Joining of Sintered Alumina Substrates and LTCC Green Tapes via Cold Low Pressure Lamination

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Abstract

In microelectronics there is a continuous trend for devices of higher integration and improved heat dissipation. For the manufacture of ceramic based microelectronic devices the following technologies can be applied. Thick-film hybrid technology uses sintered ceramic substrates, mostly Al_2O_3*, which are screen printed with functional pastes, followed by firing at 850 °C. Alumina substrates provide very good heat conductivity (25 W/mK), but there are only two sides to carry a metallization. An improved miniaturization can be accomplished by multilayer systems using the LTCC technology. LTCC devices are manufactured by screen-printing, stacking and lamination of ceramic green tapes, followed by co-firing. A drawback of LTCCs is their low heat conductivity (3 W/mK) due to their high glass content. By combining hybrid and LTCC technology the advantages of both methods like good thermal conductivity and high multilayer integration, can be joined.*

Because the failure rate is too high to laminate green tapes on sintered ceramic substrates via thermo compression, Cold Low Pressure Lamination (CLPL) has been used as an alternative lamination process. CLPL is a lamination method, where the joining of the components is performed at room temperature by application of very low pressure (<5 MPa) by using a double sided adhesive tape. During heat treatment the adhesive film keeps the tapes together until the adhesive is completely decomposed; during further temperature increase the tapes are joined by sintering. The paper describes the materials used and processing steps to join the sintered material with the green tapes and discusses effects which occur during firing. These effects like edge curl and crack formation are mainly due to stresses which occur during constrained sintering. Their control can be influenced by changing process parameters.

Keywords: joining, lamination, Cold Low Pressure Lamination, LTCC, alumina substrate

1 Introduction

In microelectronic devices for automotive applications, power electronics and other applications with high thermal dissipation losses, the requirements for thermal management are increasing. The hybrid technology is typically used if heat dissipation is needed. Sintered substrates are metallized via screen printing, followed by drying and firing of these conductive pastes at 850 °C. With the use of VIAs, there are two sides available for metallization. The standard substrate material is alumina (Al_2O_3) with a thermal conductivity of 25 W/mK.

Higher miniaturization can be achieved by multi-layer systems using LTCC technology. LTCC is a composite material composed of fillers, such as Al_2O_3, which are surrounded by a glass matrix from a glass of low glass transition temperature. This allows densification at relatively low sintering temperatures of 850 °C. After screen printing and stacking, the green tapes are laminated using thermo-compression, followed by co-firing of the multilayer device. A drawback of LTCC technology is the poor thermal conductivity of only 3 W/mK, due to the high content of glass phase.

During thermo-compression, the stacked green sheets are joined by applying pressure at a temperature above the glass transition point of the binder - plasticizer system of the green tape to provide material flow [1]. This is typically done around 80 ° C and 30 MPa. Because of this flow of material, it is difficult to find process parameters which avoid damage to the screen printed layouts.

To avoid deformation during sintering of multilayer LTCCs several zero-shrinkage techniques are available which suppress lateral shrinkage. They are based on pressure-assisted, sacrificial tapes or self-constrained sintering.

Joining of hybrid and LTCC technology is not obvious. Combination of a sintered material with a green sheet is not possible via thermo-compression because of the high failure rate. Therefore, Cold Low Pressure Lamination (CLPL) is used to join the alumina substrate and the LTCC green tapes, as it works at low pressures (<5 MPa). At this process, joining of the materials takes place by means of a double-sided adhesive tape. During burnout of the thermoplastic binder system, the adhesive tape provides capillary and adhesive forces, which keeps the joining partners together. During sintering, a complete joining is achieved by diffusion processes. In addition, a constrained sintering [2] effect occurs in the green sheet because of the sintered substrate, which suppresses lateral shrinkage. Thus, deformation of the green body by shrinkage can be avoided.

