Effect of Dielectric Selection on Sintering Behavior of LTCC

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Abstract

Low Temperature Cofired Ceramic (LTCC) is a versatile ceramic packaging technology. LTCC's functionality and performance can be enhanced with the incorporation of passive devices such as resistors, capacitors, and inductors into the structure of the substrate. This incorporation, which eliminates surface mount solder joints is desirable for improving system reliability and allows for increased package density. Commercially available dielectrics for embedding capacitors into LTCC are limited and the incorporation of such systems into LTCC substrates poses processing challenges related to material compatibility. This paper will review multiple dielectric systems and techniques for embedding capacitors into LTCC as well as present new data related to the cofiring behavior of LTCC and custom dielectrics.

Keywords: LTCC, buried passives, buried capacitors, DuPont 951™, barium titanate

I. Introduction

Ceramic multichip module (MCM – C) technology has advanced through the increasing integration of active and passive devices. The incorporation of passive devices into integrated circuits (ICs) can be limited by material compatibility and cost restraints [1, 2]. Another method for increasing MCM integration is to incorporate into the substrate. Researchers and material suppliers have presented multiple ways to incorporate passives into LTCC substrates with varying degrees of success. Material compatibility between the passive device and the substrate is a challenge when incorporating the devices with LTCC, but one that can be managed through material selection and processing modifications.

LTCC offers the opportunity to incorporate passive devices, specifically capacitors, by utilizing the substrate as the dielectric, printing dielectrics on the surface or internally, and embedding specially formulated ceramic tape internally. Utilizing the substrate as the dielectric limits the capacitance to low values or requires large areas to achieve useful values. Printing or embedding high dielectric constant dielectrics tapes is a practical approach for incorporating high value capacitors into LTCC substrates.

II. Technology Review

There are a limited number of commercially available high K dielectric pastes that can be utilized for creating surface or embedded high value capacitors in LTCC. There are a limited number of commercially available high K dielectric tapes that are compatible with LTCC materials and processing [3].

While the use of the LTCC is a simple way to embed a capacitor into the LTCC structure, it is not effective for high capacitance. The most commonly referenced method of incorporating embedded high K capacitors into LTCC is by screen printing a high K dielectric paste. This method is advantageous because of the ability to define the dielectric area precisely and position it accurately with respect to the electrodes. A disadvantage is the limited selection of dielectrics that are available and the difficulty in formulating custom dielectric pastes. Another disadvantage is the tendency for increased interaction of the paste with the unfired LTCC tape through solvent transport.

Embedding high K dielectric tapes can also be used to create high value internal capacitors in LTCC. This can be done selectively or as an entire layer within the structure. Tape casting is a widely used method of fabricating custom dielectric ceramics for new applications. Tape casting provides a flexible format for incorporating a variety of formulations with low capital investment during the early development stages of high K material development for embedded capacitors in LTCC. A second advantage to using high K dielectric tape for the embedded capacitors is that thicker dielectric layers are achievable that can be used to compensate for the electrode dielectric interaction zone that can occur with some of the metallizations. A third advantage is that special screens are not required to define the dielectric area. Disadvantages include challenges with sizing and placement of the dielectric tape if it is selectively applied to specific locations and sintering mismatches if the formulation is not optimized for the LTCC tape system being used.

An expected common disadvantage of the second and third methods is the CTE mismatch between the dielectric and the LTCC material, causing significant stress buildup during environmental testing. Dielectric shrinkage mismatch to the LTCC substrate can also cause undesirable convex or concave topography in the substrate, or cracking in either the LTCC or the capacitor dielectric. [5]

III. Experimental Procedure

This paper discusses two portions of evaluations related to the embedding of a new tape into DuPont 951 LTCC tape. The first portion consists of understanding the separate and combined sintering behaviors of the new dielectric tape and LTCC tapes. The second portion consists of fabricating functional embedded high K capacitors into an LTCC multilayer structure.

