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Co-firing of PZT–PMS–PZN/Ag Multilayer Actuator Prepared by Tape-Casting Method

Abstract: Multilayer ceramic actuator composed of piezoelectrically active $0.90Pb(Zr_{0.48}Ti_{0.52})O_3$–$0.05Pb(Mn_{1/3}Sb_{2/3})O_3$–$0.05Pb(Zn_{1/3}Nb_{2/3})O_3$ (PZT–PMS–PZN) layers and electrically conducting silver metal layers were fabricated. Low-temperature sinterable PZT–PMS–PZN ceramics were used as the piezoelectric layers, and silver paste was used as the conductive inner-electrode layers. PZT–PMS–PZN powder, adding 1% CuO as a sintering agent, was prepared by a conventional solid-state reaction process. The green sheet was fabricated by tape-casting method. Silver inner-electrode was printed on the green-tape sheets and the sheets were stacked, warm-pressed, and sintered at 900°C for 4 h after burning out the organic additions. The sintered multilayer ceramics actuator shows distinct layers with few defects in the interfaces. The thicknesses of the piezoelectric and conducting layers are about 40 and 6 μm, respectively. The multilayer PZT–PMS–PZN actuator, composed of 60 active piezoelectric layers, presents a longitudinal displacement of about 2.7 μm at an applied voltage of 80 V, approximately 0.1% of the total thickness of itself.

Keywords: multilayer actuator, PZT–PMS–PZN, tape casting, co-firing, piezoelectric

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Introduction

By utilizing the converse piezoelectric effect of piezoelectric ceramic materials, deformation can be produced in piezoelectric actuator and controlling displacement and force can be achieved. This displacement and force output also has several merits such as high control precision, fast response, good linearity and large generative force (Jaffe et al. 1971; Lee et al. 2009). On account of the excellent performances of piezoelectric actuator, the applications have been largely broadened in lots of fields, including electronic industry, communication and medical science (Xu 1991). Usually, monolayer piezoelectric actuator needs high applied voltage (above 200 V) and the electric-field-induced displacement is less than 0.1% in polycrystalline materials, although the displacement increases up to 1% in single crystals (Park and Shrout 1997), which cannot meet the current requirements of using in mechanical electronics components with miniaturization, integration and high performances due to the high applied driving voltage. In order to overcome these drawbacks, a multilayer-ceramic actuator is generally indispensible, in which piezoelectric layers and inner-electrode layers stack alternately to build a parallel structure series in machinery and electricity.

On the other hand, low-temperature co-fired ceramic (LTCC) has been an effective and reliable method to fabricate multilayer piezoelectric ceramic actuators (Li et al. 2001). In the process, the thin ceramic sheets, with thickness below 100 μm, can be prepared by tape-casting technique. When co-firing with inner-electrode layers, ceramic layers sinter each other closely. However, only precious metals such as Pt and Pd can be used as internal metallic electrode materials on account of a great amount of piezoelectric ceramics having a sintering temperature generally above 1,100°C (Zuo et al. 2002). These inner-electrode materials are expensive and show instable chemical property in oxygen atmosphere. A feasible way is to decrease sintering temperatures considerably, so that other economical and stable materials, such as silver, can be used as the inner electrodes.

The quaternary piezoelectric ceramic system $Pb(Zr_{0.48}Ti_{0.52})O_3$–$Pb(Mn_{1/3}Sb_{2/3})O_3$–$Pb(Zn_{1/3}Nb_{2/3})O_3$ (PZT–PMS–PZN) has outstanding electromechanical properties, which can be applied in piezoelectric actuators and transformers (Yang et al. 2005, 2006). According to the

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previous research, when 1% CuO was added into PZT–PMS–PZN system, the ceramics can be sintered fully dense at 900°C and present good electrical properties (Wan et al. 2010). Based on the result, this paper demonstrates the preparation of multilayer piezoelectric actuators, following the displacement output tests and the microstructure observations of the multilayer stacks.

Experimental Procedure

The ceramic powder with a composition of 0.90Pb$(Zr_{0.48}Ti_{0.52})O_3$–0.05Pb$(Mn_{1/3}Sb_{2/3})O_3$–0.05Pb$(Zn_{1/3}Nb_{2/3})O_3$ was prepared by a conventional solid-state reaction process. PbO, ZrO_2, TiO_2, MnO_2, Sb_2O_3, ZnO and Nb_2O_5 of high purity of 99.9% were used as the starting materials. The oxides were mixed using conventional ball milling for 24 h in a nylon jar with zirconia ball and anhydrous ethanol as the milling media. After ball milling, the mixture was dried in the drying oven and subsequently calcined at 850°C for 4 h. About 1% CuO additive was added to the calcined powder and then remilled for 24 h.

