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New lamination technique to join ceramic green tapes for the manufacturing of multilayer devices

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Abstract

Lamination is a quality determining processing step in the manufacturing of ceramic multilayer devices. The common method to join stacked ceramic green tapes is thermo-compression. The binder phases of two adjacent green tapes are joined together at elevated temperatures and pressures. This causes mass flow, which prevents the production of complex 3D structures or fine patterns. The new lamination technique is based on a temporary gluing step. The green tapes are stuck together by an adhesive tape at room temperature under very low pressures. Even though the tapes are separated by the adhesive tape, the ceramic green tapes are joined homogeneously after sintering. In contrast to the thermo-compression method, the new cold low pressure lamination technique reduces deformations, and has a high potential for the lamination of undercut, complex 3D structures with small lines and spaces of the metallization. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Lamination is an important processing step in ceramics multilayer manufacturing of packages, high integrated multilayer circuits (LTCC, low temperature co-fired ceramics), capacitors, actuators, and gas sensors.[1–4] Green tapes are the basic product for these multilayer devices. The tapes contain ceramic powders, which are dispersed in a matrix of a binder/plasticizer mixture which give the ceramic green tapes flexibility for subsequent multilayer processing. The tapes are punched, and for most functional applications metallized by screen printing. The tapes are stacked and laminated. After binder burnout and cofiring the final ceramic product is obtained. In multilayer manufacturing, lamination must result in homogeneous junctions between the pure green tapes or between green tapes and metallized areas. After sintering the original interfaces should not be visible anymore. The interface between metallization and ceramic must be cavity-free. The quality of the laminated interface, and its stability during the binder burnout and sintering process strongly determine the quality of the final product.

1.1. Thermo-compresssion

The common method used today to join stacked metallized ceramic green tapes is thermo-compression. This technique is based on joining the binder phases of two adjacent green tapes together at elevated temperatures up to 80 °C and pressures up to 30 MPa.[5] A holding time at peak temperature of typically 3–10 min is needed to reach a homogeneous temperature profile in the entire stack. During this process, a thorough interpenetration of the powder particles in both unsintered tapes must be achieved. This requires specific green tape microstructures with regard to the ratio of powder, binder, and porosity.[3,6] During this process mass flow occurs, which is caused by the applied pressure and temperature and by the porosity of the green tapes.

In the case of printed patterns like circuits or electrodes, the metallized areas must be embedded in the green tape during lamination. If the flow rate of the binder system and the porosity of the tapes are optimised the metallized parts are completely pressed into the green tapes. Hard binders, low porosity of the tapes, or unfavourable lamination conditions can cause small cavities at the interface tape/metallization or other density gradients. These defects can be the origin of delaminations and cracks during binder burnout and sintering.[7] In the manufacturing of capacitors and piezo actuators a controlled mass flow of the entire stack volume is desired to achieve an additional densification of the stacked and metallized tapes, thus density gradients are eliminated. In the manufacturing of other devices like high

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integrated circuits, packages or gas sensors the mass flow can cause changes in the desired position of vias, circuit lines or electrode patterns in different layers relative to each other. For this reason the thermo-compression method cannot be used to produce complex 3D structures or fine patterns. The disadvantage of the thermo-compression method can be summerized as follows:

- discontinuous process at elevated temperatures and pressures;
- waiting period to reach homogeneous T-profile in the stack;
- uncontrolled mass flow, which prevents the formation of fine structures or of spatially complex three dimensional structures;
- inhomogeneous pressure distribution in complex shapes, which results in delaminations;
- limited to specific microstructures of the green tape;
- in the case of green tapes from water based slurries, the different binder systems used are less suitable for the thermo-compression.

1.2. Cold low pressure lamination

Cold low pressure lamination (CLPL) is a new lamination technique which was developed in Erlangen to overcome the mentioned disadvantages.[8,9] This technique is based on a temporary gluing step. The green tapes are stuck together by a double sided adhesive tape at room temperature under very low pressures. Even though the tapes are separated by the adhesive tape and interpenetration of the particles cannot occur, the tapes are joined homogeneously after sintering. In contrast to the thermo-compression method, the new cold low pressure lamination technique reduces deformations, and has a high potential for the lamination of undercut, complex 3D structures with small lines and spaces of the metallization. Advantages of this new technique are:

- carried out at room temperature at low pressures;
- no waiting period to reach homogeneous T-profile in the stack;
- suitable for continuous manufacturing processes;
- no uncontrolled mass flow, suitable to build up fine structures or spatially complex three dimensional structures;
- no inhomogeneous pressure distribution in complex shapes;
- works also in the case of green tapes from water based binder systems;
- low investment costs;
- alternative technique for growing market.