2 Experimental Procedure

2.1 Materials

Sintered alumina thickfilm substrates Rubalit 708 S, CeramTec GmbH Marktredwitz, Germany) with 96% Al_2O_3 had been used. Some of these substrates were metalized by screen printing with a silver paste. The LTCC green tapes were provided by Heraeus GmbH (Hanau, Germany). This Heratape CT with a thickness of 130 µm is based on a borosilicate glass matrix, filled with alumina particles as a filler. It is free of lead, cadmium and nickel to hold up environmental legislation. The coefficient thermal expansion (CTE) is 7.0 ppm/K (25 – 700°C) which is similar to the CTE of the alumina substrates with 7.1 ppm/K (25 – 700°C). Thus, thermal stresses can be avoided during heat treatment. For Cold Low pressure Lamination a double sided adhesive tape from Beiersdorf AG (Hamburg, Germany) was applied (Fig. 1). The backing is made of polyethyleneterephthalate (PET) and functions as a carrier tape for the adhesive films. The total thickness of the double sided adhesive film is 12 µm.

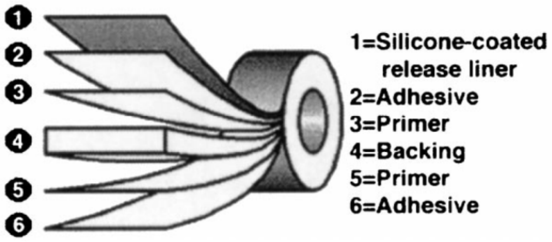

Fig. 1: Structure of the double-sided adhesive tape (Beiersdorf AG, Hamburg, Germany)

The laminates were made of a green tape which was glued to the alumina substrate using the adhesive tape. The processing steps where similar to these used for producing laminates only consisting of green tapes as shown by Piwonski and Schindler [3,4].

2.2 Lamination and Firing Process

The lamination process is shown in Fig. 2. In the first step the double sided adhesive tape was attached to the alumina substrate using a soft rubber roll. It is essential for the quality of the laminates to avoid dust or air-pockets between the ceramic

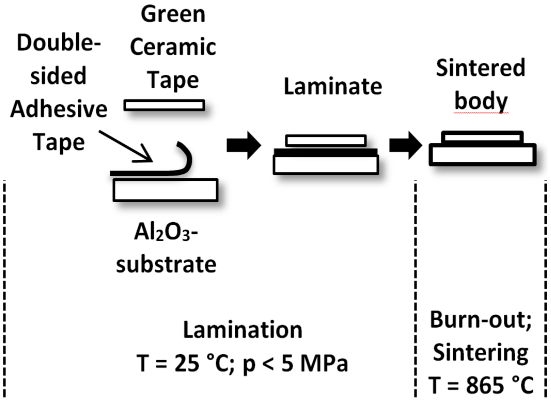

Fig. 2: Processing steps in Cold Low Pressure Lamination

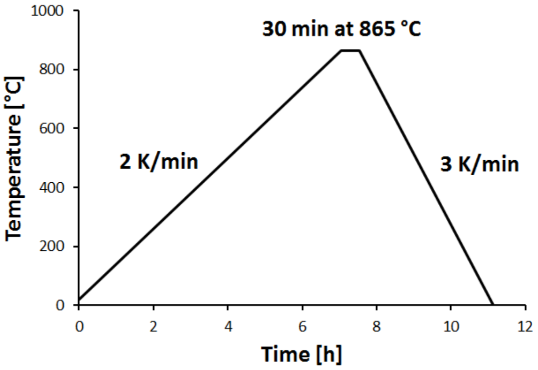

Fig. 3: Firing scheme of the laminates.

surface and the adhesive tape. Then the release liner was removed. In the second step, the LTCC was applied to the adhesive film at room temperature and a pressure of appr. 5 MPa. Subsequently, the laminate was subject to binder burn-out and sintering. For firing of the laminated structure composed of alumina substrate – adhesive film – LTCC green tape the data provided by Heraeus for sintering of the LTCC tape was used (Fig. 3).

