TECHNOLOGY OF EMBEDDED STRUCTURES IN LOW TEMPERATURE CO-FIRED CERAMICS (LTCC)

PHD THESIS

Eszter Horváth

Head of the department: Prof. Dr. Gábor Harsányi

Supervisor of the work: Prof. Dr. Gábor Harsányi

Budapest University of Technology and Economics

Department of Electronics Technology

2012

Motivations and aims of the research

Nowadays, the high reliability, miniaturized electronics applications with high integrity (containing layer resistor and capacitor), ceramic or glass-ceramic substrates are often used because of better mechanical properties than the conventional plastic printing wiring board (PWB). The glass-ceramics (also known as **L**ow **T**emperature **C**o-fired **C**eramics, briefly LTCC) substrate is a dominant one in this area compared to the high temperature co-fired pure ceramics. LTCC substrates are currently used in aerospace industry, power electronics, some medical applications (hearing aids, pacemakers), armaments industry, automotive industry (Anti-lock braking system, Electronic Stability Program) and certain telecommunication (wifi and bluetooth) devices. LTCC is often used in microsystems due to its excellent physical and chemical properties (e.g. chemical inactivity, hermecity, similar thermal coefficient to silicon, high temperature stability). In case of chemical and biosensors, it is an important issue to be able to obtain information from and handle the smallest possible sample, because often we have only very small amount of sample. Therefore, LTCC technology – which is suitable to realize 3 dimensional structures – is often utilized as the substrate for sensors and microfluidic devices, and also provides the opportunity to integrate the fluidic part and the electronics module into one unit. The major part of electronics is possible to be buried in the substrate using internal wiring, buried capacitors, inductors and resistors, which results in significant size reduction. Screen printing is the layer deposition technique to realize surface and buried passive components in LTCC technology, which is one of the easiest and but cheap solution. However, the properties of passive components are highly affected by the printing quality, so great attention should be paid to select the appropriate printing parameters and squeegee.

The key step of realizing microfluidic components in LTCC is channel formation. Earlier the most important steps of the fluidic device's production like lamination and co-firing were optimized empirically, however the physical-chemical background of these processes wasn't investigated. The channel formed in the multilayer substrate is created by aligning together and laminating the layers containing the cut channel profiles. During lamination the pressure can cause sagging of the channel covering layers. To prevent this phenomenon the channel is filled with a so-called sacrificial volume material (SVM) before lamination. Afterwards the multilayer substrate is co-fired – the organic materials evaporate and decompose from the glass-ceramic substrate and the glass matrix melts and slurs. This results in shrinkage and structure solidification of the substrate. Using sacrificial volume materials that can be co-fired is simpler, because it does not have to be removed before co-firing but the suggested heating profile has to be modified to let the SVM completely decompose without damaging the structure.

Based on reviewing the literature it was found out that the aforementioned phenomenon in the course of lamination, screen printing and co-firing were examined mostly empirically or using excessive simplifications. Therefore this research is concentrated on these technological steps of the process of LTCC based embedded structure production.

Open issues of the research field

One of the main problems of screen printing is the short lifecycle of the screen, which can be even shorter using poorly adjusted parameters. This technology is being optimized from the 1960's. After the first empirical parameter optimization methods [1] mathematical models appeared [2, 3], but they didn't take into consideration the flexibility of the screen nor the effect of the geometry. Researches have been made to improve the geometry of the squeegee, but the problem cannot be addressed without knowing the geometry of the screen. Another important process parameter of screen printing is the off-contact distance – choosing it appropriately the adhesion and overstrain of the screen can be avoided.