Piezoelectric ceramic sheets were fabricated by tape-casting slurry containing a mixture of powers, solvent, dispersant, organic binders, and plasticizer. The powers were first mixed with solvent comprised of methyl ethyl ketone and anhydrous ethanol with a weight ratio of 7:3 and dispersant of corn oil for 4 h by ball milling. Then, polyvinyl butyral (PVB) using as the binder and dibutyl phthalate (DBP) using as the plasticizer were added to the mixed slurry, and then ball milled again for 8 h to obtain castable slurry (Lee et al. 2009; Choi et al. 2008; Yoo et al. 2007). The green sheets were tape-casted by a doctor blade method with thickness of 60 μm and dried at room temperature. Then silver paste was printed on the green sheets as inner electrodes, the thickness was controlled to 6 μm, through diluting silver paste and improving screen printing process. The tapes were warm-pressed to stack up to desired layers at 50°C, and then the multilayer stacks were heated to 550°C with Al_2O_3 packing power at a slow heating rate of 15°C/h for burning out the organic binder, effectively preventing deformation and cracking. The samples were sintered at 900°C for 4 h in a sealed alumina crucible. To investigate the properties of sintered multilayer actuator, external electrodes were prepared by applying a thin silver paste to both lateral sides of the actuator to connect the inner electrodes, followed by heat treatment at 700°C for 10 min to provide robust electrodes (Yoon et al. 2004). At last, the samples were poled under electric field of 3–4 kV/mm at 120°C for 20 min in the silicon oil bath.

The phase structure of the sintered samples were determined by X-ray diffraction (Bruker D8 Advanced, Cu Kα radiation, $\lambda = 0.15418$ nm). The electric properties of the samples were measured by Precision Impedance Analyzer (Agilent 4294A, USA). The microstructure of the multilayer ceramics was observed by a field emission scanning electron microscope (Hitachi SU8010, Japan). The field-induced displacement was monitored by using the laser displacement sensor (KEYENCE LK-H020, Japan).

Results and Discussions

XRD diffraction pattern of PZT–PMS–PZN layer is shown in Figure 1. When the PZT–PMS–PZN, adding sintering agent, is sintered at 900°C for 4 h, pure perovskite peaks are exhibited without any other second phase. Despite the low sintering temperature used (900°C), the piezoelectric ceramic also presents good properties: $d_{33} = 320$ pC/N, $d_{31} = 170$ pC/N, $k_p = 0.56$, $k_{31} = 0.29$, $Q_m = 942$, tan $\delta = 0.6\%$, $\varepsilon_{33}{}^T/\varepsilon_0 = 1{,}284$ and $T_c = 300°C$. Although the properties drop a bit compared to those of the monolayer ceramics sintered at higher temperature (Yang et al. 2005, 2006), the measured piezoelectric properties still indicate good applicability for use as a high-power actuator.

Figure 2 exhibits SEM images of the sintered multilayer actuator. As shown in Figure 2(a), it is composed of the ceramic layers interpolated with the inner-electrode

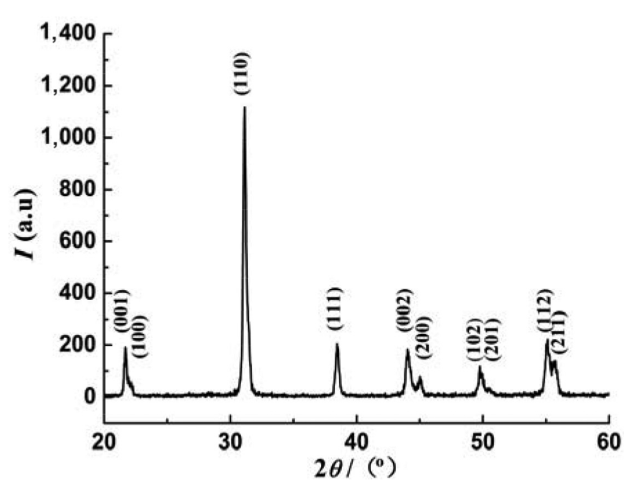

Figure 1: XRD pattern of PZT–PMS–PZN ceramic sintered at 900°C.

