

Novel cold chemical lamination bonding technique—A simple LTCC thermistor-based flow sensor

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Abstract

The LTCC technique enables fabrication of microfluidic devices. The structures consist of channels, chambers and screen-printed passives. The lamination is a quality-determining process in the manufacture of the fluidic modules. The commonly used bonding method is thermocompression. The tapes are joined together at high pressure (up to 30 MPa) and temperature (up to 80 °C) for 2–15 min. Although these parameters allow good LTCC module encapsulation, the quality of the chamber geometry is strongly affected by high pressure and temperature. The cold chemical lamination (CCL) technique presented in this paper, a solvent-based method, largely avoids these problems. A film of a special solvent is deposited on the green tape, and softens the surface. The tape layers are then stacked and compressed at low pressure, below 100 kPa, at room temperature. The fabrication of a simple LTCC thermistor-based flow sensor is presented here to compare both lamination methods. The test device consists of one buried thermistor screen printed on a bridge hanging in a gas/liquid channel. The basic sensor parameters (measurement range, working temperature, output signal, working pressure and measurement error) are analyzed.

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Lamination is a quality determining processing step in fabrication of LTCC multilayer devices for packaging,^{1–3} fluidic microsystems,^{4–6} and sensors.^{7,8} The common LTCC tape bonding method is thermo-compression. The tapes are joined at high temperature, up to 80 °C, and pressures, up to 30 MPa, for 2–15 min. The temperature softens the tapes, facilitating pressure bonding. This method has several advantages: thermo-compression gives good encapsulation of the final device, strong bonding, and allows the resulting structure to have more than 40 layers. However, there is one important problem: high pressure and temperature cause deformation of the manufactured structure. The problem can be reduced by using a fugitive phase (sacrificial material) intended to disappear during the firing process.^{8,9}

The first alternative method of bonding LTCC tapes was described by Roosen.^{10–12} The technique, called cold low pressure lamination (CLPL), used double-sided adhesive tape to

temporarily glue the green ceramic tapes. During the firing process, the adhesive tape melted and diffused into the LTCC tapes. The method has many advantages: the bond is made at room temperature and low pressure, about 5 MPa, thereby decreasing the deformation of the chambers and channels. However, it also has disadvantages: the increase of pressure in the chamber during firing destroys modules with closed chambers, and the laminated stack may crack during the firing process. The method is therefore best used for making open chambers and channels.

The solvent-base lamination was presented by Suppakarn.¹³ The alumina green tape was covered by a mixture of ethanol, toluene, and poly(propylene glycol) – PPG. The mixture film was applied to the tape surface and the next tape was added on the top. The procedure was repeated until the object was completed. Then, the complete stack was rolled by an alumina (0.45 kg) cylinder. Finally, the module was compressed at 0.5 kPa for 5 min. The method enabled strong bonding to be achieved. However, the solvent influence on passive components was not investigated, and the lamination quality of closed chambers was not presented.

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Another adhesive-based method was presented by Rocha.¹⁴ The layers were joined by adhesive liquid, and several different substances (e.g. natural honey) were analyzed. The LTCC tapes were covered by the organic liquid film, and then stacked together, gluing the layers with the liquid. The temporary gluing process was realized at very low pressure at room temperature, achieving good bonding without deformation of the chambers. Non-metallized, metallized tapes and chambers were successfully processed using this technique. However, the influence on the basic electrical properties of the screen-printed passives was not reported.

The next gluing-based method was presented by Gurauskis.^{15–18} A film of the gluing agent, 5 wt.% binder Mowilith DM765E distilled water dilution was applied, using a paint brush, on a top surface of a green ceramic piece. The gluing process was carried out at low pressure about 15 MPa at room temperature, achieving good bonding quality. The method is dedicated to water-based slurries. The influence of the gluing agent on the electrical properties of the screen-printed passives was not described.