The critical point of the technology of LTCC based embedded structures is lamination, where the laminating pressure is decreased compared to the prescribed one to avoid deformation [4, 5]. This way although it results in less channel sag, the probability of delamination between the layers is increasing. Applying adhesives between the layers can reduce delamination effects [6], but using this method gives rise to further problems e.g. if the adhesive isn't applied evenly or doesn't have the appropriate thickness. Furthermore the application is an additional technological step. In contrast using the conventional laminating pressure the channel covering layers are deformed [7]. To address this problem it is widely accepted to fill the channel with different sacrificial volume materials before lamination and examine the rate of sag, but so far the geometry of the channel wasn't taken into consideration. Furthermore the channel sag was only examined using cross-sections and this way the delamination couldn't be discovered for the whole substrate area. In the literature even nowadays trial-end-error optimization methods are being used for determining the technological parameters for realizing embedded channels with different geometries. To fully describe the lamination process the mechanical behavior of the substrate and the sacrificial volume material – in this case starch – is needed. Although LTCC is being used for about 30 years, the raw glass-ceramic substrate wasn't examined yet for mechanical stress. Many investigations were made to determine the physical parameters of starch because it is an important raw material of the chemical and the food industry as well. Several types of starch are commercially available, which have different grain structures thus different physical, chemical and mechanical properties. One of the most important raw materials of pharmaceutical production is starch, but only its volume change was examined due to pressure and die compaction [8-11]. These examinations are not sufficient to determine the plasticity parameters of starch, which is necessary to describe the deformation during lamination. In case of granular materials additional mechanical parameters can be determined with the so-called Jenike shear test machine: cohesion and internal friction angle. To provide the yield criterion the amount of air contained by the loose structured starch has to be also known.

Another important aspect of the technology is the adjustment of the co-firing heat profile. The choice of sacrificial volume material plays an important role here as well. Only materials that completely devolatilize during burnout and don't react chemically with the raw glass-ceramic can be used. The gases evolving from the decomposing sacrificial volume material can damage the structure. Decreasing the tangent of the heating profile the quantity of evolving gases per time unit can be reduced. The evolving gas quantity of certain (carbon based) sacrificial volume material can be controlled by modifying the inert gas/oxygen ratio of the burnout atmosphere [8]. However when the LTCC substrate contains embedded channels, the decomposition properties of the sacrificial volume material have to be taken into consideration because the sudden evaporating portions of the material can damage the channel structure. Without using a kinetic model and verifying it the results for parameter optimization won't be adequate.

Aims and methods of the research

Reviewing the open issues and main deficiencies of the discussed research areas, the following goals were set:

- increase the life expectancy of the screen while keeping the printing quality the same;
- optimize the geometry of the squeegee to reduce the stress in the screen;
- develop a non-destructive inspection technology which gives full details about the laminate and the delamination of the structure, and based on this optimize the lamination parameters;
- determine the mechanical parameters of LTCC and starch;
- develop a model such that the appropriate sacrificial volume material (powder, solid material) can be chosen and the expected deformation can be determined given the channel geometry;
- optimize the heating profile adjustment based on the thermal analysis of the sacrificial volume material.

New scientific results

Thesis Group 1: the effect of the squeegee to the arising tension in the screen and the compensation of reduced screen tension by the modification of the off-contact distance.

Thesis I: using finite element strength analysis and measuring the deformation of the stainless steel screen with direct emulsion mask due to load I showed that by optimizing the shape of the squeegee the arising tension in the screen can be reduced to 50%.

For the analysis finite element model was realized, and the elastic parameters were determined using the modified Voigt expression. The screen tension was given as an x and y displacement, which corresponds to line stress of the pre-stress condition. The deformation of the screen was modeled for different loads (40-80 N) at 11 different positions and the results were verified by measurements. For the experiment an individual measurement setup was made, which is suitable to examine the deformation of the screen for different loads. This was followed by adopting the typical contact distance in the model and the stress distribution was calculated. Due to the conventional shape of the squeegee the stress concentrates around the edges. With the appropriate squeegee round off the stress in the screen could be significantly reduced (to half), so the life expectancy is increased.

Related publications with Thesis I: L1, L2, L7

Thesis II: in order to maintain the printing quality of the stainless steel screen with direct emulsion mask I have define the relationship between the screen tension and the contact distance for the screen tensions of 2-3 N/mm.