Development of High K Dielectric Tape

In this study, a unique dielectric tape was evaluated for compatibility with DuPont 951. The dielectric tape was composed of $BaTiO_3$ (NEB, Ferro Co., Cleveland, OH) as well as a unique glass powder developed and fabricated at the Missouri University of Science and Technology. The BaO-B_2O_3-Al_2O_3-SiO_2 glass was added to the dielectric powder to promote liquid-phase sintering and ultimately increase the dielectric breakdown strength of the sintered material. The specific glass composition and preparation method have been previously documented by Young et al. [4].

For this evaluation, 20 vol% of the custom glass powder was added to the NEB within the dielectric tape; the composition of the tape will be referred to hereafter as NEB20. Based on previous studies [6], this relatively high concentration of glass in the tape was desired in order to lower the sintering temperature of the NEB dielectric and bring it closer to the optimum sintering temperature of the DuPont tape (850°C). Previous research by Young et al. showed that a 20 vol% glass addition lowered the sintering temperature of the $BaTiO_3$ material from approximately 1300°C to 990°C.

To fabricate the NEB20 dielectric sheets, a slurry was prepared containing 80 vol.% NEB and 20 vol% of the custom glass powder. The NEB powder, Ba-glass powder, solvent, and dispersant were ball milled for 24 hours using ZrO_2 milling media. After adding the binder and plasticizers, the slurry was milled for another 24 hours and then rolled slowly for 24 hours. The slurry was cast on silicone coated mylar to produce tape with a dried thickness of 0.00375 ± 0.00025 inch. The NEB20 tape was flexible, uniform, and easily handled. To determine the appropriate co-firing temperature for NEB20 and DuPont 951 tapes, the sintering behavior and linear shrinkage of both tapes were investigated separately.

Sintering Evaluation of NEB20 and LTCC

Single layer disks were cut from the NEB20 and DuPont 951 tapes. After binder burnout at 400°C, single layer disks of NEB20 were sintered at 850°C, 900°C and 950°C for hold times of 1 or 2 hours.

In order to explore if a symmetrical structure of NEB20 and DuPont 951 can eliminate the effect of mismatch in sintering temperature and shrinkage, a sandwich structure laminate of NEB20 and DuPont 951 was prepared. Several ~1.25 inch disks of DuPont 951 and NEB20 tape were prepared. The laminate consisted of 2 DuPont 951 disks (250 µm thick each) sandwiched between two NEB20 disks. The stack was uniaxially pressed at room temperature with an applied pressure of 2.5 kpsi for 2 min, followed by cold isostatic pressing at room temperature and a pressure of 35 kpsi for 5 min. The samples then underwent binder burnout at a heating rate of 0.25°C/min to 400°C for 4 hours in air, followed by co-firing at various temperatures from 850 to 950°C.

Three DuPont 951 tape disks were stacked alternatively with four NEB20 tape disks to obtain seven layer stacks. Both the DuPont and NEB20 disks had the same diameter. To investigate if the stacking order of the top and bottom surfaces of the as-cast tape affects the flatness of co-fired multilayer parts, three order combinations with differing top and bottom layers of the tape disks were prepared (we left out a detailed discussion of the ordering as the results show no major differences). The three multilayer stacks with different order combinations of tape were laminated directly by isostatic pressing at room temperature, underwent binder burnout at a heating rate of 0.25°C/min to 400°C for 4 hours in air, followed by co-firing at 850°C for 0.5 hours.

Fabrication of Embedded Capacitors

For this evaluation, 8-layer panels of DuPont 951PX were fabricated (Figure 1). Layer 1 will refer to the bottom layer of the stack, while Layer 8 will refer to the top layer of the stack. Discrete pieces of the NEB20 tape were inserted between layers 4 and 5 in each panel to create embedded capacitors.

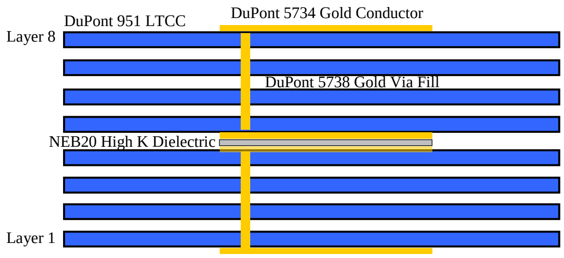


Figure 1: Cross section schematic of embedded high K dielectric embedded capacitor in LTCC.

