butyl benzyl phthalate (S-160) typically used in a tape casting process<sup>15</sup> and water are examined. The quality of the bonding and geometry of the cavities are analyzed. A Nd:YAG laser (AUREL NAVS 30 laser trimming and cutting system) is used to cut design in the LTCC tapes. Such laser is used typically for thick film resistors trimming. However, it is possible to use the laser for cutting, thanks to a special software.<sup>5</sup> The test structures consist of a top part, a bottom part and a middle part with chamber. The cavities are squares with a side long 2, 5 or 10 mm before firing process. The cavities are one or two LTCC green tapes high. Not smooth walls of the chambers in all presented figures are caused by too low laser power. The open and the close cavities are examined. The ceramic tapes are laminated using CCL and common thermo-compression lamination. The CCL is done at room temperature under atmospheric pressure. The thermo-compression is done at 70 °C, at 5 and 10 MPa pressure for 10 min. The progressive thermo-compression lamination is used. It is based on bonding of the tapes in a few steps. In the first layers from the top and the bottom part of the structure are bonded. In the second step the parts laminated in the first step with are bonded layers from the middle part. Such solution permits to decrease the deformation of the module during the lamination process. The structures are co-fired in common cofiring cycle recommended by DuPont. Two different burning cycles are used: longer for DuPont 943 and shorter for DuPont 951 LTCC tape. Co-fired structures are cut by diamond blade. The LTCC structure is examined by the optical and the scanning electron microscope.

## 3. Results and discussion

The first observation of the bonding quality is made by an optical microscope. It is found that the DuPont thinner 9450,

DuPont thinner 8250, DuPont thinner 4036 are not a good solvents. The use of them does not give strong bonding between the LTCC tapes in case of the close chambers. The open one can be achieved. The water do not give good bonding quality in both cases. The best bonding between the LTCC tapes, without deformation of the chambers, is achieved by using thinner DuPont 4553 and plasticizers S-160. The same good bonding is achieved in case of both LTCC tapes (DuPont 943 and DuPont 951). The structures laminated with thinner DuPont 4553 and with standard thermo-compression method are observed by scanning electron microscope.

### 3.1. Lamination of DuPont 951 A2 tape

The bonding quality and a chamber geometry can be observed in Fig. 1. The LTCC structure is laminated by the standard thermo-compression method. The chamber is co-fired as the close chamber. The cavity dimensions are 10 mm long, 10 mm wide and 165  $\mu\text{m}$  high (before firing process). The bonding is good. The delaminations are not visible. However, the chamber deformation can be observed. The cross section of the chamber middle part is presented in Fig. 1(b). The cross-section of the chamber left part is shown in Fig. 1(a) and (c). The presented chambers are deformed significantly. It is caused by too high pressure. The fugitive phase is needed to create better geometry of the chamber.

The bonding quality and the chamber deformation are shown in Fig. 2. The LTCC structure is laminated by the standard thermo-compression method. The chamber is co-fired as close one. The cavity dimension in Fig. 2(a) is 5 mm long, 5 mm wide and 165  $\mu\text{m}$  high and in Fig. 2(b) is 2 mm long, 2 mm wide and 165  $\mu\text{m}$  high before firing process. The bonding in Fig. 2(a) is strong. Delaminations are not visible. However, the chamber in Fig. 2(a) is deformed. The top and the bottom part of the structure are laminated in the middle of the chamber cross-section. In Fig. 2(b) chamber is not deformed any more. However, the bonding quality is very weak. The borders of the LTCC tapes can be recognized easily. The higher pressure must be used to ensure good bonding quality.

The bonding quality and the chamber deformation are shown in Fig. 3. The LTCC structure is laminated by the cold chemical lamination. DuPont 4553 thinner is used. The chamber is co-fired as a close one. Fig. 2(a)–(c) present the cavities 10 mm long, 10 mm wide and 330  $\mu\text{m}$  high before firing process. The bonding is strong for the structures presented in Fig. 3(a) and (b). Delaminations are not visible. Fig. 3(b) presents a middle part of the cavity cross-section. It is not deformed. Two layers high cavity without deformations can be achieved. Fig. 3(c) presents structure with delamination. It is caused by main difficulty of the CCL method. The solvent film cannot be too thin. Otherwise, ceramic tapes are not melted enough. Good bonding quality in this case is not possible.