In the course of screen printing right after the squeegee passes on, the screen and the substrate separate overcoming the adhesion between the paste and the screen. The tension of the screen is decreasing due to the repeated screen printing, so the restoring force setting the screen into its original position is reduced. As a result, the printing becomes blurred, which can be corrected by increasing the contact distance. In the finite element model the contact distance between the screen and the substrate was determined for reduced screen tensions in order to reach constant restoring force. My model specifies the correct distance between the screen and the substrate for different screen tensions to eliminate smearing effects.

Related publications with Thesis II: L1, L2, L7

Thesis Group 2: non-destructive testing method supplemented with image processing for optimizing the lamination parameters of LTCC substrate containing embedded channel.

Thesis III/a: I have established a new testing methodology with the concept of delamination rate (the ratio of the delaminated and the substrate area) to optimize the lamination parameters, which can be used to quantitatively qualify the degree of delamination and the method is proved by tests as well.

I have developed an acoustic microscopy based non-destructive test method supplemented with image processing technique to evaluate the lamination process of LTCC substrate containing buried fluidic channel. The image processing algorithm was implemented in Matlab. I have introduced the concept of delamination rate, which is the ratio of the delaminated and the substrate area. Evaluating the microscopic images by this algorithm and determining the delamination rate the degree of delamination can be quantitatively qualified.

Related publications with Thesis III/a: L3

Thesis III/b: using the calculation method of delamination rate the delamination can be apparently avoided in the test range (temperature: 40-70 °C, pressure 10-20 MPa, time: 5-15 minutes) using the lamination pressure of 20 MPa and a 5 minutes lamination time.

The testing method formulated in thesis III/a was applied for non-destructive quantitative qualification of lamination process of LTCC substrate containing buried fluidic channel. Using this method it was proven that the quality of the lamination depends on the applied pressure, but the lamination time does not affect significantly the global delaminated area. Taking into account the previous findings the lamination parameters of LTCC substrate containing buried structure were determined.

Related publications with Thesis III/b: L3, L8

Thesis IV: a finite element model was created to determine the maximum value of channel sag for channels with higher than 1:10 (width:length) ratio, and using this value as a threshold the appropriate type (solid or powder) of sacrificial volume material to be applied can be selected.

A finite element mechanical model was developed to describe the geometrical distortion of the SVM filled fluidic channel in LTCC substrate due to lamination process. The required material parameters of LTCC and starch (sacrificial volume material) were determined by compressive tests and shear box measurements. The stress-strain relationship of starch is nonlinear, the amount of strain is dependent of the hydrostatic stress. The plastic deformation of starch was described by the Drucker-Prager model. The stress-strain function of LTCC determined by the measurements was added to the finite element model, Von Mises plasticity criteria and isotropic hardening model were applied. Based on this model the features of the optimal sacrificial volume material used in LTCC containing embedded channel were determined considering the width of the channel and the applicability of the sacrificial volume material. The model was validated by experiments.

Related publications with Thesis IV: L4, L5, R1

Thesis V: theoretical model was made to describe the decomposition of starch used as SVM in LTCC substrate containing embedded channel, in order to determine the length and rate of deceleration period to be inserted into the heating profile of LTCC structure to restrict the intensity of gas evaporation to avoid channel damage, and the applicability of the modified thermal profile was proven up to 45 mm channel length.

According to the model the decomposition of starch and pressure generation occurs together. Evaluating the measurement results of thermal analysis it was determined that starch corresponds to the Arrhenius-type degradation process. The decomposition of the material showed consecutive characteristics, so the solution of differential equations was given using numerical method (Rosenbrock method). For the evaluation the method of least squares was used, which determined the kinetic parameters of starch, and then the results of the calculation were compared to the measurement results. Using the model I gave the length and slope of the deceleration phase, which has to be inserted into the heating profile of the LTCC. The new settings have limited the intensity of gas releasing, but have not unnecessarily slowed down the process and using the new profile the samples were not damaged due to the lower pressure.

Related publications with Thesis V: L6, L8, K1

The application of my results

In my Ph.D dissertation I have illustrated the application of my results with several examples. In this section I will summarize these.