2.3 Methods for Materials Characterization

To understand the materials behavior during heat treatment, the LTCC green tape and the adhesive tape were characterized by thermogravimetric analysis (TGA). In addition, the green tapes were characterized by means of differential scanning calorimetry (DSC), dilatometry and uniaxial viscosity measurement. All measurements were carried out with the heating profile provided by Heraeus. The TGA and DSC experiments were performed in a STA 429 and DSC 404 from Netzsch Gerätebau GmbH, Germany. Shrinkage was determined with a DIL 402 from Netzsch Gerätebau GmbH. A thermo-mechanical analysis (TMA) was carried out in a TMA 60H from Shimadzu, Japan to measure the uniaxial viscosity in a temperature range of 500 to 900°C.

To produce a sample big enough for dilatometry and uniaxial viscosity measurement, several layers of LTCC green tape were stacked and laminated at 80 °C with a pressure of 25 MPa via thermo-compression. The samples were debindered by heating up to 500°C at 2 K/min and a holding time of 1 hour to remove the organic additives. This was done to avoid incorrect measurements due to delamination of the samples in the lower temperature regime. For the TMA measurement the applied load was 1.1 N which is equivalent to a stress of approximately 20 kPa [5,6]. The load was alternately applied every 25 °C and hold for one minute.

2.4 Methods for Laminate Characterization

To determine the quality of the lamination polished cross sections were analyzed using a scanning electron microscope. The surface profile of the sintered laminate was scanned using confocal microscopy (µsurf explorer, Nanofocus AG, Oberhausen, Germany) to characterize crack and warpage formation. To observe the quality of the lamination and the decomposition of the adhesive tape during binder burn-out and sintering, samples were removed from the furnace at temperatures of 200 °C and every 50°C up to 450°C and every 40°C between 480 up to 860°C. The samples were cut, polished and analyzed by means of electron and optical microscopy.

A direct optical observation of green tapes and laminates in vertical and horizontal direction during heat treatment was carried out using an optical dilatometer [7]. Two cameras combined with a background lightning allow the observation of the tapes or laminates which are located inside the furnace in the intercept point of the two optical axis. Thus, the shrinkage and distortion behavior of the samples can be monitored during heat treatment.

3 Results and Discussion

3.1 Quality of the Laminates

The polished samples of the interface region between LTCC and the Al_2O_3 substrate show a very good bonding between the two layers as seen in Fig. 4. In the interface no delamination or pores can be detected in spite of the massive amount of organic components in the laminate which comprises binder, plasticizer and adhesive film.

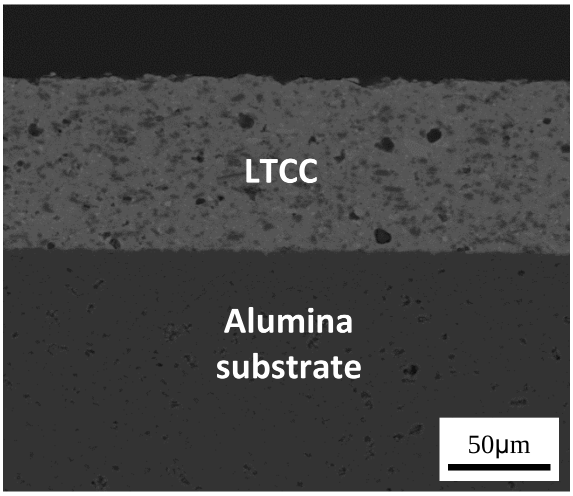

Fig. 4: SEM image of the sintered laminate indicating a good bonding between the LTCC and the alumina

At room temperature and intermediate temperatures the adhesive tape holds the LTCC in place on top of the alumina substrate. Fig. 5 shows two laminates which were removed from the furnace at different firing temperatures of 250 and 400 °C, respectively. The rest of the adhesive tape is still visible, but the figures also indicate the decrease of the adhesive film thickness with increasing temperature. It has to be taken into account that both, the binder and the adhesive film are composed of thermoplastics, which melt during the binder-burnout process, given rise to capillary forces. At 400 °C the binder-plasticizer system is nearly decomposed. With increasing temperature decomposition of the adhesive tape continues, and volume of the adhesive shrinks, but capillary and adhesive forces are still active keeping the substrate