2. Experimental

Two green tapes were used for lamination experiments using the technique of cold low pressure lamination. One green tape was tape cast from a solvent based Al_2O_3 slurry and dried.[2] The green tape had a thickness of 725 µm and consists of 92 mass% Al_2O_3 (mean particle size: 3.5 µm) and 8 mass% organic additives: polyvinyl butyral was used as a binder and dibutylphthalate as a plasticizer. The other green tape was a commercial LTCC tape (Green Tape 951, DuPont, USA) with a thickness of 165 µm. It consists of 89 mass% inorganics and 11 mass% organic additives, based on PMMA, and was metallized with different patterns using an Ag–Pd metal paste.

To determine the pore size of the green tapes, the tapes were heated up to a temperature which is 50 °C below the temperature of shrinkage onset. The medium pore diameter of the Al_2O_3 tape was 0.65 µm and of the LTCC tape 0.55 µm (Hg-Porosimeter 2000, Carlo Erba Instr., Italy), the alumina tape had a porosity of 40 vol.%, the LTCC tape 43 vol.%. The tapes were cut into 30 mm square pieces.

For gluing, two double-sided adhesive tapes (Beiersdorf AG, Hamburg, Germany) were used. The adhesive layers of both tapes were based on polyacrylate. In one case the intermediate carrier of the two adhesive layers was a polyester film of 12 µm thickness. In the other case the intermediate carrier consists of a polyimide film of 25 µm thickness. Thermogravimetric analysis (Du Pont Instr., USA) and the dynamic viscosity (Rheometer CSM 50, Bohlin Instr., Sweden, cone and plate geometry, oscillation mode of 1 Hz) of the pure carrier films, of the pure adhesive component and of the binder system of the green tapes were determined.

Laminates were fabricated by joining three to four pieces of green tape at room temperature by means of double sided adhesive tapes (Fig. 1). The tapes were non-metallized Al_2O_3 tapes or metallized LTCC tapes, respectively. The laminates were burnt out carefully in air up to 600°C, followed by sintering at 1650°C (2 h) in the case of the Al_2O_3 tapes and at 870°C (20 min) for the LTCC tapes. At 400°C some of the Al_2O_3 laminates were removed from the furnace. The fracture surface of these prefired laminates as well as the cross-section of all sintered laminates were investigated by scanning electron microscopy.

3. Results

The fracture surfaces of the prefired Al_2O_3 laminates demonstrate that the thickness of the adhesive tape

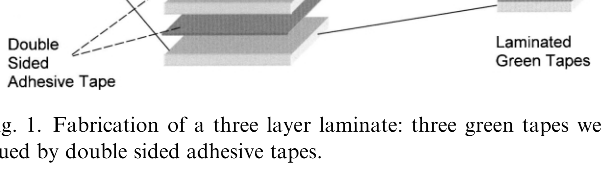

Fig. 1. Fabrication of a three layer laminate: three green tapes were glued by double sided adhesive tapes.

between two green tapes decreases with increasing temperature. Fig. 2 shows the cross-section of the laminate from the polyester based Al_2O_3 tape after the removal from the kiln at 400°C: a polymer layer of 5 μm thickness is left. At slightly higher temperatures this polymer layer has disappeared completely. After sintering the three ceramic green tapes were transferred to a homogeneous body in which interfacial layers are not visible anymore [Fig. 3(a)].

In contrast to this, prefired Al_2O_3 laminates from the polyimide based adhesive tape exhibit no adhesive strength. After sintering no joining was observed, the three original layers were completely separated from each other [Fig. 3(b)]. This demonstrates that the joining mechanism is controlled by the chemistry of the double sided adhesive tape and by its changes of the consistency and rheological behaviour during the heat treatment. The measurement of the viscosity of the carrier film from the adhesive tape and of the pure binder system of the Al_2O_3 green tapes in dependence on temperature showed strong differences (Fig. 4). The polyester based film shows a very low viscosity polymer melt at much higher temperatures than the pure binder system of the Al_2O_3 green tape. This polymer melt at the interface of the porous microstructure of the green tapes causes capillary forces. The polyimide based tape decomposes without melting, thus no viscosity could be determined, and no capillary forces can occur.

