Engineering aspects of multilayer piezoceramic actuators

V.A. Golovnin¹, I.A. Kaplunov², A.I. Ivanova², R.M. Grechishkin²

¹Research Institute ELPA, JSC, Zelenograd, Russia
²Tver State University, Tver, Russia

e-mail: rmgrech@yandex.ru

Abstract. With the increasing demand for multilayer ceramic chip components a full understanding of the co-firing of ceramics with metal electrodes becomes important. In the present work the processing of a piezoelectric monolithic actuator by stacking and cofiring Ag-Pd electrode tape cast layers was studied. The inter-diffusion and microstructure of the co-fired interface of PZT ferroelectrics and Ag-Pd metal electrode were examined by scanning electron microscopy (SEM) and energy-dispersive microanalysis. No strong structural distortions and interdiffusion were observed at the co-fired ceramic-electrode interface.

1. Introduction
Piezoelectric multilayer monolithic stacks (MLMS) are increasingly used in a great variety of high-precision actuators and microelectromechanical systems (MEMS) [1-3]. The application-specific adjustment of the working parameters of the MLMS is achieved by the choice of the piezoelectric material (mainly high-strain PZT ceramics), MLMS geometry, electrode material, etc. The microstructure of the MLMS is defined generally by the sintering process, though the result depends on the composition of PZT and electrode [4-8]. The volumetric ratio of the PZT/electrode interfaces increases inevitably with the miniaturization trend of the chip devices, so it is imperative to investigate the co-fired interfaces in greater detail. In the present work we study the cofiring process with the aid of scanning electron microscope (JEOL JSM 6510LV) and X-ray microanalysis (Inca+, Oxford Instruments).

2. Experimental
Fabrication of the samples included paste preparation, multiple printing and drying, isostatic compression, co-firing, cutting, electrical connection, poling and testing. 70%Ag-30%Pd powders with a liquidus temperature of ~1150°C served to form the inner electrodes.

Before firing green multilayer piezoelements were formed by ceramic powder layers (91.7±1 and 8.3±1 wt% (40 vol% of organic binder) with a thickness of ~50 μm interlaced with Ag-Pd paste with both polished layers ~5 μm thick. The specific surface of the PZT-46 powder was in the limits of 10000…11000 cm²/g.

At 160 °C the butyral resin decomposes releasing water and oil aldehyde, while butyl benzyl phthalate (SANTICIZER 160) flares at 199 °C. The weight loss during heat treatment at 380 °C amounts to 7.8 wt%; the weight loss process starts at ~90 °C (warm-up time 1000 s) and finishes at ~400 °C (warm-up time 4500 s).

The sintered microstructure was investigated with both mechanically polished or fractured perpendicular sections of the samples. The flexural deformation and hysteresis characteristics of
prepared actuators were measured in a quasistatic mode with the aid of a microscope. The thickness of the multilayer samples is defined by the number (typically 20) and thickness of the individual layers (~50 μm).

3. Results and discussion
The thermogravimetric analysis (TGA) (figure 1) shows that the organic components of green multilayers start to burn at temperatures as low as 50 °C, while the main burning out of the plasticizer and binder proceeds in the 200 to 400 °C range of temperatures. The differential thermal analysis (DTA) data (figure 1) are indicative of intense organic compound decomposition processes at temperatures up to 500 °C. In this view great care should be taken in the proper choice of slow enough heating rates of the green multilayer composite.

Figure 1. TGA and DTA curves of the green workpiece under temperature ramp of 800 °C / 2.5 hrs

Figure 2 shows a SEM micrograph of the multilayer sample surface in the region of partial delamination of the outer Ag electrode. It is seen that the underneath ceramic is dense and there are hardly any pores in the field of view. It should be noted that the porosity of the outer electrode is markedly higher than that of ceramics. The typical size of the pores is about 3 μm and they propagate deep into the metal (figure 3).

Figure 2. Multilayer sample surface in the region of outer Ag electrode partial delamination observed in back-scattered (BEC) and secondary electrons (SEI). Left parts of the images – exposed ceramics, right parts – silver electrodes
Figure 3. Pore and crystallite structure of the outer Ag electrode

Figure 4 shows the fractured profile of the multilayer sample in the region of the inner Ag/Pd/70/30 electrode. This figure reveals that the internal electrode is well defined. The metal is closely bonded to the ceramic and practically no pores are observed along the interface.

In the lower parts of figure 4 are given the results of local microanalysis of the composition of the composite in the regions of inner electrode and the interelectrode area.

Figure 4. SEM microstructure morphology of the internal electrode area (a) and EDS analysis (b) of the Ag-Pd electrode and of bulk material. Areas of interest are marked with rectangles
The microanalysis has shown that the atomic composition of the electrode after co-firing coincides with that of the initial Ag-Pd powder except for Pb traces observed in the electrode at distances ≤ 500 nm from the PZT/Ag-Pd interface.

Strain of the finished multilayer sample as a function of electric field is presented in figure 5.

Figure 5. Strain vs voltage hysteresis curve of a finished multilayer actuator. Fitting equations for the descending and ascending parts of the loop are

\[ y_1 = -0.0012x^2 - 0.3928x + 0.7282 \]

and

\[ y_2 = 0.0008x^2 + 0.2005x - 0.0082 \]

4. Conclusion

The process for the preparation of multilayer actuators was described starting from PZT and Ag-Pd individual layers produced by by tape casting. Uniform layers with minimal porosity well interconnected with the co-fired internal 90%Ag-10%Pd electrodes were obtained. The microanalysis has shown that the atomic composition of the Ag-Pd electrode after co-firing coincides with that of the initial powder except for Pb traces observed in the electrode at distances ≤ 500 nm from the PZT/Ag-Pd interface. Low-hysteresis multilayer actuator with a strain of ~30 µm with applied voltage of ~100 V is demonstrated.

References

[1] Heywang W, Lubitz K, Wersing W 2008 Piezoelectricity – Evolution and Future of a Technology (Berlin/Heidelberg: Springer)
[2] Kushnir U, Rabinovitch O Advanced piezoelectric-ferroelectric stack actuator 2009 Sens. & Act. A 150 102-9
[3] Zhu D, Beeby S, Tudor J, White N, and Harris N 2011 Improving output power of piezoelectric energy harvesters using multilayer structures Proc. Eng. 25 199-202
[4] Kühnlein T, Stiegelschmitt A, Roosen A, Rauscher M 2013 Development of a model for the sintering of PZT multilayer ceramics and their dielectric properties J. Eur. Ceram. Soc. 33 991-1000
[5] Galassi C, Camporesi G, Fabbri G, Costa A L, Roncarì E 2001 Processing of a multilayer bender type actuator J. Eur. Ceram. Soc. 21 2011-4
[6] Cao J L, Wang X H, Gui Z L, Li L T 2008 Structural investigation s of the co-fired interface of Pb-based relaxor ferroelectrics and Ag-Pd electrode Ceram. Intern. 34 2025-8
[7] Zuo R, Li L, Gui Z 2001 Interfacial development and microstructural imperfection of multilayer ceramic chips with Ag/Pd electrodes Ceram. Intern. 27 889-93
[8] Yao K, Zhu W 1998 Improved preparation procedure and properties for a multilayer piezoelectric thick-film actuator Sens. & Act. A 71 139-43