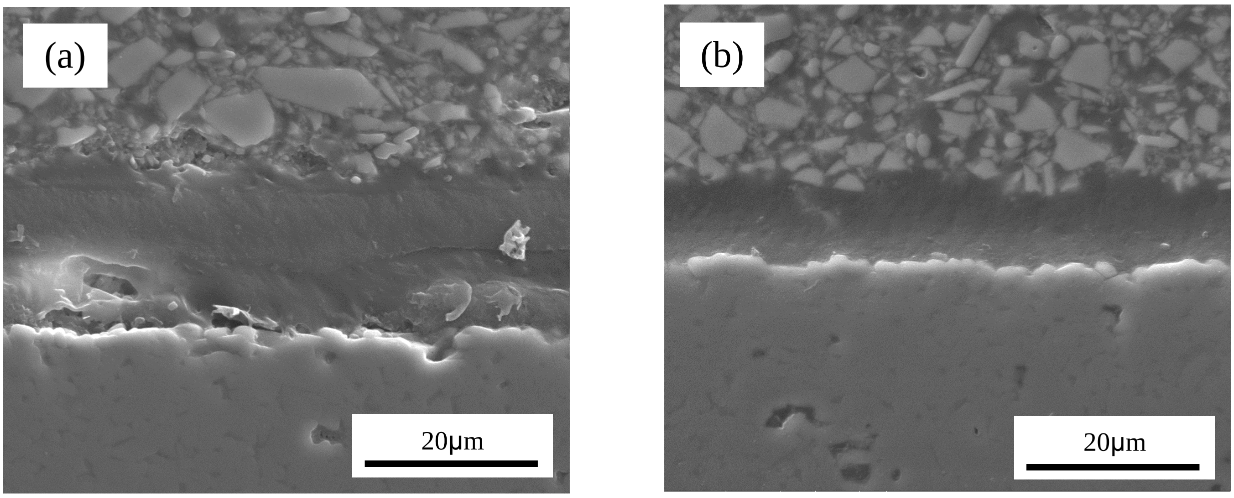

Fig. 5: SEM images of the interface region of the laminates after removal from the furnace at different temperatures: (a) 250 °C, (b) 400 °C. It can be seen that the adhesive shrinks and that the two layers are approaching each other.

and LTCC tape together, while both are approaching each other. At higher temperatures, the glass phase of the alumina substrate as well as the LTCC will exhibit interdiffusion, resulting in a very good joining between both parts. During this process the LTCC densifies in a constrained sintering process, which leads to stresses during shrinkage and cooling. These stresses can result in defect formation.

3.2 Edge Curl

Such a defect is delamination of the LTCC edges and an edge curl effect of the LTCC relative to the alumina substrate as shown in Fig. 6. The edge curl effect is widely known in sintering of green tapes, but its origins are not described in literature. It is unwanted because it can interfere with subsequent processes.

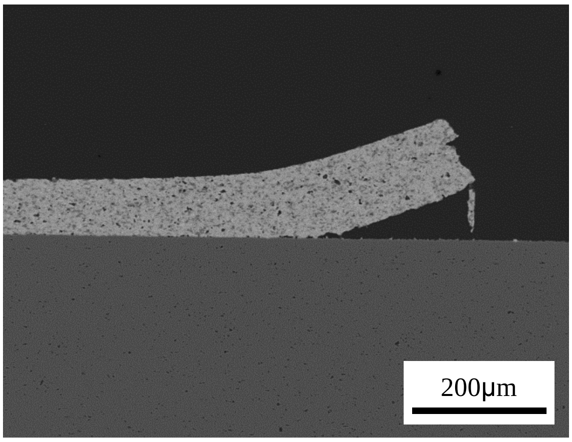

Fig. 1: SEM image of a pronounced edge curl effect of the sintered 3-layer LTCC laminate on the alumina substrate after firing.

To understand the edge curl effect, several experiments were conducted to determine the factors involved. As a measure for the edge curl, the distance between the LTCC surface and the maximum height of the edge was determined. Following this definition, the edge curl in Fig. 6 is 85 µm.