Cold chemical lamination (CCL), a solvent-based lamination method, is presented in this paper. A film of solvent is applied on a tape surface using a paint brush. The solvent melts the surface. The next tapes are added on the top by the same method. Then the stack is compressed by pressures below 100 kPa at room temperature. The lamination is based on the diffusion process. The method has many advantages: the process is realized at low pressure and temperature, thereby decreasing the deformation of the chambers and channels. Moreover, the bonding is strong and no other materials are used. The solvent is vaporized completely and the method allows manufacturing of modules having large close chambers. A fabrication of a simple LTCC thermistor-based flow sensor is presented below for comparison of both lamination methods. The test device consists of one buried thermistor screen printed on a bridge hanging in a gas/liquid channel. The thermistor is used as a heater and a temperature sensor, which permits the measurement of gas and liquid flow velocity. DuPont LTCC tape 951AT (thickness 114 μm before firing process), DuPont NTC thermistor paste NT40 and a solvent agent (DuPont thinner 4553) were used in the manufacturing process. The basic sensor parameters (measurement range, working temperature, output signal, working pressure, measurement error) and the essential electrical thermistor features (thermistor constant B , sheet resistance R_{\square} , standard deviation of resistance and constant B) are determined. The bonding quality and chamber geometry are analyzed by a scanning electron microscope (SEM).

1. Experiments

1.1. Flow sensors design and manufacturing process

The metallization (DuPont 6146 PdAg paste) and thermistors (DuPont NT40 paste), with surface area 1.8 mm \times 0.3 mm (6 squares) were screen-printed on the LTCC green tape. The channel shapes were cut in tapes using the Nd-YAG laser (Aurel

NAVS 30 laser trimming and cutting system).¹⁹ The design of the sensor is presented in Fig. 1. The sensor consists of: four bottom part layers (Fig. 1(a)), two bottom gas channel part layers (Fig. 1(b)), one bottom bridge part layers with a thermistor (Fig. 1(c)), one top gas channel part layer (Fig. 1(d)), two top bridge part layers (Fig. 1(e)), and four top part layers (Fig. 1(f)).

Eight sensors were manufactured by the CCL and the thermo-compression methods. The thermo-compression process was carried out at 10 MPa pressure, temperature 70 °C for 10 min. In the CCL method, DuPont thinner 4553 was applied on the green tape surface using a paint brush. Both structures were sintered at 875 °C peak temperature, for 15 min. The total firing cycle was equal to 90 min.

The cross-section of the thermo-compressed flow sensor is presented in Fig. 2. The delaminations visible in Fig. 2 are caused by the process pressure and temperature being too low. However, the sagging rate of the bridge, an effect of the pressure and temperature being too high, is significant. The bonding quality and the bridge geometry may be improved by using sacrificial materials (fugitive phase) intended to disappear during a cofiring process,^{10,9} but an influence of the fugitive phase on the electrical properties of screen-printed passives has to be analyzed.

The cross-section of the CCL module is shown in Fig. 3. The bridge is not deformed and its sagging rate is very low. The lamination quality is at the same level as in the thermo-compressed modules. The wall of the channel geometry is weak, because of tapes displacement. A position of each layer may be corrected during the stacking process in the thermo-compression. The CCL tapes are bonded immediately during stacking. The problem may be reduced by using more stable locating tape pins.

The top view of the buried channels, compared with the match size, and the final encapsulated devices are presented in Fig. 4(a) and (b), respectively. The electric wires may be soldered or wire bonded to the screen-printed pads. The gas or liquid may be delivered to the structure through “Upchurch”, connections.

1.2. Thermistor properties

The repeatability of the sensor parameters strongly depends on the thermistor’s basic electrical properties such as sheet resistance at room temperature (R), $R=f(T)$ dependence, thermistor constant B , standard deviations (σ_B , σ_R) of the constant B and sheet resistance, variability coefficient (V_R , V_B) of the sheet resistance and constant B .