Fig. 4 presents the bonding quality and the chamber deformation. The ceramic tapes are bonded by the cold chemical lamination. DuPont 4553 thinner is used. The chamber is co-fired as close one. Fig. 4(a) and (b) present structure with cavity 5 mm long, 5 mm wide and 165  $\mu\text{m}$  high (before firing pro-

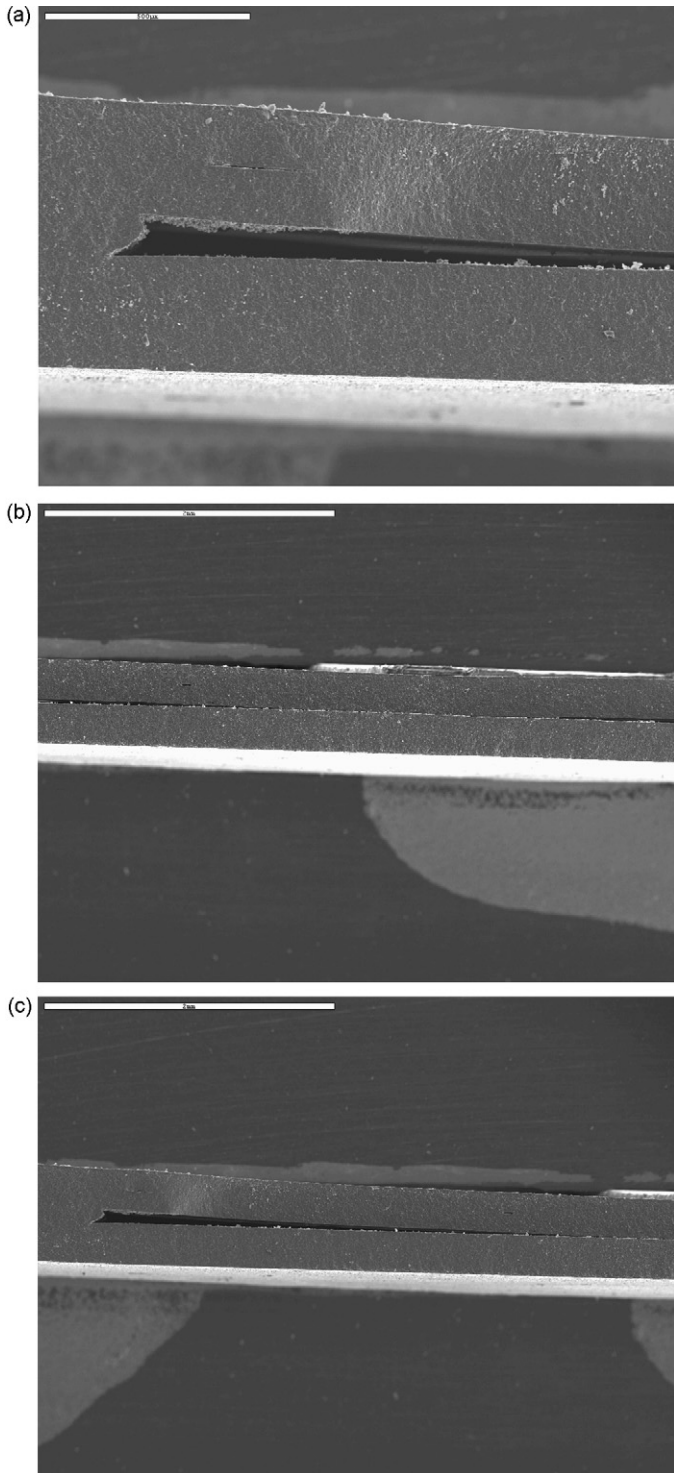


Fig. 1. SEM micrograph of a cross-section of the co-fired LTCC structure (DuPont 951A2) laminated by the thermo-compression method. The chamber is a square with the side long 10 mm, high of the cavity is equal to 165  $\mu\text{m}$  before firing, (a) and (c) cross-section of the chamber left part, (b) cross-section of the chamber middle part.

cess). In both cases the bonding quality is very good. The LTCC tapes are laminated strongly. Fig. 4(b) shows deformation in the middle of the chamber cross-section. This defect is made after lamination process. The middle part of the soft structure above

