DEVELOPMENT OF LTCC-MATERIALS AND THEIR APPLICATIONS - AN OVERVIEW

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Abstract: The first Low Temperature Co-fired Ceramics (LTCC) tapes were developed more than thirty years ago. During the past decades, LTCC have spread around many different fields of application. This contribution gives an overview of the development of new materials used in combination with typical LTCC tapes, as well as on their application for new devices. Besides commercially offered materials like self-constrained tapes, high-k tapes or ferromagnetic tapes, newly developed LTCC compositions and their possible applications are presented and discussed.

Razvoj LTCC materialov in njihova uporaba - pregled

Kjučne besede: LTCC materiali, pregled, sintranje, debeloplastni prevodniki, uporaba

Izvleček: Keramika z nizko temperaturo žganja (LTCC - Low Temperature Co-fired Ceramics) je bila razvita pred več kot tridesetimi leti. V preteklih desetletjih se je njena uporaba razširila na mnoga področja. V tem prispevku podajamo pregled razvoja novih materialov, ki se uporabljajo v kombinaciji s tipičnimi LTCC folijami in aplikacije v novih napravah. Poleg komercialno dostopnih LTCC materialov, kot so na primer folije, ki se med žganjem ne krčijo, folije z visoko dielektričnostjo in feroelektrične folije bomo predstavili in opisali tudi aplikacije novih LTCC folij s posebnimi lastnostmi.

1 Introduction

The origin of LTCC development were needs of a multilayer technology comparable with already existed HTCC but at lower costs with possible application of high conductives that are well known from thick-film technology. Introduction of glass-ceramics compositions allowed decreasing of firing temperature and thus using almost all precious metal pastes (gold, silver, silver-platinum or silver- palladium). Due to cost issues, silver-based inks dominate.

During the past three decades, LTCC was expanded from rather small but profitable military and spacecraft markets into the radio frequency and automotive area. Moreover, besides already industry-established applications, new areas, most of them micro- or mesosystems were investigated. Flexibility and easy processing of unfired tapes as well as ability to construct three-dimensional ceramic devices in one single sintering process opened not only the door for new opportunities but also new requirements occurred.

The development of LTCC-technology can be described from different points of view. In this paper, three ways of LTCC development were considered. First and very important is the development of LTCC materials - tapes and pastes. Second, technological development like novel processing of tapes and pastes or joining of LTCC with other materials. And third, the development of LTCC applications besides radio-frequency applications, like different kind of physical and chemical sensors, chemical micro-reactors and others.

Detailed describing of above-mentioned kinds of LTCC developments would go beyond the scope of this paper. Nevertheless, we show you some, in our opinion, milestones in LTCC-development, a few very interesting ideas of LTCC applications, as well as some trends in R&D of LTCC.

2 Development of LTCC Materials and technology itself

Typical LTCC tapes mostly consist of glass-ceramics composites. Dependent on the ceramic powder content and the sintering mechanism, LTCC tapes can be divided into four groups /1/: "glass-free" ceramics (C), glass-bonded ceramics (GBC), glass ceramic composites (GCC) and glass ceramics (GC).

The low temperature sintering process is well known and well described in the literature (for example in /2/ or /3/. Typical for tape technology is the shrinkage of tapes during firing. The shrinkage differs dependent on material composition, lamination parameters, metallization coverage and firing parameters. For typical LTCC tapes, the shrinkage varies between 12-15% in x-y direction and 18-35% in z-direction. The shrinkage of the substrate, especially in x-y direction, can be disadvantageous for multilayer technology. Therefore, zero-shrinkage methods were introduced.

2.1 Zero-Shrinkage Process

Generally, it is not possible to avoid completely shrinkage of the tapes during firing, due to densification process and sintering viscosity /4/. At first, methods to join unfired LTCC and ceramics like Tape on Substrate (TOS) or LTCC-on-Metal (LTCC-M) have been introduced. These methods use classical LTCC tapes. The LTCC tapes are laminated on fired ceramics and the solid part prevents shrinking. Therefore, the ceramic part has to be bigger than the LTCC tapes.

In other words, these techniques describe ceramic elements with integrated LTCC, not LTCC structures itself. The newest of such system is called substrate-bonded tape system (SBTS) and was presented by ESL /5/. In the 1990's, the first zero-shrinkage method was introduced. Here, one should notice that not all methods called "zero-shrinkage" cause a zero-shrinking of the whole substrate. These methods are intended (and have been developed) for minimizing shrinking of the substrate in its plane directions.

2.1.1 PAS, PLAS

Pressure Less Assisted Sintering (PLAS) and Pressure Assisted Sintering (PAS) are very similar in use. To avoid shrinkage, in PLAS additional release tapes are introduced. Such release tapes consist of alumina and are laminated on the top and the bottom of the structure. During firing, the structure is "fixed" in x-y direction, since these sacrificial layers shrink at higher temperatures. After firing, the release layers are removed. This simple method has some disadvantages. First, it is impossible to print metallic layers on the top or bottom of the green sample - such elements can only deposited by post-firing. The zero-shrinkage effect is limited by thickness of the sample, i.e., if the sample contains more than 20 layers, the inner layers shrink like typical LTCC materials and some unwanted effects occur.

