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Investigation of interactions between co-fired LTCC components

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Abstract

In spite of many advantages it offers for micro-electronic packaging, the low temperature co-fired ceramic (LTCC) technology has yet certain issues to be solved. Physical and chemical reactions, for instance, within and between the passive electronic components (thick film conductors, resistors—TFR) and the LTCC tapes (green sheets) during processing are one of the major challenges of this technology. This study aims to better understand and control the nature of such interactions, which have a direct effect on properties. The work is conducted on PTC (positive temperature coefficient) resistors, which are screen printed on (co-fired) and in (buried) LTCC sheets and fired at various temperatures. The final properties such as temperature coefficient of resistance (TCR) and sheet resistance are evaluated in terms of processing parameters using scanning electron microscopy (SEM), dilatometry and electro dispersive X-ray (EDXS) analysis as characterization tools. The results show that the TFR properties of buried samples deviate more strongly from the expected values compared to those of co-fired ones. It is primarily related to the destroyed conductor and resistor lines, which is closely related to the entrapped gases in buried structures as a result of organic burnout of printed inks.

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1. Introduction

Low temperature co-fired ceramic (LTCC) technology is recently addressed to be a key approach for smart packaging.^{1–5} Although its use initiated in the telecommunication filed due to the excellent dielectric properties of the LTCC tapes, its application areas have diversified in the recent years.^{6–9} Ease of machinability of tapes for 3-D structuration, high production volume and reliability at low processing costs, hermeticity of the circuits have attracted the interest of sensor and mesosystem technologies rapidly. Moreover, integration of passive electronic components as screen printable pastes into the tapes and low firing temperatures permitting the use of low resistive terminations have made this technology indispensable for many applications. It is based on LTCC tapes, which sinter below 950 °C and provide a medium with electrical and mechanical functions for realization of desired

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products. However, the integration of components raises several potentially problematic issues, both physical and chemical. The former arise mainly from the differential shrinkage mismatch of the components and the latter from the chemical interactions of the components during firing. In either case, the extent of these effects must be controlled in order to guarantee high reliability. This work aims to better understand the processing-property relationship of the selected TFR's fired on LTCC tapes.

The effects of processing method (co-fired on/in the tapes) and the firing temperature on the thick film conductors, resistors (TFR) properties (temperature coefficient of resistance (TCR) and sheet resistance) are studied and the results are interpreted using scanning electron microscopy (SEM), dilatometry and compositional analysis.

2. Experimental procedure

The LTCC tape we chose during this study was DuPont (DP) 951-AX^{10,11} which, according to our experience, has well-controlled shrinkage behaviour. On the other hand we

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The layout	Number of resistors	Length	Width
	5	1.5	1.5
	1	0.3	1.5
	1	0.6	1.5
2.2	1	1	1.5
	1	5	1.5

Fig. 1. The layout for test patterns with description of resistor specifications.

used Au-based compositions as terminations (ESL 8837 and DP 5744) and ruthenium-based PTC resistor ink (ESL 2612 with TCR of 2400 ppm/K and sheet resistance of $100 \ \Omega/\Box$ at 25 °C) due to the reduced deformation obtained on the co-fired tapes by these pastes compared to other systems available at our laboratory.

We applied the conductor-resistor combinations in two variants. In the first one, the pastes were screen-printed on the LTCC sheets and co-fired, whereas in the second one the printed layer was laminated with an additional layer to make a buried structure. Although they were both co-fired, from here on the first set of samples will be referred to as co-fired and the second set as buried for convenience. Screen-printing was made according to the layout shown in Fig. 1, where the number and the geometry of the resistors are also summarized. After drying the pastes in air and then in a lab oven at 120 °C, the structures were fired according to a two-step firing profile. They were heated up to 440 °C with a heating rate of 5 °C/min (same rate is applied after the dwell time up to the peak temperature) and kept at that temperature for two hours for organic burnout. This was followed by peak firing temperature of 850, 875 and 900 °C for each structure consecutively for 25 min to check the effect of firing temperature on properties. Finally, the samples were cooled down to room temperature by a rate of 10 °C/min. For each conductor-resistor combination, 4 samples were fired. In order to obtain sufficient statistical interpretation, the resistors with dimensions of 1.5 mm:1.5 mm were selected since each test pattern has five times of this resistor geometry.

The resistor values were measured three times at 30, 65 and 100 $^{\circ}$ C between consecutive pad pairs (Fig. 1), while the temperature was recorded using a Pt-1000 PTC resistor. The measurements were made by Keithley 2000 multimeters and Keithley 7000 scanner using four-wire method. TCR value was calculated based on the resistances at 30 and 100 $^{\circ}$ C according to the following relation.

$$TCR = \frac{10^6 (R_{100} - R_{30})}{R_{30}(T_{100} - T_{30})}$$
(1)

where R is the resistance at a certain temperature, T. The standard deviation (S.D.), on the other hand, was calculated by

S.D. =
$$\sqrt{\frac{n \sum y^2 - (\sum y)}{n(n-1)}}$$
 (2)

where n is the number of values y (sheet resistance or TCR).

