

Structural and Electrical Properties of PZT(40/60)/PZT(60/40) Thick Films via a Screen-printing process

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Ferroelectric PZT heterolayered thick films were fabricated via an the alkoxide-based sol-gel method. PZT(40/60) and PZT(60/40) paste were created and alternately screen-printed onto Al_2O_3 substrates. A press-treatment was introduced to obtain a good densification of the screen-printed films. The porosity of the thick films decreased as the applied pressure increased, and thick films pressed at 0.6 ton/cm^2 showed a dense micro-structure with a thickness of approximately $76 \mu\text{m}$. The remanent polarization and coercive field increased as the applied pressure increased. The values for the PZT thick films pressed at 0.6 ton/cm^2 were $17.04 \mu\text{C/cm}^2$ and 78.09 kV/cm .

Keywords: PZT ceramics, ferroelectric, thick films, screen-printing, remanent polarization.

1. INTRODUCTION

Ferroelectric $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ (PZT) material is one of the most important electric ceramics materials for use in sensors, actuators and filters on account of its unique properties. Interest in applying PZT films to microactuators has increased recently. In most cases, films thicker than $10 \mu\text{m}$ are required to obtain a large force consistently, though the optimum thickness depends on the structure and substrate material of the actuator. Generally, PZT thick films are fabricated on substrates of materials such as Al_2O_3 and ZrO_2 using a screen-printing method. Sintering temperatures above 1000°C are required to fabricated dense thick films^[1,2]. The screen-printing method is especially useful for a high productivity and good cost performance and has played a pivotal role in bringing the films to the stage of commercial mass production. Although it would be highly desirable to screen-print other components such as capacitors, varistors or sensors, currently this cannot be done while maintaining the electrical properties of the corresponding ceramics. One of the main problems inherent to this technology is the lack of compactness of the screen printed layers^[3].

In this study, PZT heterolayered thick films were prepared using the screen printing techniques in which they were alternately screen-printed on high-purity alumina substrates using PZT(60/40) and PZT(40/60) pastes. A press-treatment of a green film printed onto the substrates was introduced to

obtain a good densification of screen-printed films without the use of an inorganic binder. Additionally, the structural and dielectric properties of the thick films were investigated for the possible fabrication of various transducers and electronic devices.

2. EXPERIMENT PROCEDURE

PZT(40/60) and PZT(60/40) powders with excess Pb-acetate 10mol% were prepared from Pb acetate trihydrate ($\text{Pb}(\text{CH}_3\text{CO}_2)_2 \cdot 3\text{H}_2\text{O}$), Zr propoxide ($\text{Zr}(\text{OCH}_2\text{CH}_2\text{CH}_3)_4$) and Ti iso-propoxide ($\text{Ti}[\text{OCH}(\text{CH}_3)_2]_4$) as the starting materials and 2-methoxyethanol ($\text{CH}_3\text{OCH}_2\text{CH}_2\text{OH}$) as the solvent using a sol-gel method^[4].

The screen-printable pastes were prepared by kneading ground PZT powder with 30 wt% of an organic vehicle (Ferro B75001) in a non-bubbling kneader (NBK-1, Kyoto Electro.). Pt bottom electrodes were screen-printed onto a high-purity alumina substrate ($15 \times 15 \times 1 \text{ mm}^3$). PZT(40/60) paste was screen-printed onto the substrates to form the first layer. These PZT(40/60) films were dried, and then the PZT(60/40) paste was thenscreen-printed and dried over the PZT(40/60) films to form a second layer under identical conditions. This procedure was repeated four times. After removal of the solvents, the screen-printed films were pressed at 0, 0.2, 0.4 and 0.6 ton/cm^2 using a hydraulic press. These heterolayered PZT thick films were sintered at 1050°C for 2h in PbO atmosphere. The crystalline structures of the PZT heterolayered thick films were analyzed by X-ray diffraction (XRD) with $\text{CuK}\alpha$ emission. The surface and

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cross-sectional microstructures of films were examined using scanning electron microscopy (SEM). The upper electrodes were fabricated by the screen-printing of Ag paste. After poling with a field of 30 kV/cm for 30 min at 120°C, the dielectric properties of the specimens were measured using an LCR-meter (ANDO 4301) at 1 KHz. Ferroelectric properties were measured using a ferroelectric tester (Radiant, RT-66A).