PAS means that the sample is fired in a special pressure furnace under a low uniaxial pressure, up to 50 kN /6/. Thus, the sample is fixed due to the applied pressure. Similar to PLAS, some shrinkage in homogeneities in z-direction can take place, but pressure adjusting can avoid it, even for samples with a higher number of layers.

Another advantage of PAS or PLAS is that almost all typical LTCC tapes can be used. The quality of zero-shrinkage effect (deformation and stress of structure, shrinkage anisotropy) depends on material chemistry, glass crystallization and microstructures /7/.

Both methods, originally proposed by DuPont have been established in industry a few years ago and at present almost all LTCC foundries offer PAS or PLAS for LTCC manufacturing.

2.1.2 Self-Constrained Sintering (SCS)

In spite of being in industrial application, both above-described methods have disadvantages. It is either technological limitation (PLAS) or additional expenses for new equipment (PAS). In 2002, Heraeus presented the self-constrained tape HL-2000 (a part of the so-called Heralock-System with Ag, Au and Ag/Pd conductors for co-firing) /8-10/. The SCS method requires neither special equipment nor additional release tapes. The self-constrained tape consists of three layers of different compositions. Both outer layers have the same chemical composition, whereas the inner layer, which serves as a locking layer or constraint layer, is different. It sinters at a higher temperature and therefore locks the compound. Laminating and firing conditions as well as electrical properties are very similar to classical LTCC tapes. The shrinkage is only about 0.2 % (shrinkage tolerance ± 0.02%) in x- and y-axes and 32 % in z-axis, respectively /1/. At present, Heralock is the only commercially available SCS system. In 2004, DuPont patented similar layer alignment with two primary layers and constraint layer inside /11/. This tape, however, is at the moment not commercially available. The constrained sintering of multilayer laminate with layers containing $CaO-B_2O_3-SiO_2$ glass and pure alumina has been reported as well. The x-shrinkage in this case was not as good as for HL2000, but, depending of alumina particles size, in the range of only 2.0% /12/.

2.2 Tapes for special purposes

One advantage of LTCC technology is its possibility to join tapes with different electrical properties. Very interesting are tapes with a high dielectric constant, e or *k* (high *k*-tapes) and ferroelectric tapes. The main issue in this respect is their compatibility with standard LTCC materials and the required low firing temperature below 1000C /13/.

2.2.1 High k-materials

Typical LTCC tapes have k-values in range of 5-9. This relatively low value is required in RF-applications to minimize parasitic capacitances and to increase signal speed. Although for the design of integrated capacitors this mentioned k-value is to low. In 2002, ESL brought a series of dielectric tapes with dielectric constant between 50-250 to the market (412xx-Series /14, 15/). It is an important feature that they can be joined with commercially available standard LTCC products of DuPont or Ferro. The dissipation factor is about 1% and temperature variations are comparable with X7R characteristics. From non-commercial products, different low-temperature tapes based on $CaZrO_3$-$CaTiO_3$ /16/, bismuth-titanate ceramics /17-21/ or zinc-titanate /22, 23/ were investigated. To decrease the firing temperature, different composition of lithium-borosilicate glasses were added to the dielectric composition. Middle dielectric constant compositions ($30 < k < 50$) were obtained with a low temperature coefficient (about 2 ppm/K /22/). Addition of zinc-borosilicate glasses to $BaTiO_3$ enabled to reduce the firing temperature down to 900 C with dielectric constant of about 1000 /19/.

2.2.2 Ferromagnetic materials

Ferroelectric tapes are generally used for buried inductors to increase the inductance of such structures and to save space. Here, ESL offers a magnetic tape with a permeability between 50 and 500. Its permeability value depends strongly on firing temperature. The highest permeability (μ = 1100) was achieved by firing at 1030 C /15/. Unfortunately, typical silver pastes cannot be used as coils material because due to the lower melting point of silver.

Other tested composites contain NiZn ferrites with a permeability of 500 /24/. To lower their sintering temperature, small amounts of AgO were added. Another group reported on tapes of hexaferrites of $BaFe_{12}O_{19}$ /25/ that are compatible with standard LTCC. Low firing temperature was obtained by adding reactive glasses. Here, however a much lower permeability of only about 20 was achieved.

2.3 Photoimageable pastes on LTCC

Thick-film photoimageable conductor and dielectric pastes on alumina have been known for many years (Fodel System from DuPont or KQ-Series from Heraeus) /26/. In the 1990's DuPont introduced Fodel pastes for LTCC /27/. First, silver pastes were presented followed in 2000 by a family of resistor pastes /28/. The advantage of this product is that the photolithographic process can be carried out on unfired materials. In contrast to typical photo-lithography, developing of the structure occurs with environmental friendly 1% Na_2CO_3 solution. At the moment, the minimum line width and space distance is between 30 - 40 µm for conductors /29-30/. First investigations on Fodel microresistors showed that it is possible to produce 50x50 μm^2 resistors with only a small dependence of the sheet resistance on resistor length providing also a good long-term stability /31-32/.