Prior to use, the 951PX tape was conditioned at 100°C for 60 minutes, and the NEB20 tape was conditioned at 100°C for 15 minutes. Unique patterns of 0.010” diameter vias were punched into each of the eight tape layers using a Pacific Trinetics Corp. (PTC) Automatic Punching System.

The internal vias in each 951PX layer were filled with a gold conductor paste (DuPont 5738) using AMI screen printers, and the printed tape was dried at 70°C for 6 minutes. Conductors were printed on the top of Layer 4, bottom of Layer 5, and top of Layer 8 using a gold conductor paste (DuPont 5734), and the prints were dried at 70°C for 6 minutes. The ultimate fired thickness of the electrodes was ~11 - 17 µm .

Discrete pieces of NEB20 tape (~ 5 mm x 5 mm) were affixed on top of select electrodes on Layer 4 of the 951PX (figure 2). Note that the exposed portion of bottom electrodes in figure 2 are electrically isolated from the top electrode, which is not shown in this picture.

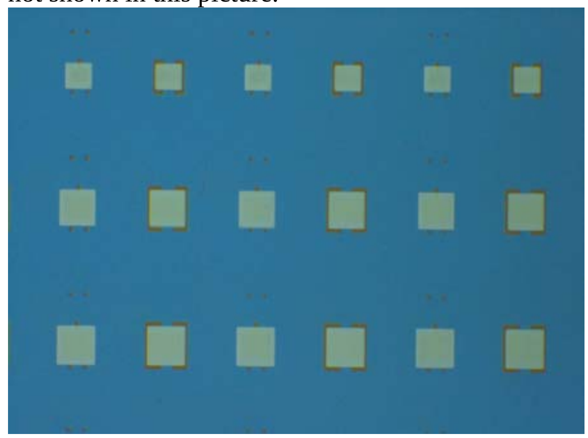


Figure 2: Top view of layer 4 showing bottom gold electrode with NEB20 tape dielectric squares.

The eight layers were aligned, stacked, and then laminated using a PTC isostatic press. After preheating in the press for 5 minutes at 70°C, the panels were pressed at 3000 psi for 10 minutes. The panels were then placed into an exhausted box furnace and heated at 2°C/min to 450° C, held for 3 hours, then heated at 2°C/min to the sintering temperature. The sintering temperatures and times for the various panels are as follows: Panels 1 & 2 – 900°C for 1 hour, Panels 3 & 4 – 900°C for 15 minutes, Panels 5 & 6 – 900°C for 1.5 hours, Panels 7 & 8 – 850°C for 15 minutes, Panels 9 & 10 – 950°C for 15 minutes.

After firing, a gold conductor (DuPont 5771) was printed on bottom of each panel to complete the connection with the top layer probe structure. The panels were dried at 150°C for 10 minutes and then fired at 850°C for 9 minutes.

IV. Results and Discussion

Sintering Shrinkage of NEB20

The percent linear shrinkage of the NEB20 disks was determined by measuring the change in diameter of eight disks for each sintering condition and the results are plotted in Figure 3. The shrinkage of DuPont 951 tape sintered at 850°C for 1 hour is also included in Figure 3 for comparison. As shown, the amount of shrinkage of the NEB20 increases with sintering temperature and hold time up to 950°C. Hold times of 1 and 2 hours at 950°C did not affect the shrinkage, therefore the NEB20 should be sintered to near full density at 950°C. Subsequent microstructure analysis (discussed below) also supports the conclusion that the sintered full density is achieved at 950°C.