The standard deviation coefficients of the sheet resistance and constant B are given by Eqs. (1) and (2) (where: x_i sheet resistance or constant B of “ i ” thermistor, μ_R – average value of the thermistor sheet resistance, μ_B – average value of constant B , N – number of thermistors). The variability coefficients of the sheet resistance and constant B are given by Eqs. (3) and (4), respectively (where: σ_R , σ_B – standard deviation of the sheet resistance and constant B , respectively, R_A , B_A – average value of sheet resistance and constant B , respectively). The constant B is given by Eq. (4) (where: R_T – measured resistance, T – temperature of measured element in Kelvin degree, T_{ref} – reference

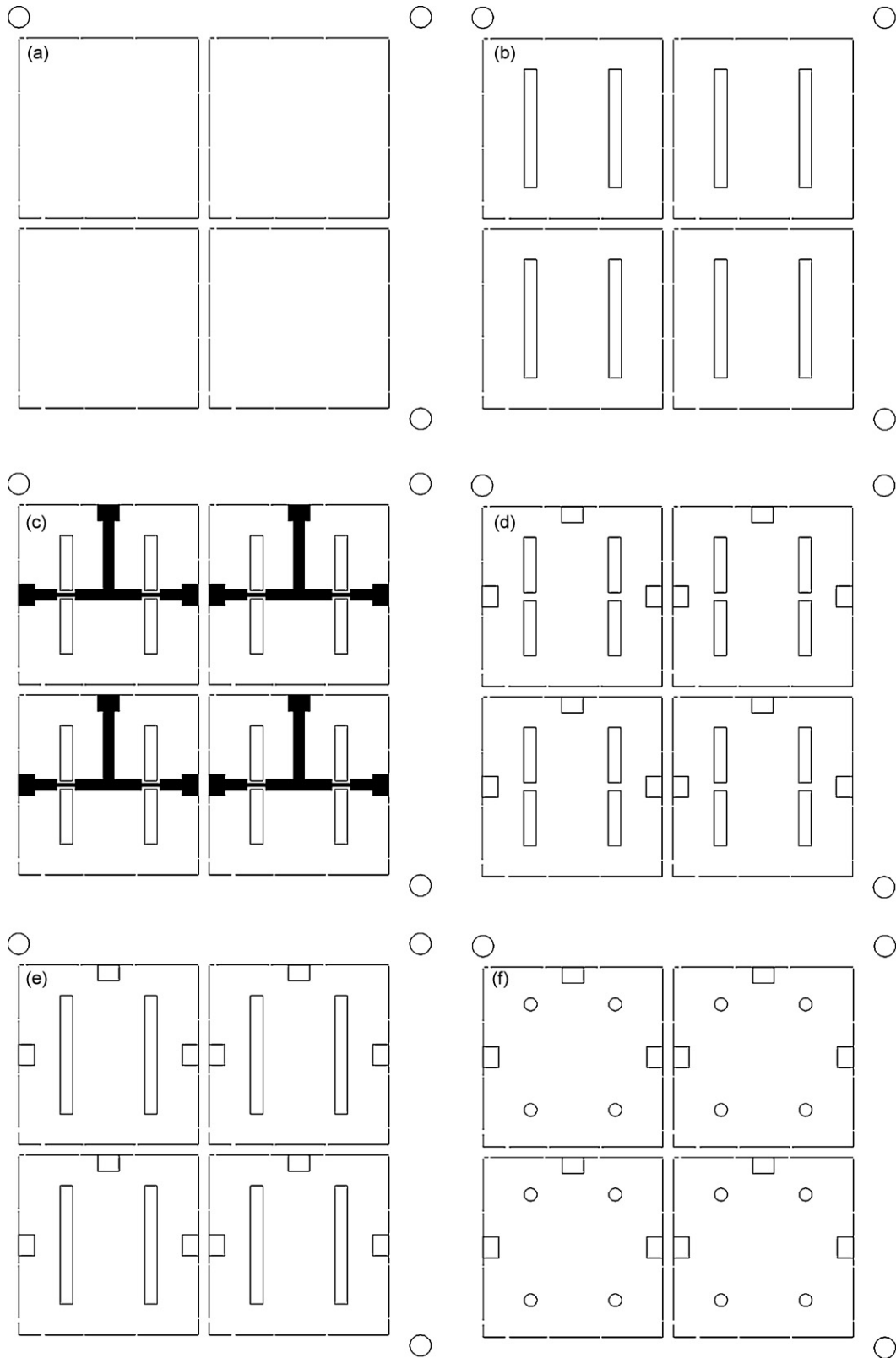


Fig. 1. Design of the sensor: (a) bottom part, (b) bottom gas channel part, (c) bottom bridge part with thermistors, (d) top bridge part, (e) top gas channel part, and (f) top part.

