NOVEL BONDING METHOD OF LOW TEMPERATURE COFIRED CERAMIC TAPES*

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The lamination determines quality and geometry of the fabricated ceramic microsystems. The thermo-compression method is commonly used. In this technique the Low Temperature Co-Fired Ceramics (LTCC) tapes are joined together at high pressure and temperature. Cold Chemical Lamination (CCL) is presented in the paper. It is a solvent-base method used for green ceramic tape bonding. The tapes are covered by film of the special liquid. Then the ceramics are put in a stack and laminated at low pressure below 0.5 MPa. The lamination quality is investigated. The cross-section of the close chambers is examined by optical and Scanning Electron Microscopy. The solvent influences the basic electrical properties of the thermistors composition (ESL NTC-2114), sheet resistance at a room temperature, R=f(T) dependence, B constant and a long term stability is analysed. The lead free ESL 41020 tape is used during the experiment. The thermistor composition is screen printed on the LTCC substrate. The R=f(T) dependence is measured by the Agilent 34970A data acquisition unit. Long-term stability is investigated by annealing at 150°C for 200 h.

Key words: LTCC, thermistor, lamination

1. INTRODUCTION

The lamination is the most important technological step during manufacturing process of the LTCC devices [1-6]. The thermo-compression is well known method.

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The tapes are joined at high pressure and temperature up to 20 MPa and 80°C, respectively. The method permits to achieve strong bonding. However, during the process the chambers and channels are deformed. The problem can be reduced by using the sacrificial materials [7-8] which evaporate completely during the co-firing process.

Alternative method of the green tape bonding was presented by Roosen [9-11]. It was named Cold Low Pressure Lamination (CLPL). The LTCC tapes were joined by special two side adhesive tapes. However, there was no possibility of manufacturing close chambers. The influence of the method on the electrical properties of screen printed passives was not investigated.

The solvent-base method was presented by Suppakarn [12]. The method was used to join alumina and PZT composition. The lamination was not used for bonding LTCC green tapes. There was no investigation of the solvents influence on the electrical parameters of the passive components.

Another adhesive-based method was presented by Rocha [13]. The organic liquid film was applied on the LTCC green tape surface, by spinning or screen-printing method. Then the tapes were stacked together and glued by the adhesive. The lamination process was carried out at low pressure at room temperature. Good bonding quality and chambers without any deformations and modules with metalized tapes were fabricated by the method. Several different types of adhesive was investigated e.g. natural honey. However, the influence on the basic electrical properties of the screen-printed passives was not described.

Another adhesive-base lamination method of green tapes was presented by Burdon et al. [14]. the film of special room-temperature adhesive agent (pressure-sensitive adhesives, e.g. the Flexcryl 1653) was applied on the green tape surface by the standard coating techniques. The spraying and spin-coating method were recommended. The tapes were joined by room temperature adhesive penetrating into the surfaces of the bonded layers. Laminated structure were compressed by 700 kPa at room temperature. The method enables fabrication of modules which consist of passive components, open and close chambers. However, the influence of the adhesive agent on the basic electrical properties of the screen-printed passives was not described.

In the paper is presented an alternative bonding method, called Cold Chemical Lamination. The quality of the bonded LTCC tapes is presented in the paper. The solvent is screen printed on the LTCC bonded tapes. Quality of the lamination and the solvent influence on the basic thermistor electrical properties are investigated in the paper.

2. LAMINATION QUALITY

The test structure consists of ten ESL 41020 tapes. The bottom, the middle and the top part of the structure are respectively 4, 2 and 4 tapes high. The thickness of one tape is approximately 115 μ m. The test close chambers are fabricated in two types. The cavity top surface is equal to 5x5 mm² in the first and to 2x2 mm² in the second. The chamber is two layers high. The DuPont 4553 thinner is applied on the top surface of each tape by screen printing method (Screen 500 mesh). The tapes are put in one stack and then pressed by low force below 0.5 MPa at room temperature. The co-firing process determines quality of the bonding. The burning cycle has to be long enough to ensure evaporation of the solvent before sintering of the LTCC modules. The gas bubbles are entrapped between tapes if the process is too short. The author recommends the cofiring cycle presented in Tab. 1. The ESL does not describe the cofiring process cycle. The company recommends only the peak temperature at 875°C, and firing at peak temperature for 10 minutes.