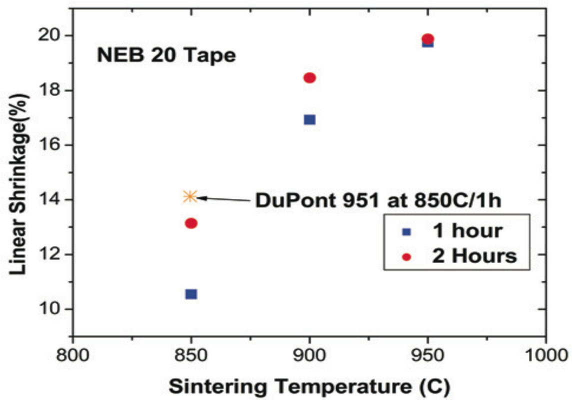


Figure 3: Linear shrinkage of NEB20 disks at various temperatures for hold times of 1 and 2 hours.

Microstructure of Sintered NEB20

Figure 4 shows the surface microstructure of DuPont 951 tape sintered at 850°C for 1 hour. As seen, the ceramic grains are surrounded by an amorphous phase, but a number of larger pores (1 to 3 µm in size) exist in the sintered tape cross sections obtained by fracture. Figures 5 to 7 compare the microstructures of the surfaces and cross sections of the NEB20 disks sintered at 850, 900, and 950°C, respectively. The micrographs indicate that the densification process for NEB20 is still proceeding, but incomplete, between 850 and 950°C for hold times up to 2 hours. There is approximately 20% porosity remaining in the NEB20 sample sintered at 850°C (Figure 5), while the amount of porosity was reduced to <5% in the sample sintered at 950°C (Figure 7). Furthermore, the higher magnification SEM images in Figures 5, 6, and 7 show that the $BaTiO_3$ grain size increases with increasing sintering temperatures from 850°C to 950°C. The grain size ranges ~ 1 µm to ~ 2 µm over the temperature range discussed.

Based on the results of the sintering shrinkage and microstructural investigations, it can be concluded that the sintering temperature and shrinkage of NEB20 tape still did not match with the DuPont 951 tape, because the T_g of the Ba-glass may be much higher than the T_g of the glass in the DuPont 951. Therefore, further increases in the Ba-glass fraction are not expected to further reduce its sintering temperature or further improve the match with the sintering temperature of DuPont 951. However, a symmetrical structure may balance the effect of mismatch in sintering shrinkage between the NEB20 and DuPont 951 to keep co-fired devices flat and functional.

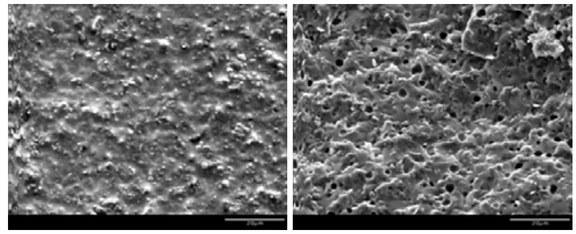


Figure 4: Microstructure development of DuPont 951 sintered at 850°C for 1 hour (left - DuPont 951 Surface, right - DuPont 951 cross section).

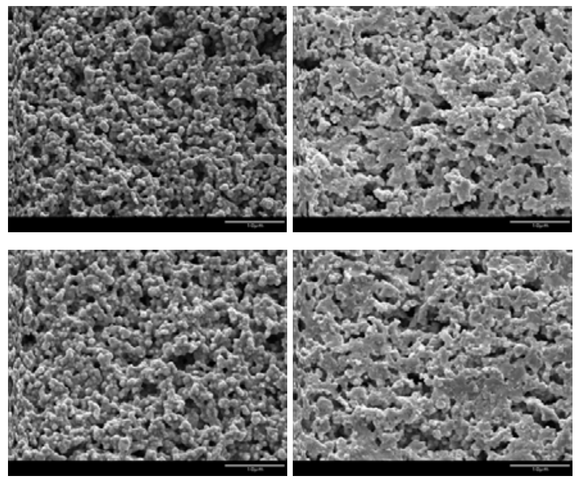


Figure 5: Microstructure development of NEB20 sintered at 850°C for 1 or 2 hours (top left - NEB20 surface 850°C/1h, top right - NEB20 cross section 850°C/1h, bottom left - NEB20 surface 850°C/2h, bottom right - NEB20 cross section 850°C/2h).