3. Results and discussions

3.1. TCR and standard deviation

TCR and standard deviation data is presented in Fig. 2a and b, each figure corresponding to different component pairs and processing variables. Results demonstrate that the co-fired samples have TCR values, which are closer to the producer specifications, with much narrower deviation compared to the buried ones. Bearing in mind various effects leading to this result, we started our investigation with the most directly observable one. It was the deformation at the conductor-resistor intersecting regions (contacts) in the form of blisters, which formed mechanical distortion on the substrate surface. Although the differential shrinkage is well known for leading to such results it was more dominant in the co-fired samples and not in the buried ones, where the exact origin of deformation is yet being studied. Our current efforts for a better understanding of this effect mainly focus on the temperature range at which LTCC is still porous whilst the organics are completely burnt out. Thus, deformation, which takes place up



Fig. 2. . TCR and standard deviation at different firing conditions for. (a) ESL 8837-ESL 2612 (left) and (b) DP 5744-ESL 2612.



Fig. 3. SEM images of buried ESL 8837–ESL 2612 component fired at 850 °C (left). Deformation of the structure due to delamination/entrapped gases after organic burn-out. The image on the right hand side shows the discontinuous conductor line due to extremely fine thickness not bearing migrating material.

to this temperature, can be linked to delamination localized around the tape-paste interface and/or accelerated organics decomposition and gas entrapment due to fast heating rate. However, lack of deformation at this stage indicates that this effect takes place at a higher temperature, which can arise from the chemical reactions within or between components (e.g. reduction of certain elements resulting in blistering, etc. ...). Fig. 3 shows the SEM analysis of buried components, with excessive deformation.

On the other hand, the major source of deformation on the co-fired samples was rather different. It was observed that DP 5744 ink created more deformation compared to other composition, which was mainly due to two reasons. The first one was related to the expansion of the paste, which started at 700 °C and continued up to 850 °C (Fig. 4), deforming the non-sintered-tape during the same temperature range. The second factor, which contributed to the increased deformation, was related to the nature of the pastes. ESL 8837 ink having an organo metallic character resulted in a very thin-fired thickness



Fig. 4. Dilatometry analysis of the tape and the conductors used. DP 5744–Au ink starts expansion at 700 $^{\circ}$ C, which is around the onset temperature for the tape shrinkage.

of around 1 μ m, whereas DP 5744 reached a thickness of 6–7 μ m (Fig. 5). It is clearly observed that the paste having higher thickness deformed the tape more than the finer one. In the buried samples, deformation is expected to be much less because the film resides on the neutral plane of the structure and the overall thickness is twice as large.

In addition to these effects, influence of the chemical contents of the components on properties was also studied. Electro dispersive X-ray (EDXS) analysis was performed following X-ray fluorescence (XRF) and XRD analysis, in order to find the elemental compositions. The major chemical interaction, which resulted in highly deviated TCR values of buried samples, was expected to be arising from the interaction of the glass constituent of the resistor with that of the substrate from both sides. In order to see the extents of this effect, microcompositional analysis was carried out on the glass content of both components in a buried structure by EDXS (Fig. 6). It was found that both the tape and the resistor had common elements such as Pb, Al, Ca, which show a gradient at different locations. This reminds a possible change of stoichiometry in the expected phases, resulting in differences in properties. The exact nature and consequences of such an interaction is the part of an on-going study and will be explained in a future article.

3.2. Sheet resistance and standard deviation

Sheet resistance of resistors with $1.5 \text{ mm} \times 1.5 \text{ mm}$ geometry was measured at 30 °C. The results are presented in Fig. 7a and b. As it can be seen from the graphs, buried samples showed a higher sheet resistance value with very high standard deviation compared to those of co-fired ones. Among the buried samples, the ones with DP 5744 paste were found to remain closer to the sheet resistance value specified by the producer than the ones with ESL 8837. The origin of this difference is believed to be due to the thickness of the conductor paths: as seen in Fig. 3, ESL 8837 conductor tends to become discontinuous and divide into islands. In buried structures, this is favoured by the interaction with the LTCC glass phase from both sides and also by the mechanical stresses induced by the lamination process.



Fig. 5. Comparison of conductor line thickness of ESL 8837 (top and down left) and DP 5744 at 875 $^{\circ}$ C (top and down right) and its consequence on the substrate deformation. Substrate is curled in the right bottom image (DP 5744).



Fig. 6. EDXS Analysis performed on the buried structure. The points of measurement can be seen on the image (right) and the corresponding elemental data for the selected elements in the glass phases is presented on the chart (left).



Fig. 7. Sheet resistance and standard deviation at different firing conditions for. (a) ESL 8837–ESL 2612 (left) and (b) DP 5744–ESL 2612.

4. Conclusion

Co-firing of LTCC tape with commercial thick film inks at different processing conditions and its effects on properties were studied. It is found that buried samples have increased deviation and unreliable TCR and sheet resistance values compared to the co-fired ones. This is linked to deformation along the contacts, which is found to be due to the gases entrapped in LTCC as a result of organics burn-out. Electron microscopy analysis shows that the contact points in these samples were destroyed and partially detached from the substrate. Moreover, the introduction of the second LTCC layer acted as an extra diffusion path, which had an influence on properties chemically, in addition to the composition gradient at the interfaces due to certain elements in the glass constituent of substrate and the resistor.

On the other hand the co-fired samples show properties closer to producer specification with lower deviations. It is observed that the thickness of the fired conductor line plays an important role in the extent of deformation in addition to differential shrinkage. This deformation is less in buried structures since the film reside on the neutral plane of the structure and the thickness is twice as large.

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