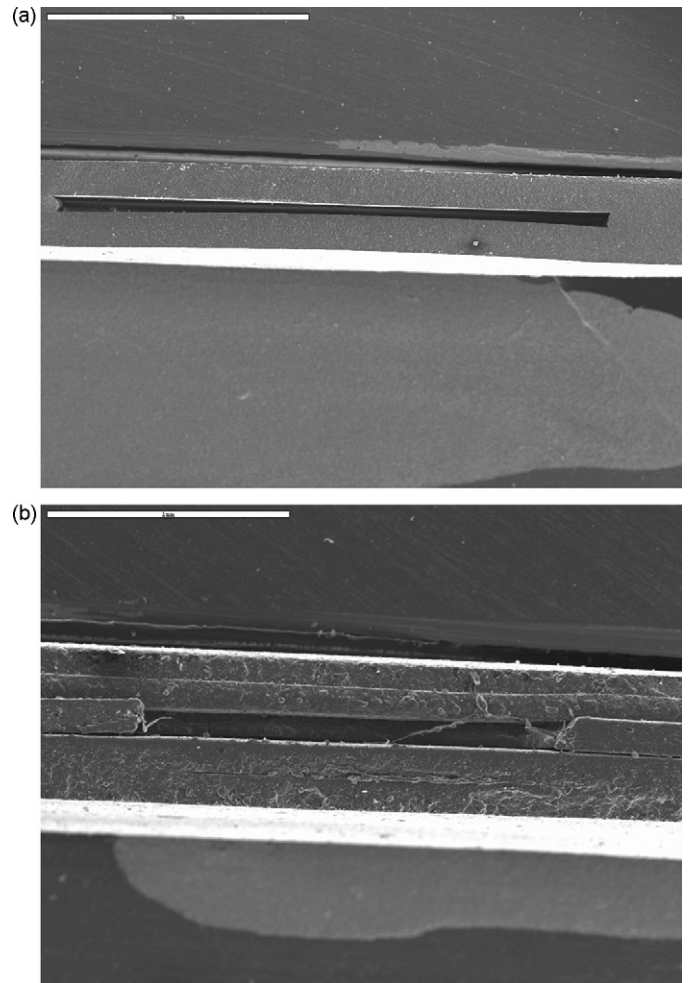


Fig. 2. SEM micrograph of the cross-section of the co-fired LTCC structure (DuPont 951A2) laminated by the thermo-compression method. The chamber is a square with the side long, (a) 5 mm and (b) 2 mm, high of the cavity is equal to 165  $\mu\text{m}$  before firing.

the chamber is pushed after bonding. The error is amplified by the fact after cold chemical lamination that the tapes are soft. The top and the bottom part of the structure cannot be pushed after the CCL process.

The deformation rate of the chamber is lower than in structures which are shown in Fig. 1. The top and the bottom part of the structure are not bonded together. The structures laminated by CCL are soft. They must be dried after bonding. It is highly recommended to use progressive lamination. In the first step the tapes from the bottom part are bonded with layers from the middle part. Then the laminated tapes are dried for about 5 min, at 100 °C. In the next step layers from the top part are laminated and dried. Then the both parts are bonded together. The solution decreases the probability of the chamber deformation. However, soft structures must be treated with care before firing. Otherwise, it causes collapse effect and deformation of the chamber geometry. The deformation made by technological error is smaller than in structure laminated with thermo-compression method, presented in Fig. 1. Structure with cavity 2 mm long, 2 mm wide and 165  $\mu\text{m}$  high (before firing process) is shown in Fig. 4(c). The green tapes are joined by CCL method. DuPont