To investigate the parameters which might influence the edge curl, the LTCC size, the LTCC thickness, the number of layers laminated via CLPL, and the side of the LTCC which is glued to the alumina surface were changed. The latter takes into account that the upper side of the green sheet, which was in contact with air during tape casting contains a higher amount of binder than the bottom side [8]. In addition, a 3-layer LTCC stack was laminated via thermo-compression and then laminated onto the alumina substrate surface via CLPL.

It could be shown that neither the size nor the side, which was glued to the surface, had any effect on the magnitude of the edge curl after firing. But with increasing thickness of the individual green sheet and number of layers laminated, the edge curl was increased. With rising number of layers the edge curl of every layer accumulated. There was no difference in edge curl for a thicker green sheet and the laminate of the same thickness, but composed of thinner layers. The LTCC stack which was laminated via thermo-compression and laminated onto the substrate via CLPL also displayed an edge curl, but of a lower value than the laminate which was fully laminated by CLPL.

To determine the starting temperature of the edge curl, a CLPL laminate, consisting of 3 layers, was laminated to an alumina substrate by CLPL and investigated in the optical dilatometer. To investigate the influence of constrained sintering on the edge curl, a second multilayer LTCC sample was produced by CLPL. But in contrast to the specimen above, this specimen was just deposited on the sintered substrate without any lamination. As shown in Fig.7, the LTCC laminate, which was not laminated to the surface, displayed severe warpage during sintering. The distortion began at 770 °C and reached its maximum at 825 °C, when the laminate exhibited the shape of a bowl. These deformations are caused by inhomogeneities of the green laminate structures causing shrinkage gradients. At higher temperature of 865 °C, the laminate transforms to a flat structure, due to the softening of the LTCCs glass phase. The sample which was laminated to the sintered Al_2O_3 substrate showed no warpage effects. The laminate adheres to the substrate which prevents any warpage. But at 830 °C the LTCC edges began to curl upwards until 865 °C, when the edge curl reached its maximum. No further effect could be observed during cooling.

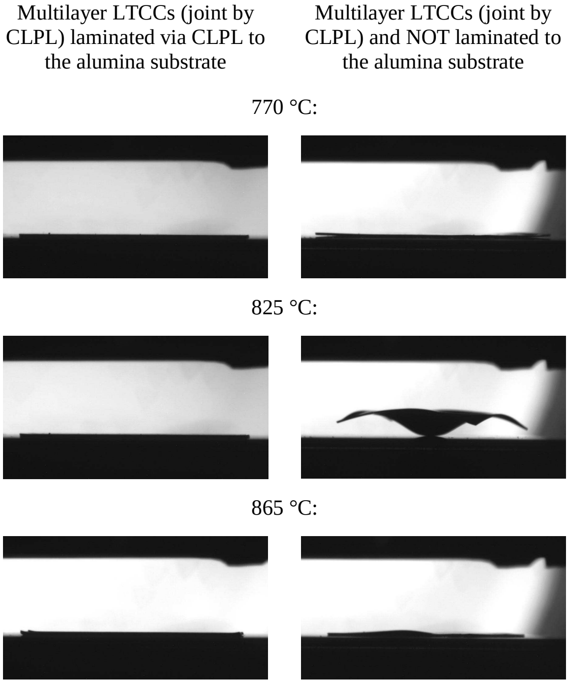

Fig. 7: Photos of multilayered LTCCs, laminated via CLPL, during sintering in an optical dilatometer

Fig. 8 shows the DSC plot of the LTCC green tape used in the study. The crystallization of the glass phase starts at 830 C. At this temperature, edge curl formation was observed, too. It seems that crystallization, which is connected with changes in density, results in stresses. Only at the edges, where the adhesion between LTCC and alumina substrate is the lowest, the stresses are relieved by edge curl formation. A lower edge curl of the LTCC laminate occurs, if the LTCC laminate was produced by thermo-compression instead of CLPL, followed by CLPL to join LTCC and the alumina substrate.