2.4 Development of technological processes

New applications require new technological processes. In the past years, a few achievements in the field of LTCC technology were reported. Here, in our opinion, interesting and promising are advances in laser patterning, construction of channels with the aid of fugitive layers or with by photolithography and integration of fired ceramic materials into unfired LTCC structures.

2.4.1. Laser structuring and application thereof

Structuring of LTCC tapes with laser is not new, but in the last years, new laser machines were introduced bringing improved resolution und repeatability in this field. Typically, for structuring of fired ceramics as well as for green tapes, CO_2-lasers were used. Such lasers can be characterized by a relatively large laser spot diameter and a high laser power. The typical Nd:YAG laser with a wavelength of 1064 nm was used in thick-film technology for resistors trimming. Cutting of ceramics was impossible in this case due to the very low absorption coefficient at this wavelength. However, it was possible to use standard Nd:YAG lasers to structure unfired LTCC tapes /33/. The cut quality was quite good, but in the case of very small structures far from perfection. The application of frequency-tripled Nd:YAG (355 nm) sources with a laser beam diameter of 20 µm could increase resolution and could minimize structure dimensions. Thus, it was possible to cut 50 µm channels in unfired tapes or vias with diameters of 50 µm /34/.

In the case of hot-plate gas sensors, hot-plate beams were as narrow as 260 µm /35/. Another laser application can be the construction of microcapacitors /36/, inductors /37/ and microresistors /38/.

2.4.2. Constructing of channels

The construction of channels in LTCC structures is a suggesting and relatively easy task. The tapes are cut prior to the lamination process. After stacking, lamination, and firing, a three-dimensional channel structure is created. The key-issue by the channel construction (especially by wide channels and cavities) is the sagging of top layers during lamination and/or during the firing processes. To avoid this phenomenon, different methods were proposed. The most popular one is to fill channels or cavities with graphite or carbon black /39-41/. Thus, flat membranes (up to 10 mm diameter) can be fabricated using a ne-sized graphite powder. However, in case of closed channels the depletion of air to oxidize the graphite and the lack of degassing can cause problems with the flatness of such elements.

Another possibility of small channel construction (down to 20 µm wide) is etching of partially fired LTCC ceramics /42/. Here, Riston foil (DuPont) is laminated on a partially fired LTCC structure and after exposure and developing, a channel structure is etched in HF-solution. After etching, the LTCC structure is finally stacked and fired.

2.4.3. Integration of fired ceramics into unfired LTCC structure

Electrical and thermal properties of LTCC tapes are, for some applications, not suitable. Therefore, a method to integrate a ceramic substrate with other thermal and/or electrical properties as standard LTCC tape was proposed /43/.

Generally, such elements are added (joined with the aid of adhesives or glasses) after firing the structure, because of the shrinkage of typical LTCC material. The introduction of mentioned SCS-tapes allowed joining of fired alumina prior firing of LTCC. Additional joining layers should improve adhesion and stability. Small modifications of this method enable integration of aluminum nitride or zirconia ceramics /44/.

3. Innovative LTCC applications

At the very beginning, LTCC applications were rather limited to military and spacecraft area. Due to excellent high frequency properties (low dielectric constant and low dissipation factor), LTCC materials found place in RF-frequency applications. Good thermal and mechanical stability extended the LTCC application area into the automotive sector.

The mentioned areas of LTCC applications are at present standard for LTCC technology. However, in the past ten years new LTCC applications emerged. This includes gas sensors /45-49/, physical sensors (pressure, force, temperature, etc.) /50-58/, chemical and biochemical devices /59-64/, fluidic structures with mixer and pumps /65-70/ and three-dimensional structures for different purposes /71-73/.

LTCC ceramics are very often used in applications where thermal treatment (heating) is necessary and therefore materials like FR4 or glass cannot be used. In contrast, LTCC technology allows integration of stable heaters and heating of the structure up to 700 C.

The increase of applications is also observed in wide considered bio-applications due to chemical stability, possibility of integration of conductive materials (electrodes) and easy structuring of unfired LTCC materials.

4 Conclusions - Future of LTCC Technology

In this paper, an overview over the development of LTCC technology during the past years is given. It is almost impossible to present all novel LTCC materials and devices in such a short summary. Nevertheless, in our opinion the presented examples can give a good impression on recent progresses in LTCC technology. A growing number of reported applications confirm the advantages of LTCC as well as that LTCC can be applied not only in RF-applications but is also suitable to be used in the microsystems field as well.

The progress in the material development eliminated the barriers and limitations of the first LTCC tapes. The Possibility of fine material processing allows creating small and reliable LTCC parts.

Here should be emphasized that LTCC cannot compete with mass product technologies like PCB. LTCC was not developed for this and nobody thought that LTCC would compete with PCB or polymer techniques, because generally ceramic technologies are expensive (materials high temperature processes, technology park, etc).

Nevertheless, the future of LTCC seems to be very promising, especially in areas where stability and reliability are important. Therefore, next LTCC applications should focus on areas, where using of low-cost materials is prohibitively impossible because of thermal, mechanical or chemical requirements. Thus, LTCC can achieve not really a big but a really profitable market niche.

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