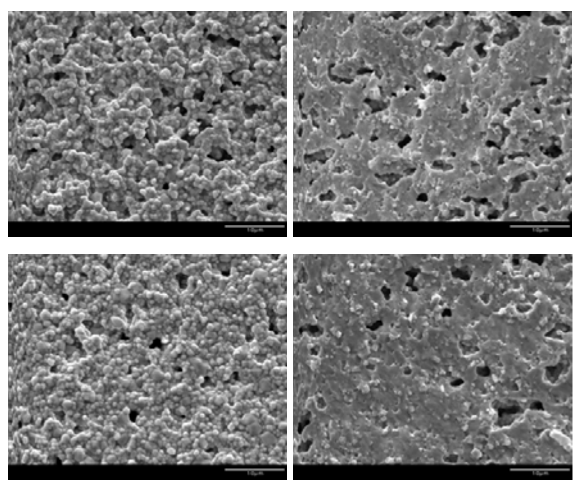


Figure 6: Microstructure development of NEB20 sintered at 900°C for 1 or 2 hours (top left - NEB20 surface 900°C/1h, top right - NEB20 cross section 900°C/1h, bottom left - NEB20 surface 900°C/2h, bottom right - NEB20 cross section 900°C/2h.

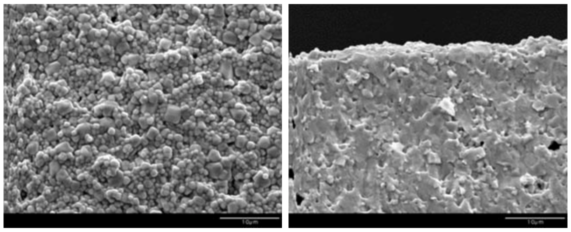


Figure 7: Microstructure development of NEB20 sintered at 950°C for 1 hour (left - NEB20 surface 950°C/1h, right - NEB20 cross section 950°C/1h).

Lamination

The cross section of the unfired stack was observed using optical microscopy and is shown in Figure 8. As seen from the image, no visible delamination between NEB20 and DuPont 951 disks was observed. However, the outer ring portion of the NEB20 disks, away from the DuPont 951 disk in the center of the sandwich stacks, did not laminate together. This is because the uniaxial pressing does not allow the NEB20 disks to conform around the 500 µm DuPont 951 thickness due to the rigid tooling used in this stage of the lamination. The deformability of NEB20 may also not be as high as DuPont 951. Consequently, an air pocket around the DuPont disk was formed.

In order to avoid the defect, additional sandwich laminates were prepared using NEB20 and DuPont 951 disks with the same diameter. After sintering at 850˚C for 0.5 hours, cracks were observed on the surface of the NEB20 disks (Figure 9). Possible origins for the cracking include: 1) lower compressibility of NEB20, compared to DuPont 951, resulting in some micro-cracking in the green NEB20 disk created by the uniaxial press; and/or 2) a mismatch in thermal expansion and/or sintering shrinkage between NEB20 and DuPont 951 tapes. In order to avoid microcracking in the green NEB20 disks, the sandwich stacks were laminated using an isostatic press at 35 kpsi at room temperature.

From images of the sandwich stacks before and after sintering (Figure 10), no visible cracking was found on the surface of NEB20 except for the outer ring portion of the NEB20 disk away from the center of the disks where the DuPont 951 disk was located. It can be concluded that the primary origin of the cracking on the NEB20 disk surface in the sintered sandwich is micro-cracking created by the uniaxial press. The cracking in the outer ring portion of the NEB20 disk may be caused by a mismatch in sintering shrinkage and/or thermal expansion coefficient between NEB20 and DuPont 951.

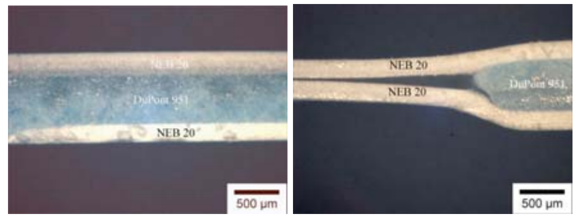


Figure 8: Optical images of the cross section of laminated NEB20 and DuPont tapes.