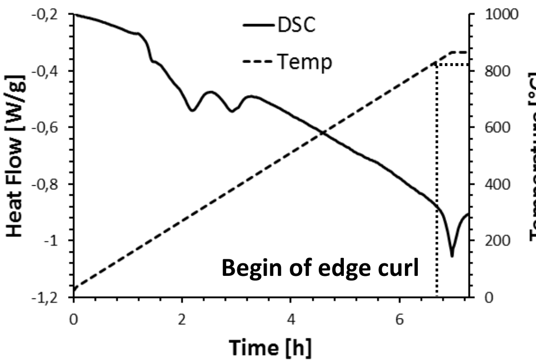

Fig. 8: DSC curve of the LTCC heated up to 865°C. Crystallization of the glass phase occurs at 830 °C, which is the temperature at which edge curl formation was observed.

3.3 Crack Formation

LTCCs laminated to a blank alumina substrate by CLPL did not show any cracks after firing. If the Al_2O_3 substrate is metalized by screen printing, defects occur in the LTCC layers which were laminated on these metalized substrates. As shown in Fig. 9 the LTCC tapes cracked just above the printed circuit line during sintering, due to tensile stresses.

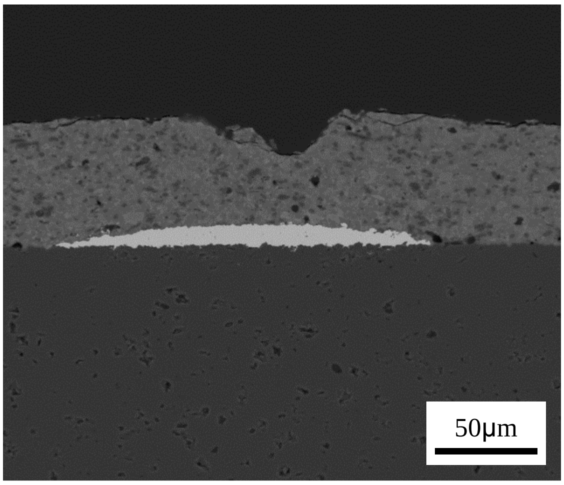

Fig. 9: V-shaped crack formation in a sintered LTCC-alumina laminate, joint by CLPL

To determine the temperature, at which the cracks in the LTCC tape occur, appropriate specimens of metallized Al_2O_3 substrates with LTCC laminated on top of it were removed from the furnace at different temperatures. The characterization by means of confocal microscopy clearly shows that–small cracks appear above the metallization line at a temperature of 680°C (Fig. 10). With rising temperature these cracks above the conductor line propagate until an extended crack is clearly visible.