In **thesis group 1 (thesis I and II)** the problems of screen printing (which is the applied layer deposition process in LTCC technology) are researched and it was suggested to modify the shape of the squeegee and change the off-contact distance to compensate for the screen tension reduction. The results discussed in the dissertation is being applied by Robert Bosch Gmbh. automotive electronics production company.

Applying the results of **thesis group 2 (thesis III/a and III/b)** the quality of lamination can be qualified quantitatively regardless of the lamination technology. Furthermore with the introduced non-destructive inspection method I verified that the bonding between the laminated LTCC layers has little lamination time dependence but significant lamination pressure dependence. Thus selecting the appropriate lamination pressure delamination can be avoided without the distortion of the geometric dimensions.

Applying the results of **thesis IV** the deformation of the channel geometry can be determined considering the deformation of the sacrificial volume material. Using the calculated distortion for channels with different widths and taking into consideration the applicability of the sacrificial volume materials I made a suggestion for choosing the appropriate type (powder or solid material) of sacrificial volume material.

In **thesis V** I used thermal analysis methods to inspect the decomposition of starch. Applying the appropriate mathematical model I partitioned the measured DTG curves into the partial DTG curves of a consecutive decomposition. The kinetic behavior of the material was investigated thoroughly and the applicability of the model was validated. Based on this I modified the applied heating profile used for LTCC sintering by inserting a deceleration phase to limit the decomposition of starch. Thus the most intensive gas releasing phase can be extended without unnecessarily slowing down the other parts of the heating profile.

Related publications with my theses

Reviewed journal articles:

[L1] **E. Horváth**, G. Hénap, Á. Török, G. Harsányi: „Mechanical Modelling and Life Cycle Optimisation of Screen Printing", Journal of Theoretical and Applied Mechanics, vol 50, no. 4, 2012, IF 0.264

[L2] **E. Horvath**, G. Henap, G. Harsanyi: Materials and Technological Developement of Screen Printing in Transportation; International Journal for Traffic and Transport Engineering, vol. 2, no. 2, 2012, 133-141 o.

[L3] **E. Horváth**, A. Török, G. Harsányi: „Design And Application Of Low Temperature Co-Fired Ceramic Substrates For Sensors In Road Vehicles", Transport, IF2.552

[L4] **E. Horváth**, G. Hénap, G. Harsányi: „Finite Element Modeling of Channel Sag in LTCC", Microelectronics Inrenational, Vol. 29, Iss. 23, pp. 145-152. IF 0.468

[L5] **E. Horváth**, G. Hénap, A. Torok, G. Harsányi: „Mechanical characterization of glass-ceramics substrate with embedded microstructure" Journ of Mat Sc-Materials in Electronics, vol. 23, iss. 12, 2012, p. 2123-2129, IF 0.927

[L6] **E. Horváth**: Thermal analysis of starch for realizing embedded channel in low temperature co-fired ceramic, Journal of Thermal Analysis and Calorimetry (under review) IF 1.604

[L7] **E. Horvath**; P. Ficzere; G. Henap; A. Torok; G. Harsanyi: Optimisation of computer aided screen printing design, Journal of Computer-Aided Design (submitted for publication) IF 1.542

[L8] **E. Horváth**, G. Harsányi: „Optimization of Fluidic Microchannel Manufacturing Processes in Low Temperature Co-Fired Ceramic Substrates", Periodica Polytechnika - Electrical Engineering, 54/1-2, 2010, 79–86 o.

Referred conference papers:

[R1] **E. Horváth,** G. Harsányi: "Modeling and Simulating Lamination of LTCC Substrate Containing Embedded Channel", 34rd International Spring Seminar on Electronics Technology, IEEE-ISSE2011, Tatranská Lomnica, Szlovákia, 2011.05.11-2010.05.15., 409 - 413. o.

Not referred conference papers:

[K1] **E. Horváth**, G. Harsányi: "Realizing Fluidic Microchannel In Low Temperature Co-Fired Ceramic Substrate", Electronic Devices and Systems IMAPS CS International Conference Proceedings, Brno, Csehország, 2009, 227-234. o.