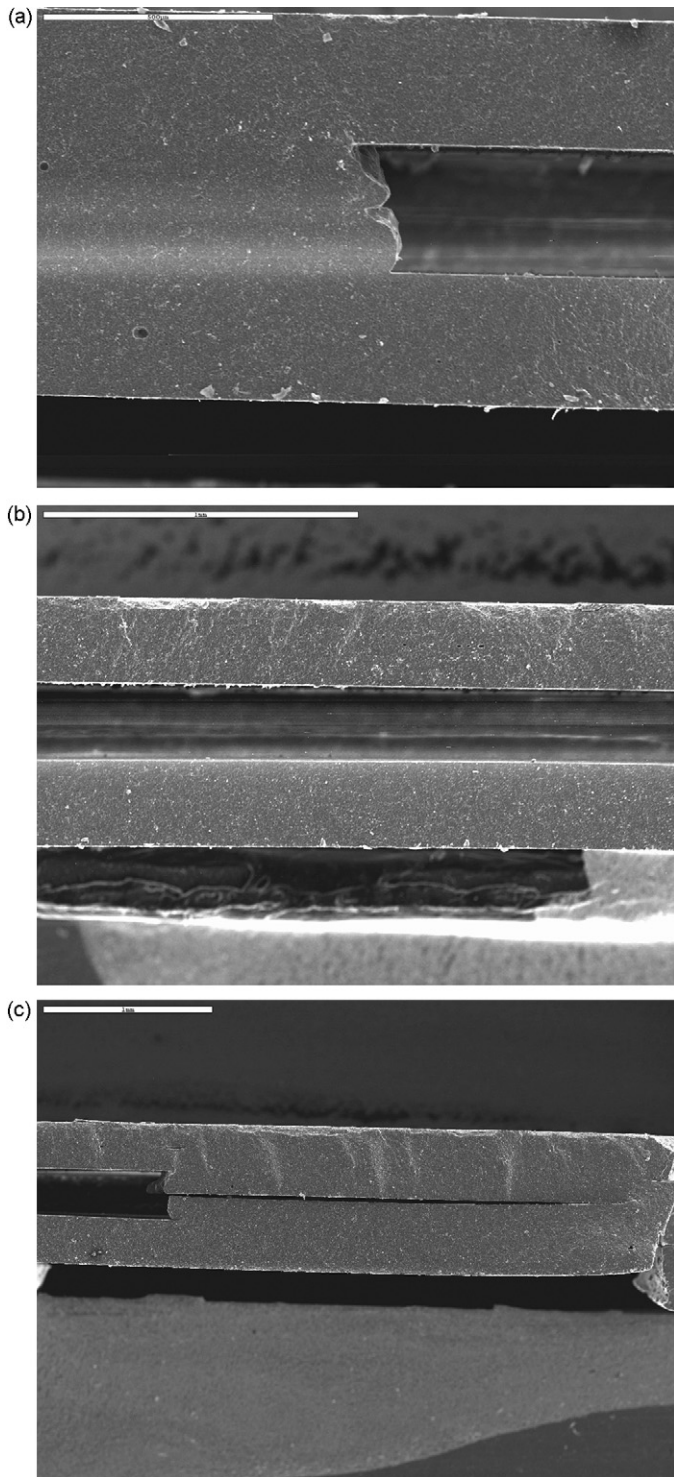


Fig. 3. SEM micrograph of the cross-section of the co-fired LTCC structure (DuPont 951A2) laminated by the CCL method. The chamber is a square with the side long, 10 mm, high of the cavity is equal to  $330\ \mu\text{m}$  (before firing), (a) left, (b) middle and (c) right part of the chamber.

thinner 4553 is used. There is delamination between the top and the middle part of the structure. It is caused by too thin film of the solvent put on the tapes during the CCL process. The bonding between other tapes are strong. The cavity is not deformed.