Table 1. Cofiring process parameters.Tabela 1. Parametry procesu spiekania.

Time [h]	3	3	3	0.25
Temperature [°C]	20-480	480	480-870	870

Fig. 1. presents optical image of a cross-section of the structures laminated by Cold Chemical Lamination. Chambers with area $2x2 \text{ mm}^2$ and $5x5 \text{ mm}^2$ are shown in Fig. 1 (a) and (b), respectively. The cavity geometry is very good for both structures. The Scanning Electron Microscope photo of test structures with the cavity area $2x2 \text{ mm}^2$ and with area $5x5 \text{ mm}^2$ are presented in Fig. 2 and 3, respectively. Images of a whole smaller chamber cross-section and its right part are shown in Fig. 2 (a) and (b). The bonding quality is very good, borders between layers can not be observed, the module encapsulation is good. The walls are smooth and the tapes displacement can not be observed. Photos of left part, upper left corner and right part of the $5x5mm^2$ area chambers are shown in Fig. 3 (a), (b) and (c), respectively. Once more, lamination quality is very good and the borders between layers can not be observed. Cavities are not deformed. The walls smoothness is acceptable. The Cold Chemical Lamination may be used for joining ESL 41020 tapes in one hermetic module. The good quality close chambers can be fabricated.



Fig. 1. Optical microscope image: (a) size of the chamber 2 x 2 mm^2 ; (b) size of the chamber 5 x 5 mm^2 .

Rys. 1. Zdjęcie wykonane za pomocą mikroskopu optycznego: (a) wielkość komory 2 x 2 mm²; (b) wielkość komory 5 x 5 mm².



Fig. 2. Scanning Electron Microscope image of the tape bonding quality of the chamber area 2x2 mm²: a) lower zoom; b) larger zoom.

Rys. 2. Zdjęcie wykonane za pomocą mikroskopu elektronowego komory 2 x 2 mm²: (a) mniejsze powiększenie; (b) większe powiększenie.

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Fig. 3. Scanning Electron Microscope image of the tape bonding quality of the chamber area $5x5 \text{ mm}^2$: a) left part, b) left upper corner, c) right part.

Rys. 3. Zdjęcie wykonane za pomocą skaningowego mikroskopu elektronowego komory $5 \ge 5 m^2$: (a) lewa strona; (b) lewy górny róg; (c) prawa strona.

Chemical elements spectrum of the whole cross-section and the one point on the border between layers are presented in Fig. 4 (a) and (b), respectively. The amount of aluminum is decreased on the border between the layers. It is believed that the solvent creates some sort of inter layer between the tapes after sintering process. However, it do not affect the bonding quality. Further investigation of this phenomena is necessary.



Fig. 4. Chemical elements spectrum of the LTCC module: (a) whole cross-section area; (b) from border between bonded tapes.

Rys. 4. Sektrum pierwiastkowe modulu LTCC: (a) całej powierzchni przekroju poprzecznego; (b) z punktu na granicy dwóch warstw.

3. INFLUENCE OF THE SOLVENT ON BASIC ELECTRICAL PROPERTIES

The influence of the solvent on the basic electrical properties is presented in Tab. 2. The thick-film paste is screen printed on the green tape substrates lami-

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nated with the CCL method. The sheet resistance of the surface components is approximately 25% higher for surface components. The thermistor constant B and the variability coefficient of resistance are also higher in this case. The value of the constant B has good repeatability and therefore the buried passives can be used in the sensor application.

Table 2. Basic electrical properties of the thermistor composition ESL NTC-2114 made onthe lead free ESL 41020 tape.

Tabela 2. Parametry elektryczne pasty termistorowej ESL NTC-2114 nadrukowanej na podłożu ESL 41020.