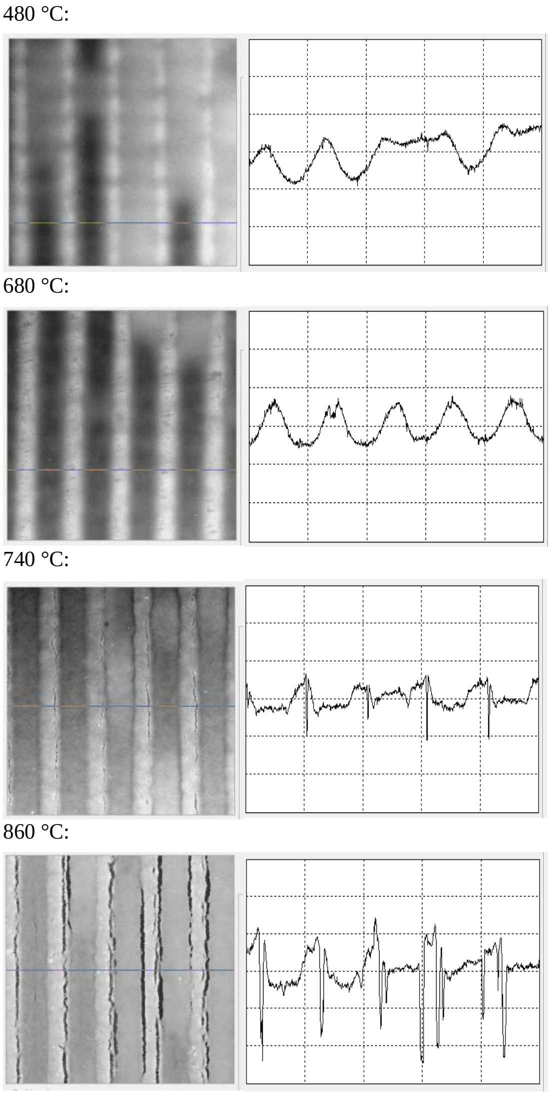

Fig. 10: Surface top view and profile of sintered LTCCs laminated via CLPL onto alumina substrates investigated using confocal microscopy. The specimens were removed from the furnace at different temperatures.

All cracks exhibit a V-like shape as shown in Fig. 9. The crack tip is typically located above the middle of the conductor line. Thus it can be assumed that crack formation starts at the surface of the LTCC, just above the highest point of the metallization. Here, the green tape, laminated onto the metallization, exhibits its highest bending. Therefore, a stress peak is formed on top of every heightening, caused by conductor lines or other metallization structures. With increasing temperature and advancing decomposition of the adhesive tape the LTCC sheet is pulled closer to the alumina and tensile stresses are rising. After the removal of the binder the tape is a weak porous network of interlocked inorganic particles held together by particle-to-particle adhesion forces. Further temperature increase results in a decrease of the viscosity (Fig. 12), which results in a shrinkage onset at 680 °C (Fig. 11) due to viscous flow [9]. At this temperature, the first cracks can be detected.

The sintered alumina substrate constrains the shrinkage of the LTCC in the x-y plane. This shrinkage occurs in z-direction. At the interface the constraining effect is very pronounced, but diminishes to the top surface of the LTCC. Here, a small degree of lateral shrinkage takes place, which causes stress formation and centers above the metallization. When the tensile stresses are higher than the adhesive forces between the particles, cracks occur. It is assumed that local weakening of the material already appears at lower temperatures than 680 °C, but they become visible as shrinkage starts.

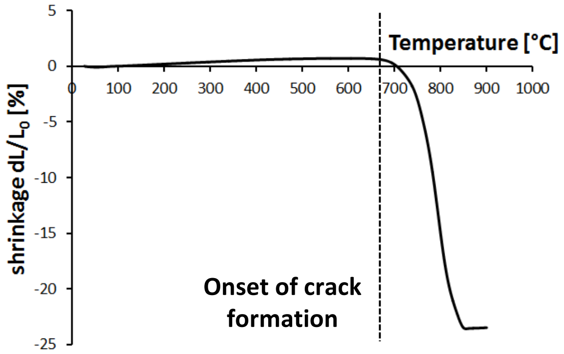

Fig. 11: Shrinkage in dependence on temperature and onset temperature of crack formation.

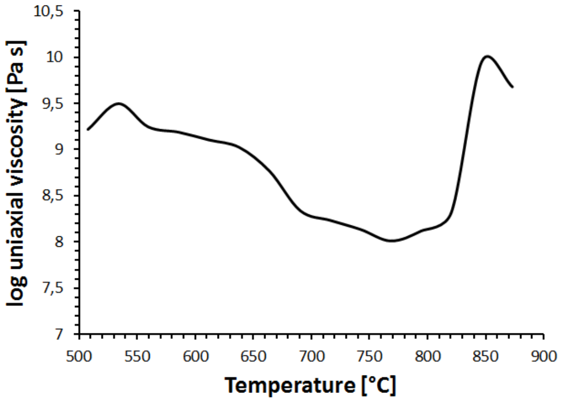

Fig. 12: Uniaxial viscosity of the LTCC tape in dependence on temperature.

With this background it is possible to adjust the process parameters and sintering profile to avoid crack formation as can be seen in Fig. 13.