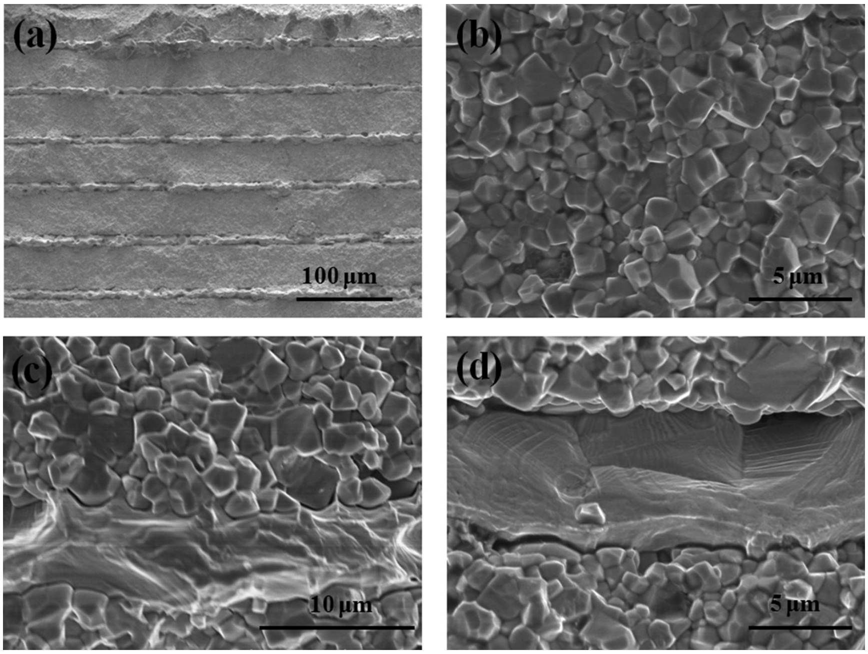

Figure 2: Cross-sectional micrographs of co-fired multilayer PZT–PMS–PZN/Ag: (a) the overall structure, (b) piezoelectric ceramics PZT–PMS–PZN layer, (c) interface and (d) inner-electrode layer.

layers. In addition, in order to avoid electrical short circuits, the inner-electrode layers are connected alternatively to the left and right lateral surfaces. The bulks are fully dense without large holes and cracks. The thickness of the piezoelectric ceramic layers is uniform, and about 40 μm after sintering. The grain size is about 1.5 μm with a fine, uniform and equiaxial structure, as shown in Figure 2(b), which also indicates that PZT–PMS–PZN is well sintered at 900°C. The multilayer piezoelectric ceramic actuators have very high demands for interface bonding between ceramic layers and inner-electrode layers, because of their often working under high electric field and high stress. In this paper, by choosing suitable silver paste, adjusting screen printing process and optimizing pressure of lamination, the interface between PZT–PMS–PZN and silver is distinct with few defects as shown in Figure 2(c). As we know, the silver electrode has no piezoelectricity, so a thinner electrode contributes to the total thickness decreasing of the multilayer stack. However, a thin electrode also affects the continuity of electric conduction after co-firing. In this work, the thickness of the inner electrode is about 6 μm, as shown in Figure 2(d).

Meanwhile, the end parts of the inner electrode of the actuator were concerned in this paper, especially. These two parts need to conduct with the external electrodes and will be subjected to a high voltage, so it is easily fragile. In this work, in order to improve the quality of the end part of the inner electrode, the end part was polished with sandpaper before preparing the external electrode. As shown in Figure 3, the straight, continuous inner electrodes and the flat end face with few defects successfully reduce the electric field concentrations and improve the conductivity.

Figure 4 shows the prepared multilayer actuator composed of 60 active PZT–PMS–PZN layers and 61 conducting Ag layers, whose overall shape is 7 mm × 5 mm × 2.8 mm. When applying a voltage in the poling direction, the longitudinal displacement of the multilayer actuator

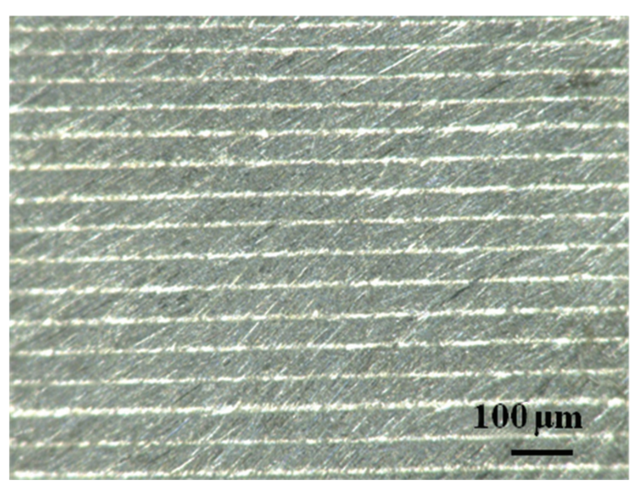

Figure 3: Optical micrograph of polished cross section for the end part of the inner electrode.