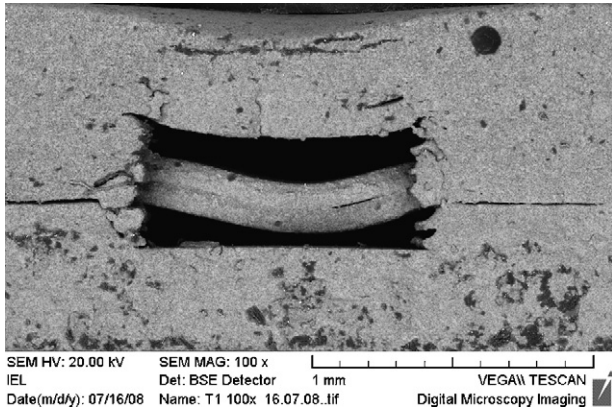


Fig. 2. SEM photo of the cross-section of the flow sensor with the LTCC tapes bonded by the thermo-compression method.

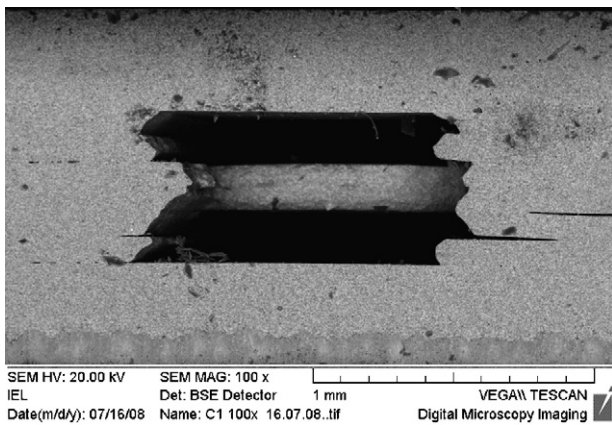


Fig. 3. SEM photo of the cross-section of the flow sensor with the LTCC tapes bonded by the CCL method.

temperature (298 K), R_{Tref} – resistance at reference temperature)

$$\sigma_R = \sqrt{\frac{\sum_{i=1}^N (x_i - \mu_R)^2}{N - 1}} \quad (1)$$

$$\sigma_B = \sqrt{\frac{\sum_{i=1}^N (x_i - \mu_B)^2}{N - 1}} \quad (2)$$

$$V_R = \frac{\sigma_R}{R_{A\Box}} \quad (3)$$

$$V_B = \frac{\sigma_B}{B_A} \quad (4)$$

$$B = \frac{\ln(R_T/R_{Tref})}{(1/T) - (1/T_{ref})} \quad (5)$$

Sheet resistance, thermistor constant B , standard deviation of sheet resistance and thermistor constant B , variability coefficient of constant B and sheet resistance for thermo-compressed and CCL modules are presented in Tables 1 and 2, respectively. The CCL affects the basic electrical properties of the thermistor-composition. Sheet resistance is about 30% and constant B is 4% higher in comparison with thermo-compressed components. The variability coefficients of resistance and constant B are 14% and 11% lower, respectively for the CCL method.