Figure 9: Sandwich specimen of NEB20/DuPont 951 laminated using a uniaxial press followed by room temperature isostatic pressing and co-firing at 850˚C for 0.5 hours.

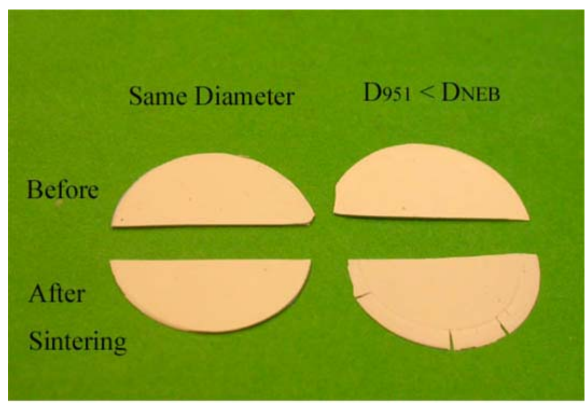


Figure 10: Sandwich specimen of NEB20/DuPont 951 laminated only by isostatic pressing at RT.

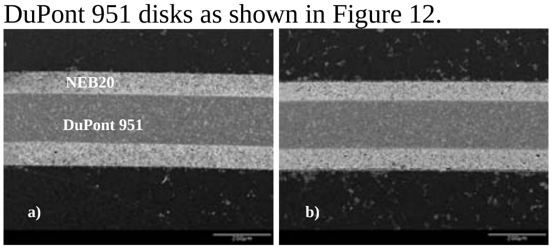
Co-firing Sandwich laminate of DuPont 951/NEB20 Tapes

The microstructure of the sandwiches sintered at various temperatures were analyzed using SEM and are compared in Figure 11. No delamination on the interface between the NEB20 disk and DuPont 951 disk was observed. It is because both the NEB20 tape and DuPont 951 tape have a certain volume fraction of glass. Based on the DuPont data sheet, the optimum sintering condition for DuPont 951 tape is 850°C for ~0.5 hours. Therefore, the glass transition temperature (T_g) of the glass in DuPont 951 tape should be lower than 850°C. At the interface between the two tapes, viscous flow most likely occurred even though this was the minimum sintering temperature, 850°C.

Severe warping was observed in a sintered laminate of two layers of NEB20 and DuPont 951 tape disks, which is attributed to a mismatch in the appropriate sintering temperatures and amount of shrinkage between the two tapes. In contrast, all of the sandwiches (NEB20/DuPont 951/NEB20) sintered at the various temperatures are flat. The flat structure is a result of the symmetric stacking sequence.

The thickness of both the NEB20 and DuPont 951 disks decreases with increasing sintering temperature. Quantitative analysis of the sintering shrinkage along the Z-direction was not performed in this study.

SEM micrographs of the sandwiches sintered at various temperatures (Figure 12) reveal some cracking in both the NEB20 and DuPont 951 layers. It is believed that the cracking is the result of mismatch in sintering shrinkage. During heating, at some temperature the DuPont 951 disk started shrinking prior to NEB20 densification. When the NEB 20 disk started shrinking, the DuPont 951 had been densified already, therefore, the thicker DuPont 951 disk would inhibit further shrinkage of the NEB disk along the X and Y directions. As a result, some cracking appeared in the NEB disks. Cracking in the NEB20 disk sintered at 850°C was minimized and the tensile stress located at the cracks in the NEB20, and applied on the DuPont 951 disk, was not large enough to cause catastrophic failure of the DuPont 951 disk. However, the cracking in the NEB20 disk became more severe with increasing sintering temperature and at the same time more tensile stress was applied on the DuPont 951 disk. As a result, severe cracking appeared on both NEB20 and DuPont 951 disks as shown in Figure 12.



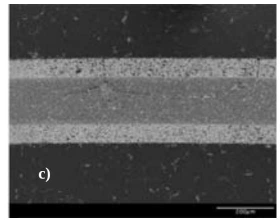


Figure 11: Microstructures of the sandwiches of NEB20/DuPont 951 sintered at various temperatures (a) - Sintered at 850°C, b) - Sintered at 900°C, c) - Sintered at 950°C).