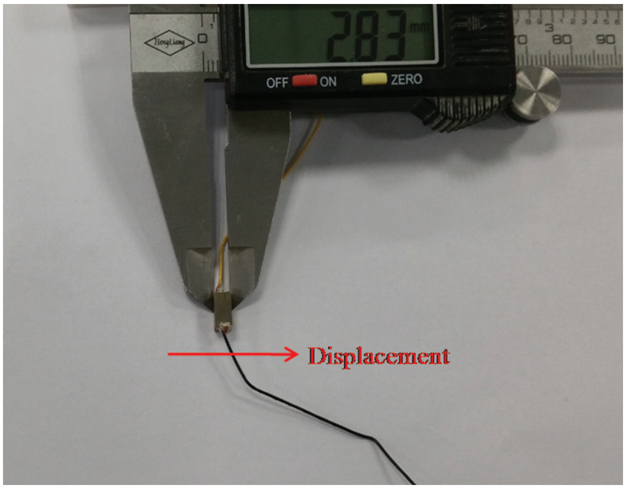

Figure 4: Photograph of the prepared multilayer actuator.

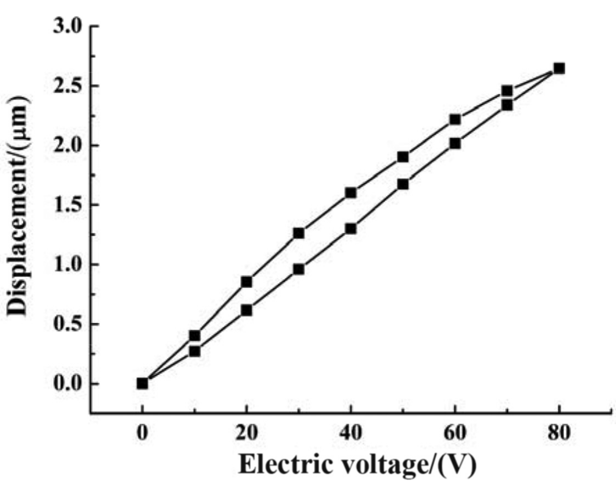

Figure 5: Longitudinal displacement of the multilayer actuator.

was measured, as shown in Figure 5. We can know that the displacement increases with the driving voltage, presenting a good linearity, and when the voltage is 80 V, the displacement is about 2.7 μm, approximately 0.1% of the total thickness of the actuator. However, when the voltage reduces, the displacement doesn't change along the previous curve, producing a hysteresis. The intrinsic trait of piezoelectric materials should mainly be responsible to the nonlinearity hysteresis. It is known that the displacement introduced by applied electric field is due to the converse piezoelectric effect and ferroelectric effect in high electric field. The inverse piezoelectric effect is linear, while according to the turning and incomplete reversibility of non-180° domain, the

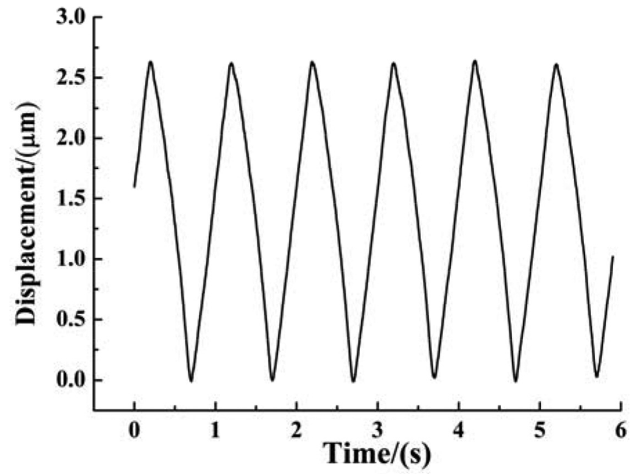

Figure 6: Dynamic displacement of the multilayer actuator.

ferroelectric effect exhibits nonlinear and hysteretic phenomenon.

The dynamic displacement was also measured as shown in Figure 6. A triangle-wave voltage was applied with the frequency of 1 Hz and the voltage (peak to peak) of 80 V, and the displacement also presents a triangle-waveform with 1 Hz frequency, showing a quick and linear response to the driving voltage signals. In addition, the displacement amplitude is also 2.7 μm in each cycle, as shown in Figure 6, which complies with the displacement measurement under static voltage loading in Figure 5.

Conclusion

In summary, PZT–PMS–PZN with 1% CuO as a sintering agent and silver can be co-fired at 900°C with few defects. A multilayer actuator composed of PZT–PMS–PZN layers and inner-electrode Ag layers was successfully fabricated by tape-casting method. The displacement of the multilayer actuator, composed of 60 piezoelectric layers, is about 2.7 μm at a voltage of 80 V, approximately 0.1% of the total thickness of itself, and presents a good linearity.

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