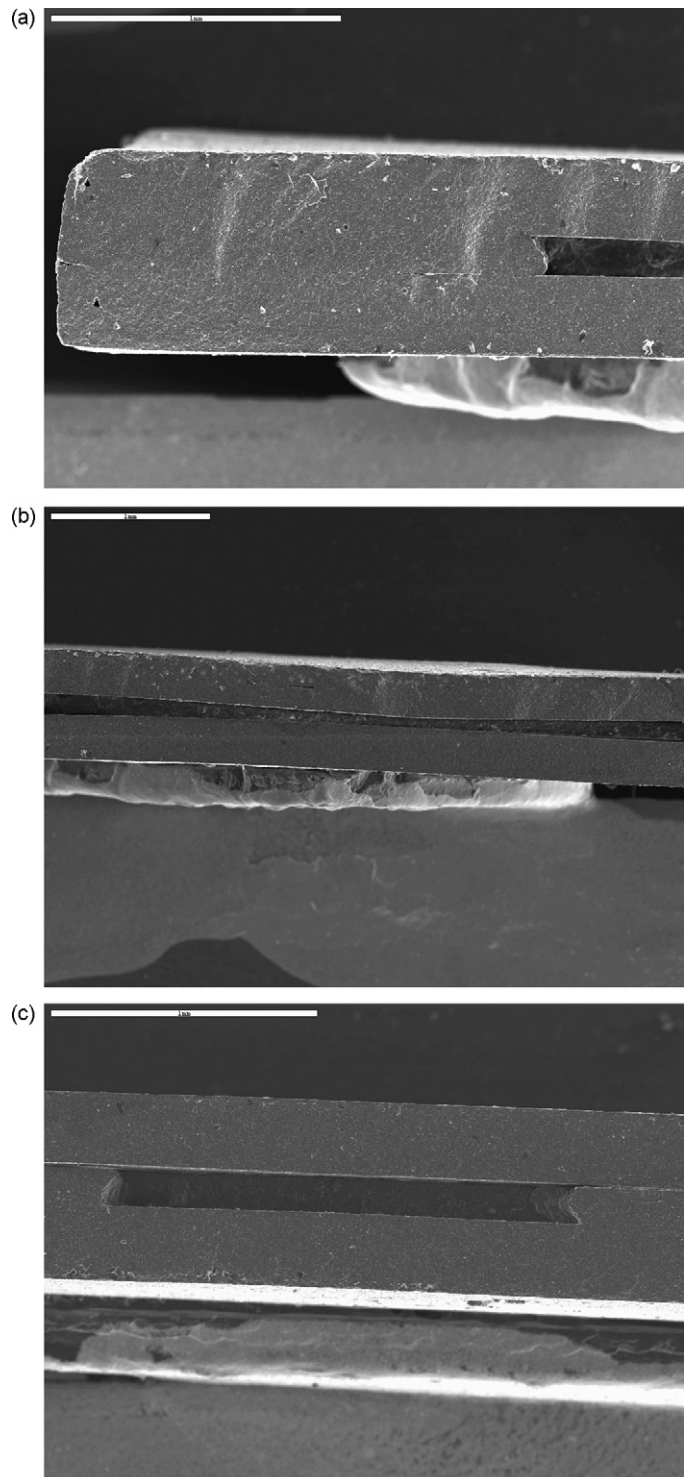


Fig. 4. SEM micrograph of the cross-section of the co-fired LTCC structure (DuPont 951A2) laminated by the CCL method. The chamber is a square with the side long, (a) and (b) 5 mm, (c) 2 mm, high of the cavity is equal to  $165\ \mu\text{m}$  before firing.

### 3.2. Lamination of DuPont 943PX tape

Fig. 5 shows the bonding quality and the chamber deformation of the structure laminated by the standard thermo-compression method. The chamber is co-fired as the close

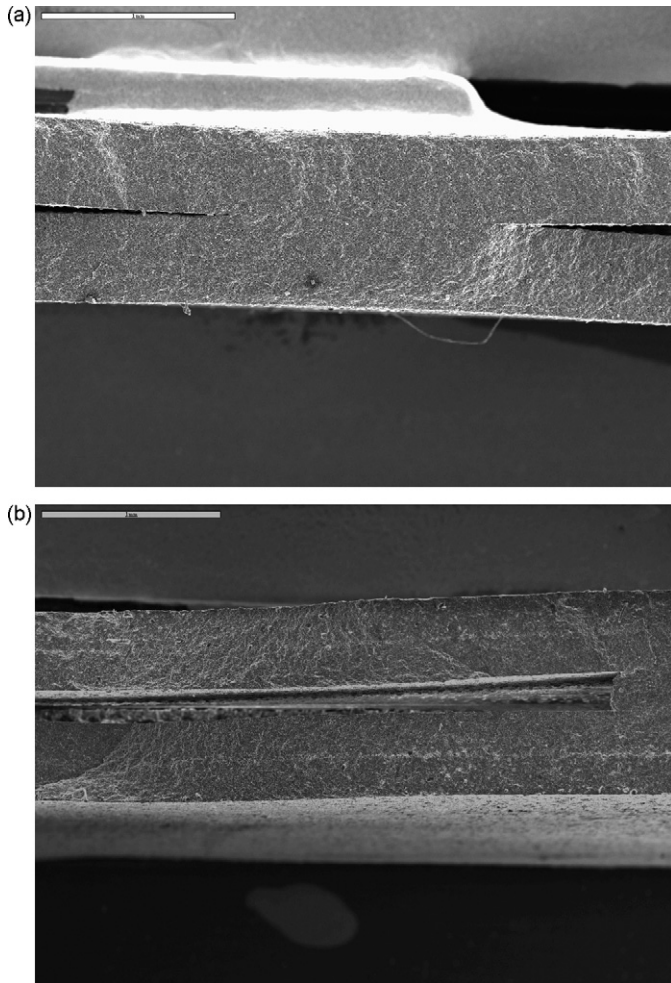


Fig. 5. SEM micrograph of the cross-section of the co-fired LTCC structure (DuPont 943PX) laminated by the thermo-compression method. The chamber is a square with the side long, (a) 10 mm and (b) 5 mm, high of the cavity is equal to 254 μm before firing.