3. RESULTS AND DISCUSSION

Figure 1 shows X-ray diffraction patterns of the PZT heterolayered thick films printed on Pt/alumina substrates. PZT thick films show the typical XRD patterns of a perovskite polycrystalline structure without a preferred orientation. A

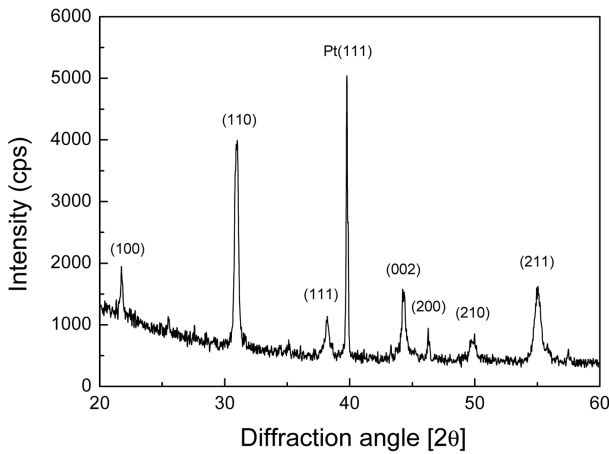


Fig. 1. X-ray diffraction patterns of the PZT heterolayered thick film.

pyrochlore phase was not observed in the images.

Figure 2 shows surface and cross-section SEM micrographs of the PZT thick films printed on Pt/alumina substrates under different applied pressures. The average grain size was approximately 1-2 μm. The rugosity and porosity of the thick films decreased as the applied pressure increased. Thick films pressed at 0.6 ton/cm² showed a dense microstructure with a thickness of approximately 76 μm.

Figure 3 shows the relative rates of the dielectric constant and dielectric loss for PZT thick films subject to a range of applied pressures. The relative dielectric constant increased and the dielectric loss decreased as the level of applied pressure increased. These properties can be understood in terms of the effect of the densification and decreasing porosity, as

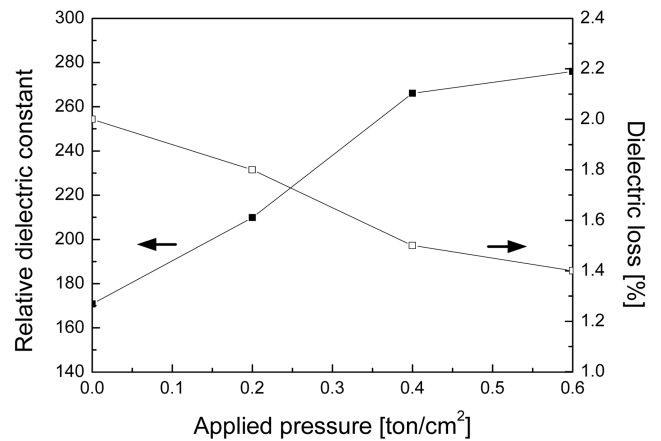


Fig. 3. Relative dielectric constant and dielectric loss of PZT heterolayered thick films as a function of the applied pressure.

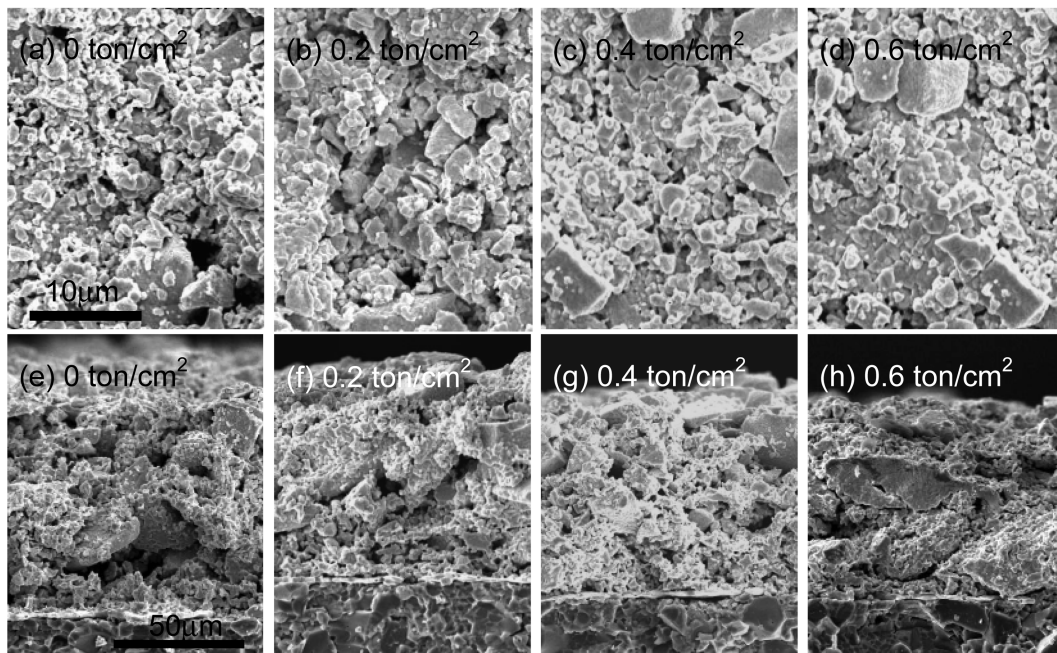


Fig. 2. SEM micrographs of the surface and cross-section morphologies in PZT heterolayered thick films under varying applied pressures.