In case of the metallized LTCC tape which was joint by the polyester based adhesive tape an excellent joining of the ceramic/ceramic and also of the metal/ceramic interface is obtained, too (Fig. 5).

4. Discussion

The joining mechanism is due to a low viscosity melt derived from the polyester carrier film of the adhesive tape, which occurs at higher temperatures of around 350°C during the binder burnout process. This liquid phase which occurs at the interface of the porous microstructure of the green tapes causes capillary forces. These forces result in an approach of the ceramic layers towards each other, and finally in a re-arrangement of the particles in the interface of the tapes. Thermo-

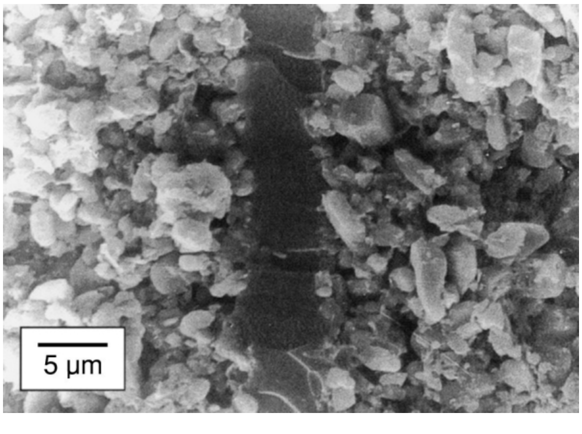

Fig. 2. Scanning electron microscopy images of the fracture surface from a prefired Al_2O_3 laminate, removed from the kiln at 400°C.

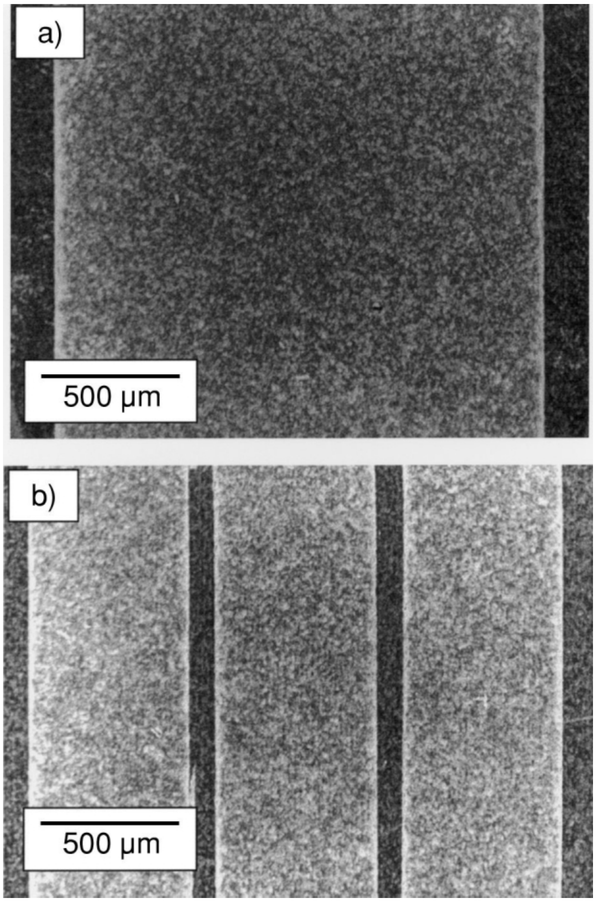

Fig. 3. Scanning electron microscopy images of polished cross-sections from three layer Al_2O_3 laminates after sintering at 1650°C using different adhesive tapes: (a) polyester carrier, (b) polyimide carrier.