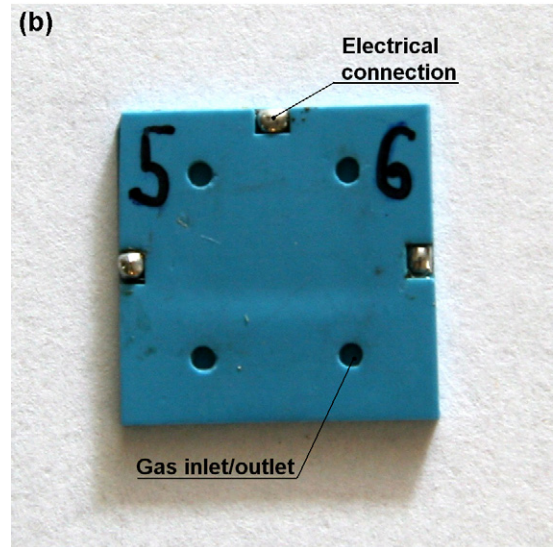
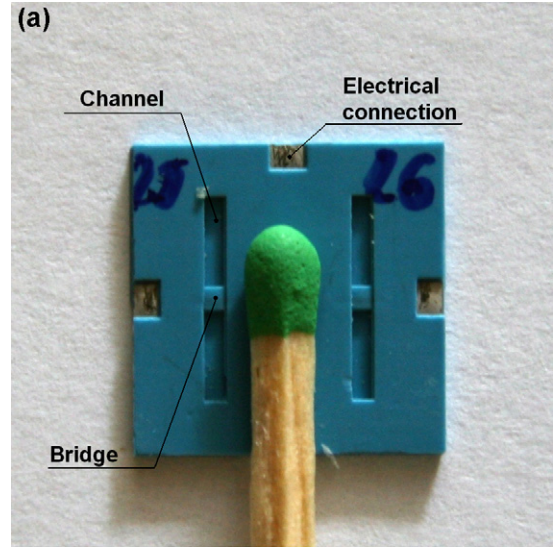


Fig. 4. Top view of the final sensor: (a) view of buried channel and (b) final structure.

Higher variability coefficients for thermo-compressed modules are caused by damage to the thermistors due to bridge deformation. The results show that the CCL method may be successfully applied in the fabrication process of the LTCC modules. Temperature characteristics of resistance for CCL and thermo-compressed modules are presented in Fig. 5. The CCL does not affect this parameter and the characteristics are linear in logarithmic scale.

Table 1

Basic electrical properties of the screen-printed thermistors on substrate bonded by thermo-compression lamination.

Parameter	Value
Sheet resistance, R_{\Box} [Q/□]	1656
Standard deviation of sheet resistance, σ_R [Ω]	757
Variability coefficient of sheet resistance, V_R [%]	46
Thermistor constant B [K]	1290
Standard deviation of constant B , σ_B [K]	261
Variability coefficient of the constant B , V_B [%]	20

Table 2

Basic electrical properties of the screen-printed thermistors on substrate bonded by thermo-compression lamination.

Parameter	Value
Sheet resistance, R_{\square} [Ω/\square]	2200
Standard deviation of sheet resistance, σ_R [Ω]	701
Variability coefficient of sheet resistance, V_R [%]	32
Thermistor constant B [K]	1340
Standard deviation of constant B , σ_B [K]	121
Variability coefficient of the constant B , V_B [%]	9

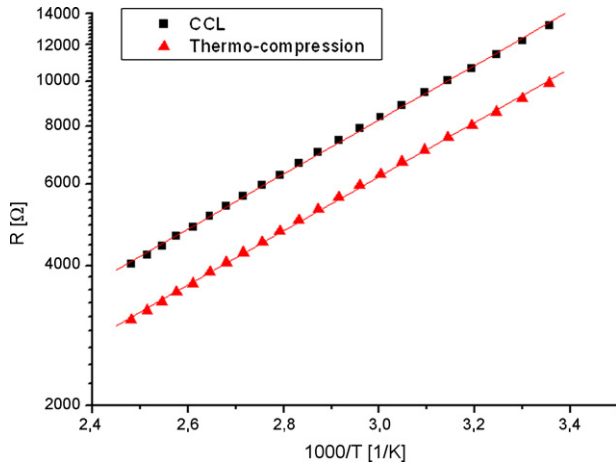


Fig. 5. Temperature characteristics of resistance for CCL and thermo-compressed modules.

Table 3

Basic parameters of the thermo-compressed sensor.

	Value
Working range	3–20 ml/s
Working pressure	<400 kPa
Max. pressure	<2 MPa
Output signal	0.5 V/ml
Max. standard deviation	3.2%
Average standard deviation	2%
Nonlinearity	3%
Working temperature	<40 °C

Table 4

Basic parameters of the CCL sensor.