Not related publications

Reviewed journal articles:

[L9] **E. Horváth**: „Embedded Thick-Film Resistors Applied in Low Temperature Co-fired Ceramic Circuit Substrates", Periodica Polytechnica-Electrical Engineering, 2008, Vol. 52, Issue 1-2, 45-57. o.

Referred conference papers:

[R1] **E. Horváth**, A. Erényi, A. Géczy, and G. Harsányi: " Optimization of breaking processes in LTCC manufacturing", 17th International Symposium for Design and Technology in Electronic Packaging , IEEE-SITME2011 Conference Proceedings, Timisoara, Romania, 2011, 153 - 155. o.

Other publications:

[M1] **Horváth E.**: „Build up technológia és a zsákfuratok fémezésének vizsgálata" I. rész, *Elektronet*, 2008, XVII:(6) 57-58. o.

[M1] **Horváth E.**: „Build up technológia és a zsákfuratok fémezésének vizsgálata" II. rész, *Elektronet*, 2008, XVII:(7) 52-53. o.

[M3] **Horváth E.**: „Az üveg-kerámia hordozók technológiai problémái", *Elektronet*, 2009 XVIII: 31-33. o.

References

[1] D.R. Kobs and D.R. Voigt, Parametric dependencies in thick film screening. *Proc. ISHM* 18, 1970, p. 1–10.

[2] L. F. Miller, Paste Transfer in the Screening Process, *Solid State Technology*, 1969, p. 46–52.

[3] Dietrich E. Riemer, Analytical model of the screen printing process: part 2. *Solid State Technology* 9, 1988, p. 85–90.

[4] Karol Malecha, Leszek J. Golonka Three-dimensional structuration of zero-shrinkage LTCC ceramics for microfluidic applications *Microelectronics Reliability* 2009;49: p. 585–591.

[5] L.E. Khoong, Y.M. Tan and Y.C. Lam, "Study of deformation and porosity evolution of low temperature co-fired ceramic for embedded structures fabrication ", *Journal of the European Ceramic Society*,2008, **29**, no. 13. p. 2737-2745.

[6] Dominik Jurkow, Henryk Roguszczak, Leszek Golonka: "Cold chemical lamination of ceramic green tapes" *Journal of the European Ceramic Society* 2009 29:p. 703–709.

[7] Karol Malecha, Leszek J. Golonka: "Microchannel fabrication process in LTCC ceramics" *Microelectronics Reliability* 2008, 48, p. 866–871.

[8] Espinoza-Vallejos, P., Zhong, J.-H., Gongora-Rubio, M. R., Sola-laguna, L. and Santiago-Aviles, J. J., "Meso (Intermediate)-scale electromechanical systems for the measurement and control of sagging in LTCC structures" *Mater. Res. Soc. Symp. Proc.*, 1998, 518, p. 3–79.

[9] K. Kawakita, K., I. Hattori, M. Kishigami. J. Powder Bulk Solids Technol. 1, 1977, 3-8 o.

[10] Heckel, R. W. Density pressure relationships in powder compaction. *Transactions of the Metallurgical Society of AIME*, 1961, 221, 671-675 o.

[11] N.A. Armstrong and R.F. Haines-Nutt: „Elastic recovery and surface area changes in compacted powder systems", *Powder Technology*, Vol. 9, Issues 5-6, May-June 1974, 287-290 o.

[12] Jenike, A. W.: "Storage and Flow of Solids" *Utah Engineers Experimental Station, Bulletin 23,* University of Utah, 1964.

[13] Irgens Fridtjov, *Continuum Mechanics*, Springer, Berlin, 2008.

Acknowledgements

First, I would like to thank my supervisor *Dr. Gábor Harsányi*, who directed and supported my work as head of the department and provided the necessary background for my research.

I would like to thank *Dr. László Jakab*, who supported the necessary background for the research in Bosch.

Thanks to *András Szabó* (Robert Bosch Electronics Ltd.) for supporting my research.

I am grateful to *Dr. Ádám Török*, who professionally and humanly supported my research work.

I would like to thank *Gábor Hénap* (BME-MM) for the help in retrieving simulation results and graphical visualization.