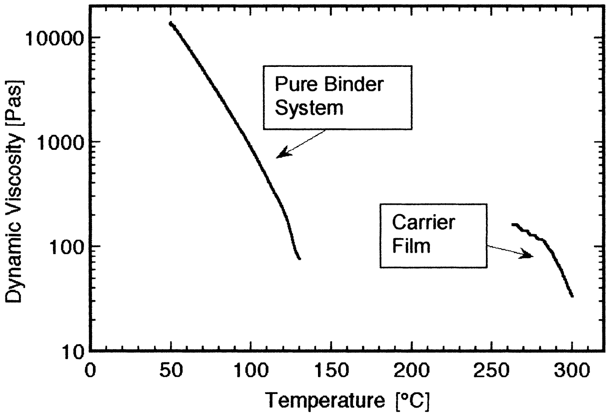

Fig. 4. Viscosity measurements as a function of temperature for the polyester carrier film of the adhesive tape and the pure binder system of the green tapes.[9]

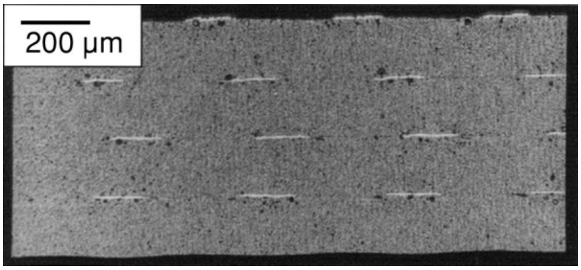

Fig. 5. Scanning electron microscopy images of a polished cross-section from a four layer LTCC laminate from polyester based adhesive tapes after sintering at 870°C.

gravimetric investigations showed that at these temperatures, the binder system of the Al_2O_3 and LTCC green tapes as well as the adhesive layers are already burned out, but sintering has not started. The movement of the particles in the presence of the melt results in an interpenetration of the particles of adjacent tapes. The following sintering process transfers this structure to defect free joining.

Further research is necessary to demonstrate the flexibility of the method, and its capability for production processes.

5. Conclusions

The presented results show that by cold low pressure lamination CLPL a joining of non-metallized and metallized tapes is obtained. The method allows lamination at low pressures and temperatures and avoids any mass flow, which occurs during the thermo-compression process. In thermo-compression this mass flow is needed to obtain an interpenetration of the particles of two adjacent tapes. In CLPL this interpenetration is achieved during the binder burnout process by the formation of a low viscosity melt from the adhesive tape carrier. This takes place in situ without any external pressure. The method is of high interest for the preparation of laminated fine, undercut, complex three-dimensional multilayer structures. The technique can also be used to join ceramic green bodies, independent on the ceramics forming process.

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References

1. *Multilayer Ceramic Devices,* ed. H. J. Jean, T. K. Gupta, K. M. Nair and K. Niwa. The American Ceramic Society, Columbus, OH, 1999.
2. Roosen, A., Basic requirements for tape casting of ceramic powders. In *Ceramic Powder Science*, ed. G. Messing, H. Hausner and S. Hirano. The American Ceramic Society, Westerville, OH, 1988, pp. 675–692.
3. Hellebrand, H., Tape casting. In *Materials Science and Technology, Vol. 17 A, Processing of Ceramic*, ed. R. J. Brook. VCH Verlagsgesellschaft, Weinheim, Germany, 1996, pp. 189–265.
4. Knickebocker, J. U., Overview of the glass-ceramic/copper substrate: a high performance multilayer package for the 1990's. *Am. Ceram. Soc. Bull.*, 1992, **71**, 1393–1401.
5. Reed, J. S., *Principles of Ceramics Processing*, 2nd edn. John Wiley & Sons, New York, 1994.
6. Gardner, R. A. and Nufer, R. W., Properties of multilayer ceramic green sheets. *Solid State Technol.*, 1997, May, pp. 38–43.
7. Pepin, J. G., Borland, W., O'Callaghan, P. and Young, R. J. S., Electrode-based causes of delaminations in multilayer ceramic capacitors. *J. Am. Ceram. Soc.*, 1989, **72**, 2289–2291.
8. Piwonski, M., Diploma thesis, University of Erlangen-Nuremberg, Germany, 1998.
9. Piwonski, M. and Roosen, A., Low pressure lamination of ceramic green tapes by gluing at room temperature. *J. Eur. Ceram. Soc.*, 1999, **19**, 263–270.