	Value
Working range	3–20 ml/s
Working pressure	<400 kPa
Max. pressure	<2 MPa
Output signal	0.5 V/ml
Max. standard deviation	5.4%
Average standard deviation	2.6%
Nonlinearity	3%
Working temperature	<40 °C

1.3. Sensor response

The flow sensor response vs. flow velocity of thermo-compressed and CCL modules are presented in Fig. 6(a) and

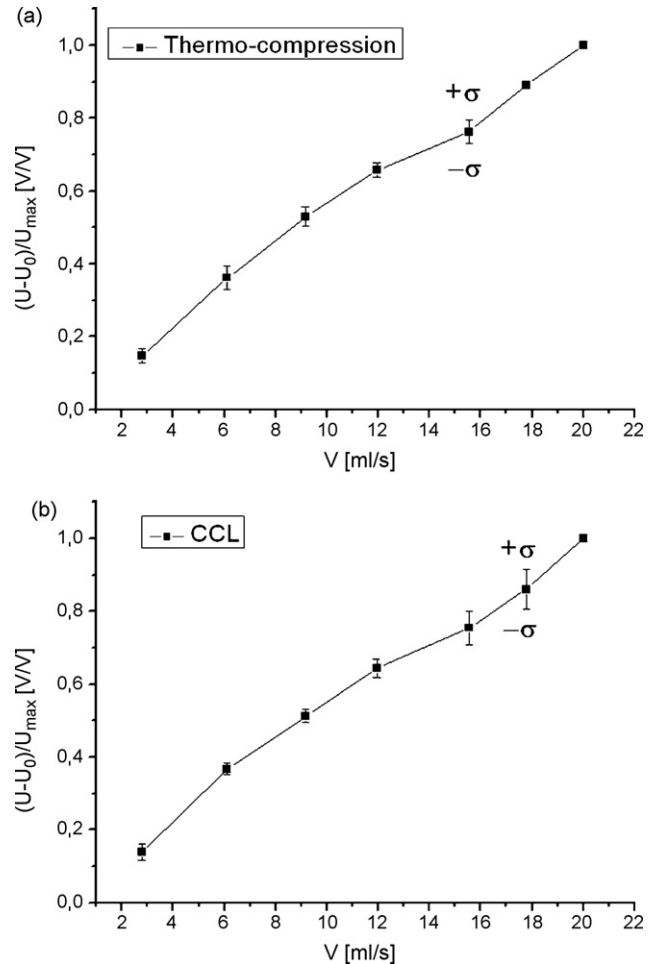


Fig. 6. Output voltage vs. flow velocity: (a) thermo-compressed module and (b) CCL module.

(b), respectively. Tables 3 and 4 show thermo-compressed and CCL sensor parameters, respectively. Both response characteristics and sensor parameters are very similar. Standard deviation is higher for CCL sensors. This is an affect of the tape displacement during lamination process. The effect can be decreased by using more stable locating tape pins.

2. Conclusion

The application of a new bonding technique in the fabrication process of LTCC sensors is presented. The influence of the chemical lamination on basic electrical properties of screen-printed thermistors, chamber geometry and bonding quality are described. Thermistor resistance and thermistor constant B are higher, while variability coefficient of resistance and constant B are lower for CCL modules in comparison with the thermo-compressed ones. The proper location of pins during stacking process in the CCL method is very important. Otherwise, the tapes will be displaced in the multilayer structure. The bonding quality and chamber geometry are very good in the CCL modules. The method gives great possibilities for LTCC technology. The cold chemical lamination may be applied in manufacturing process of the LTCC devices with screen-printed passive

components, gas/liquid channel and microreactor chamber. Future work will concentrate on other solvents, screen-printed on the LTCC tapes and the bonding ability of other commercially available tapes. Moreover, stable locating tape pins will be applied in the lamination process, to achieve better quality channel walls. be applied in manufacturing process of the LTCC devices with screen-printed passive components, gas/liquid channel and microreactor chamber. Future work will concentrate on other solvents, screen-printed on the LTCC tapes and the bonding ability of other commercially available tapes. Moreover, stable locating tape pins will be applied in the lamination process, to achieve better quality channel walls.

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