Position	Sheet resistance $R_{\Box} [\Omega/\Box]$	Standard deviation of sheet resistance $\sigma_{R} [\Omega/\Box]$	Variability coefficient of sheet resistance $V_R[\%]$	Ther- mistor con- stant, B [K]	Standard deviation of constant B $\sigma_{\rm B}$ [K]	Variabi- lity coef- ficient of constant B V _B [%]
Surface CCL	24200	5089	21	2213	96	4.3
Buried CCL	16400	4062	25	1950	49	3
Catalogue Parameters*	10000			2125		

* on alumina substrate

The R = f(T) dependences are shown in Fig. 5 (a). The thermistor constant B is lower for buried components. It is caused by higher interaction between thermistor composition and DuPont 4553 thinner. Both characteristics are linear. The long-term stability of the ESL NTC-2114 composition is investigated by annealing at 150°C for 200 h. The results are shown in Fig. 5 (b). The buried components exhibit better long term stability (resistance changes less than 5%). The surface thermistors are not so stable and the resistance changes are up to 25%. The ESL recommends stabilization of the resistance by annealing for 16 h at 150°C. The buried components after this time exhibit resistance changes below 2%. However, the surface passives are not stable after annealing.

The results can be compared with results in [15]. In the article an influence of the different thermo-compressed substrates and position of the passives on the basic electrical properties are analysed. The CCL method increases sheet resistance for the surface and decrease for the buried components. The Cold Chemical Lamination decreases the thermistor constant B for buried and do not affect the parameter B of the surface passives. The resistance variability coefficient is better in the structures laminated with thermo-compressed method. The CCL do not affect the variability

coefficient of the constant B. The Cold Chemical Lamination do not affect the long-term stability of the buried components. The surface passives in the CCL structures has better stability than in thermo-compressed modules.



Fig. 5. (a) The temperature characteristics of resistance for ESL NTC-2114 thermistor composition, (b) long-term stability of ESL NTC-2114 thermistor composition.
Rys. 5. (a) Zależność rezystancji od temperatury dla termistorów NTC ESL NTC-2114; (b) stabilność długoterminowa dla termistorów NTC ESL NTC-2114.

4. CONCLUSION

New solvent-base lamination technique is presented in the paper. The Cold Chemical Lamination is successfully used to manufacture chambers with area up to $5x5 \text{ mm}^2$ and 2 tapes high with good bonding quality of green layers, without any delaminations and deformation of chambers area up to $5x5 \text{ mm}^2$. Moreover, it was discovered that on the border between tapes an amount of aluminum is decreased in comparison with rest of the structure.

The method also can be used to fabricate modules with screen printed passives. The influence of the DP thinner 4553 on the basic thermistor electric properties is analyzed. The long-term stability is good in case of buried components. The variability coefficient of the thermistor constant B is below 3%. This permits to use the buried one for manufacturing thermistor based sensors. The buried configuration is recommended.

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NOWA METODA ŁĄCZENIA WARSTW NISKOTEMPERATURO-WEJ CERAMIKI WSPÓŁWYPALANEJ

Jakość połączenia warstw folii niskotemperaturowej ceramiki współwypalanej (LTCC) oraz geometria komór silnie zależą od procesu laminacji. Obecnie powszechnie stosowaną metodą do produkcji ceramicznych układów wielowarstwowych jest laminacja termokompresyjna. Przy jej zastosowaniu warstwy są łączone w wysokiej temperaturze i ciśnieniu. W artykule przedstawiono alternatywną metodę łączenia folii niskotemperaturowej ceramiki. Warstwy są łączone za pomocą rozpuszczalników, które zmiękczają powierzchnie łączonych folii. W kolejnym etapie warstwy są ze sobą wstępnie łączone i dociskane niewielkim ciśnieniem (poniżej 0,5 MPa). W artykule przedstawiono jakość połączeń uzyskanych za pomocą nowej metody, jak również wpływ rozpuszczalnika na parametry elektryczne grubowarstwowych elementów biernych.

Słowa kluczowe: LTCC, laminacja, termistor