one. Fig. 5(a) presents the structure with cavity 10 mm long, 10 mm wide and 254 μm high (before firing process). Fig. 5(a) shows middle part of the chamber cross-section. The top and the bottom part of the structure are laminated together. The chamber is deformed significantly. The bonding between the LTCC tapes is strong. Fig. 5(b) presents structure with cavity 5 mm long, 5 mm wide and 254 μm high before firing process. The chamber is deformed significantly. The top and the bottom part of the structure are bonded in almost all parts of the chamber. The structure with cavity dimensions 2 mm long, 2 mm wide and 254 μm high (before firing process) is not shown. The module cannot be achieved by the thermo-compression method at pressure 10 MPa. The bonding is too weak and the structure is cracked up. The higher pressure is needed for lamination of DuPont 943PX than DuPont 951A2 tape. The recommended value is about 15–20 MPa. However, so high pressure will deform all chambers inside the LTCC structure. The progressive lamination is not enough in this case.

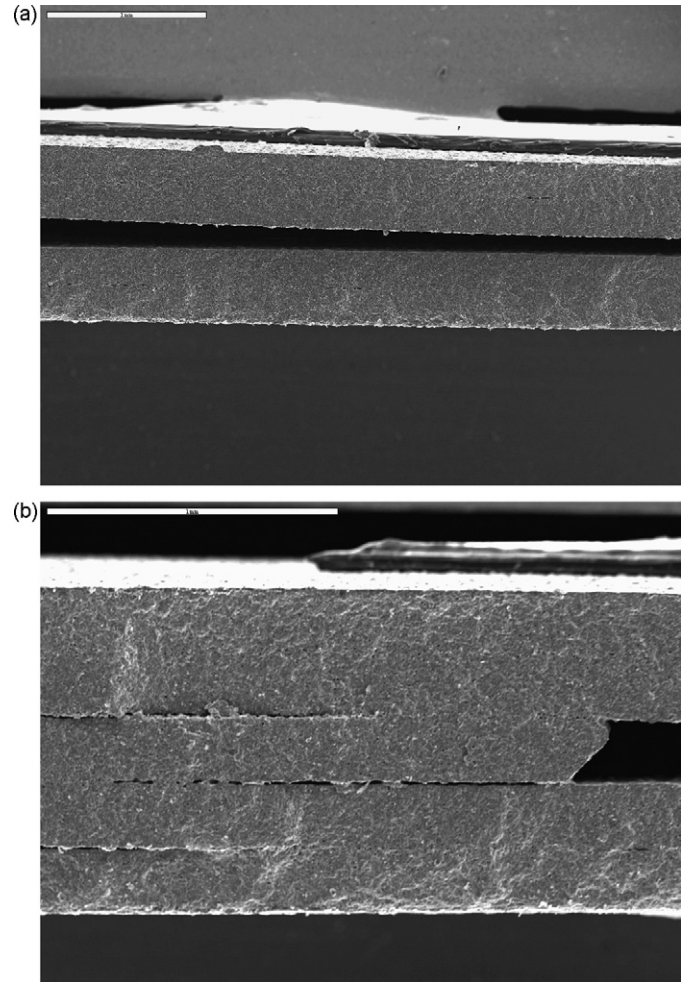


Fig. 6. SEM micrograph of the cross-section of the co-fired LTCC structure (DuPont 943PX) laminated by the CCL method. The chamber is a square with the side long 10 mm, high of the cavity is equal to 254 μm before firing, (a) cross-section of the chamber middle part, (b) cross-section of the chamber right part and (c) cross-section of the chamber left part.

Fig. 6 shows the bonding quality and the chamber deformation. The tapes are bonded by the CCL method. DuPont 4553 thinner is used. The chamber is co-fired as close one. Fig. 6 presents structure with cavity 10 mm long, 10 mm wide and 254 μm high before firing process. Fig. 6(a) shows middle part of the chamber cross-section. It is deformed, but the top and the bottom part of the structure are not laminated together. The result is better than in case of standard thermo-compression method. The bonding quality of the structures presented in Fig. 6(a) is strong. Fig. 6(b) and Fig. 6(c) show delamination of the right and left part of the chamber, respectively. They are caused by too thin film put on the LTCC tapes during the CCL process.