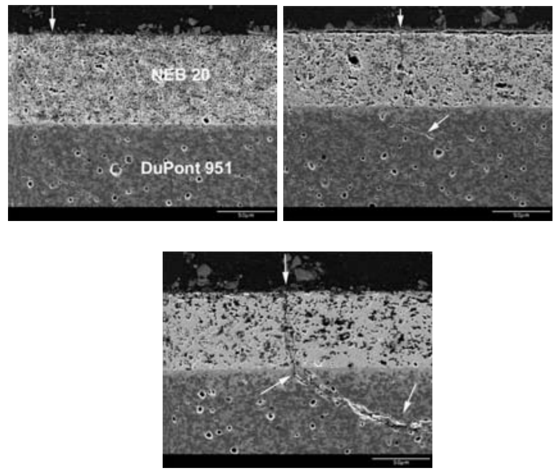


Figure 12: Microstructure of sandwiches of NEB20/DuPont 951 sintered at various temperatures (top left - Sintered at 850°C, top right - Sintered at 900°C, bottom - Sintered at 950°C).

Co-firing Multilayer Laminate of DuPont 951/NEB20 Tapes

The co-fired seven layer stacks of alternating NEB20 and DuPont 951 of the same diameter were almost flat as shown in Figures 13 and 14. The stacking order, in regard to the top vs. bottom surface of the as-cast tape, did not affect the overall flatness of the co-fired multilayer stacks of DuPont 951 and NEB 20 tape.

Figure 13: Multilayer specimens of NEB20/DuPont 951 laminated only by isostatic pressing at RT.

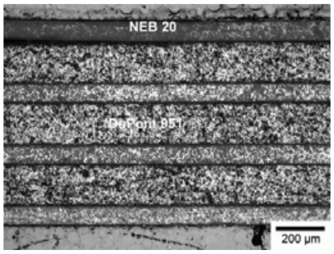


Figure 14: Cross section microstructure of the seven layer specimens of NEB20/DuPont 951 tape.

Dielectric Measurements

The capacitance of embedded NEB20 capacitors in DuPont 951 (Figures 1, 2) were measured at 1 kHz using a TENMA Model 72-6202 Multimeter (TENMA Test Equipment, Springbor, OH). Embedded capacitor structures were sintered at 850°C to 950°C for 15 to 90 minutes, however only measurements of samples sintered at 900°C for 60 minutes were taken at the time this manuscript was submitted. Fourteen capacitors (two rows of 7) of two different capacitor areas were measured on this substrate.

The average capacitance of capacitors with a designed active area of 0.027 cm^2 was 181.5 pF with a standard deviation of 59 pF. The average capacitance of capacitors with a designed active area of 0.033 cm^2 was 280.2 pF with a standard deviation of 27 pF. The ratio of the areas of the two capacitor sizes was 1.22 and the ratio of the measured values was 1.54. The dielectric constant of the NEB20 was previously reports as 1160 when sintered at 990°C[7]. The dielectric constant calculated for these capacitors for a sintered dielectric thickness of 0.003 inch is ~575 and ~730 respectively.

V. Conclusions

A custom, high K dielectric tape was cast using a formulation that was compatible, but not ideally suited with the processing conditions of DuPont 951. The high K NEB20 tape does have a sintering shrinkage mismatch with the DuPont 951 that can be managed by minimizing the area of the NEB20 embedded in the LTCC. Additional steps such as creating a balanced structure where the NEB20 is located within the LTCC stack is critical to maintaining a flat LTCC structure. Functional capacitors can be successfully embedded into DuPont 951 using the NEB20 high K tape. Sintering the embedded capacitors at temperatures below 990°C decreases the dielectric constant of the NEB20, however it remains high enough to be useful for high capacitance devices. Further characterization of the electrical performance of the capacitors is necessary to understand their applicability in RF and high speed digital LTCC packages.

VI. Acknowledgments

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VII. References

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