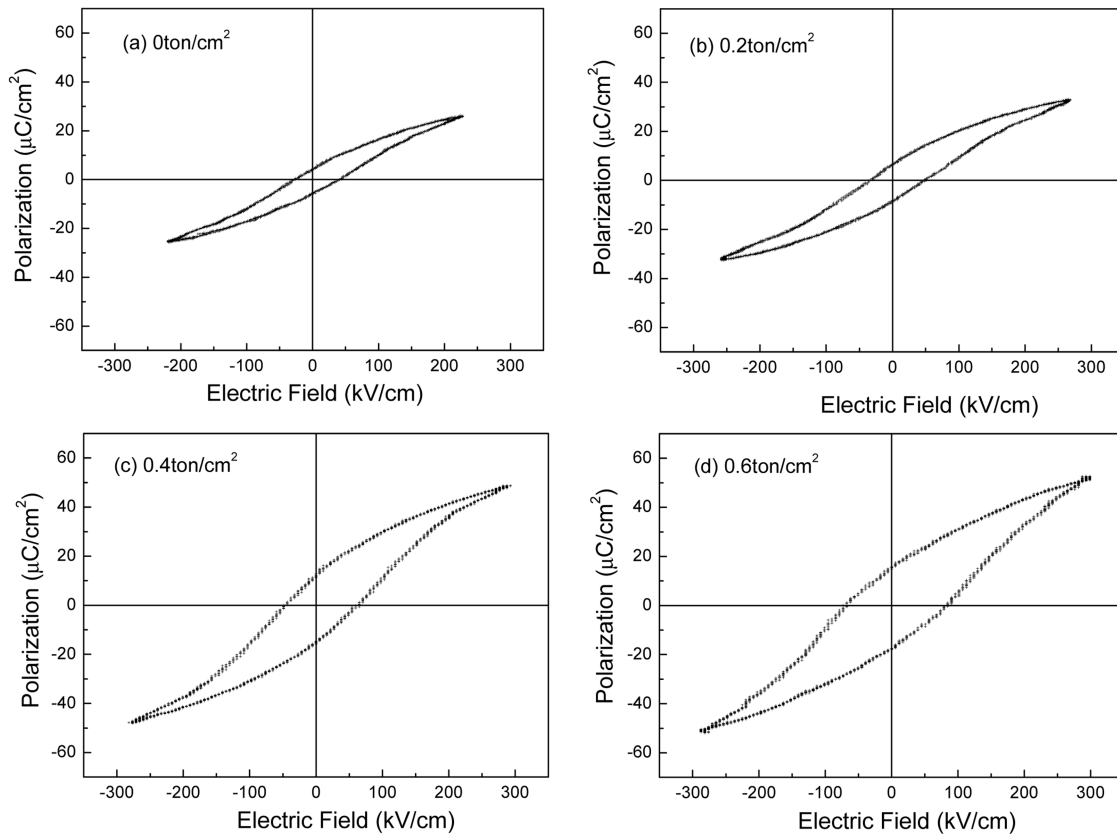


Fig. 4. P-E hysteresis loops of PZT thick films under varying applied pressures.

shown in Fig. 2. The relative dielectric constant and dielectric loss in PZT thick films pressed at 0.6 ton/cm^2 were 276 and 1.40%, respectively.

Figure 4 shows the P-E hysteresis loops of PZT thick films under various amount of applied pressure. The remanent polarization and coercive field increased as the amount of applied pressure increased. These values for PZT thick films pressed at 0.6 ton/cm^2 were $17.04\mu\text{C}/\text{cm}^2$, 78.09 kV/cm , respectively. Thus, better densification of the films is shown to be directly related to higher ferroelectric properties.

4. SUMMARY

In this study, PZT(40/60) and PZT(60/40) powders, prepared by using a sol-gel method, were mixed with an organic vehicle, allowing PZT heterolayered thick films to be fabricated via screen-printing techniques by alternately using PZT(40/60) and PZT(60/40) pastes. The effect of mechanical pressure on the electrical properties of the PZT heterolayered thick films was demonstrated. PZT thick films showed the typical XRD patterns of a perovskite polycrystalline structure without preferred orientation in the absence of a pyrochlore phase. The level of densification of the thick

films was increased as the applied pressure was increased. The relative dielectric constant and dielectric loss of the PZT thick films pressed at 0.6 ton/cm^2 were 276 and 1.40%, respectively.

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