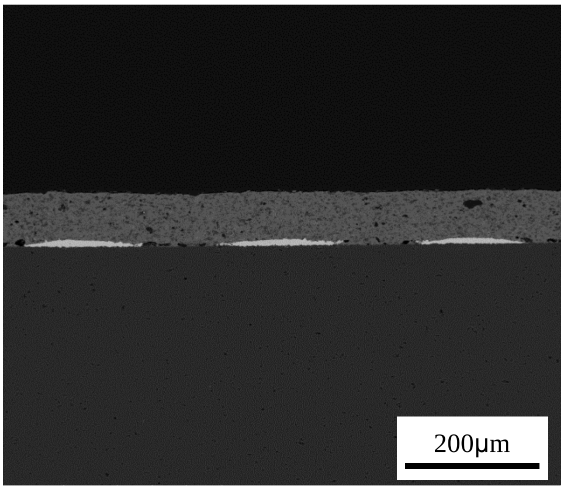

Fig. 13: SEM image of a polished cross section of a sintered LTCC, laminated onto a metallized alumina substrate using CLPL.

4 Conclusion

The paper describes the processing steps to combine hybrid and LTCC technology via Cold Low Pressure Lamination.

For non-metalized alumina and LTCC, the alumina-LTCC interface showed very good bonding between the two layers without porosity or delaminations. Independent of the joining effect, an unwanted edge curl effect occurs during sintering. Edge curl formation occurs at about 830 °C at the beginning of the crystallization temperature of the glass phase and reaches its maximum at the final temperature of 865° C. Sintering stresses that occur during firing are caused by inhomogeneities in the green body and are responsible for this effect. In case of metallized alumina substrates, crack formation in the LTCC tapes above the metallization was observed at temperatures of 680 °C. This temperature coincides with the beginning of shrinkage and thus the viscous flow of the glass phase. Tensile stresses generated by the bending of the tapes over the conductor line, which increase during densification, are responsible for tearing.

With the knowledge about the origin of these effects, the process parameters can be adjusted to avoid edge curl and crack formation.

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References

[1] H. Hellebrand, “Tape Casting, in Processing of Ceramics”, Part 1, R.J. Brook (ed.), VCH Verlagsges., Weinheim, Germany, pp. 190-265, 1996.

[2] T.J. Garino, H.K. Bowen, “Kinetics of Constrained-Film Sintering”, J. Amer. Cer. Soc. 73, pp. 251-257, 1990.

[3] M. Piwonski, A. Roosen, “Low Pressure Lamination of Ceramic Green Tapes by Gluing at Room Temperature”, J. Eur. Cer. Soc. 19, pp. 263-270, 1999.

[4] K. Schindler, A. Roosen, “Manufacture of 3D structures by cold low pressure lamination of ceramic green tapes”, J. Eur. Cer. Soc. 29, pp. 899-904, 2009.

[5] A. Mohanram, G.L. Messing, D.J. Green, “Measurement of Viscosity of Densifying Glass-Based Systems by Isothermal Cyclic Loading Dilatometry”, J. Amer. Cer. Soc. 87, pp. 192-196, 2004.

[6] R. Zuo, E. Aulbach, J. Rödel, “Experimental determination of sintering stresses and sintering viscosities”, Acta Materialia 51, pp. 4563-4574, 2003.

[7] A. Roosen, D. Pohle, M. Wagner, “Characterization of the shrinkage behavior of pure, screen printed thick film pastes and LTCC green tapes”, Proceedings of the 2004 IMAPS Conference on Ceramic Interconnect Technology, Denver, CO, USA, June, pp. 260-265, 2004.

[8] A. Roosen, “Tape casting of ceramic green tapes for multilayer device processing”, Ceramic Transactions Vol. 97, H.J. Jean, T.K. Gupta, K.M. Nair, K. Niwa (eds.), The Amer. Cer. Soc., pp. 103-121, 1999.

[9] S. Kemethmüller, M. Hagymasi, A. Stiegelschmitt, A. Roosen, „Viscous flow as the driving force for the densification of LTCC ceramics”, J. Amer. Cer. Soc. 90, pp. 64-70, 2007.