Fig. 7 shows the bonding quality and the chamber deformation. Tapes are bonded by the CCL method. DuPont thinner 4553 is used as the solvent. The chamber is co-fired as the close one. Fig. 7(a) and (b) present structure with cavity 5 mm long, 5 mm wide and 254 μm high (before firing process). The bonding between LTCC tapes is strong. Fig. 7(b) shows the middle part of the chamber cross-section. The cavity is not deformed.

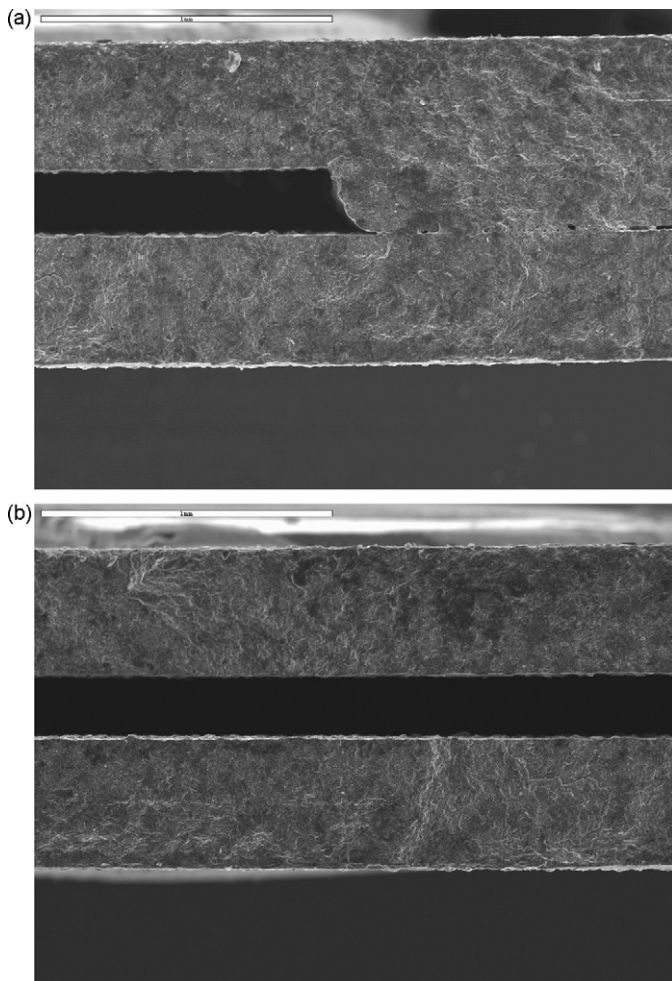


Fig. 7. SEM micrograph of the cross-section of the co-fired LTCC structure (DuPont 943PX) laminated by the CCL method. The chamber is a square with the side long: (a) and (b) 5 mm, high of the cavity is equal to 254  $\mu\text{m}$  before firing.

#### 4. Conclusion

The results show that the cold chemical lamination can be used successfully in the LTCC manufacturing process. The CCL permits to achieve strong bonding between the LTCC tapes in the multilayer structure. The close and open chambers can be made. The cold chemical lamination decreases a level of the chambers deformation. It allows on manufacturing very wide channels and chambers one layer high. It permits to fabricate sensors and others microsystems in the LTCC technology. The method is used to manufacture micromechanical sensor presented in.<sup>16,17</sup> In comparison with standard thermo-compression method the cold chemical lamination reduces deformations during the bonding process. It permits to achieve more precise design of the chambers. In comparison with cold low pressure lamination<sup>12,13</sup> there is no need of using any adhesive tapes to join the LTCC layers together. There is also no problem with delamination of the close chambers. The CCL process is quick and inexpensive.

The influence of the other solvents on the quality of the bonding are going to be analyzed, in the further works. The influence

of the solvents on the quality of the printed electronic components will be examined. The other types of the LTCC tapes will be used in further investigation. The solvent is going to be screen-printed in further researches. It is going to increase repeatability of the solvent film thickness. Moreover, the tapes after CCL will be pressure by rubber cylinder. This should increase the bonding strength.

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