Ferroelectric Properties of PZT Heterolayered Thick Films

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Abstract : PFerroelectric PZT heterolayered thick films were fabricated by the alkoxide-based sol-gel method. PZT(40/60) and PZT(60/40) paste were made and alternately screen-printed on the Al_2O_3 substrates. We have introduced a press-treatment to obtain a good densification of screen printed films. The porosity of the thick films were decreased with increasing the applied pressure and the thick films pressed at 0.6 ton/cm^2 showed the dense microstructure and thickness of about 76μm. The remanent polarization and coercive field increased with increasing applied pressure and the values for the PZT thick films pressed at 0.6 ton/cm^2 were 17.04μC/cm^2, 78.09 kV/cm, respectively.

Key Words : PZT, thick films, screen-printing, remanent polarization, coercive field.

1. Introduction

Ferroelectric Pb(Zr,Ti)O_3(PZT) material is one of the most important electric ceramics materials for use in sensors, actuators, and filters, because of their unique properties. Interest in applying PZT films to microactuators has been increasing recently. In most cases, films thicker than 10mm are required to obtain a large force effectively, though the optimum thickness depends on the structure and substrate material of the actuator. Generally, PZT thick films are fabricated on substrates, such as Al_2O_3 and ZrO_2, using a screen-printing method, and 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 brings the films to the stage of commercial mass production. Although it would be highly desirable to screen-print other components, such as capacitors, varistors, sensors, etc., currently this cannot be done while keeping 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 by 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. We have introduced a press-treatment of the green film printed on the substrates to obtain a good densification of screen printed films without inorganic binder. And the structural and dielectric properties of the thick films were investigated for fabricating various transducers and electronic devices.

2. Experimental

PZT(40/60) and PZT(60/40) powders with excess Pb-acetate 10mol% were prepared from Pb acetate trihydrate ($Pb(CH_3CO_2)_2 \cdot 3H_2O$), Zr propoxide ($Zr(OCH_2CH_2CH_3)_4$) and Ti iso-propoxide ($Ti[OCH(CH_3)_2]_4$) as the starting materials, and 2-methoxyethanol ($CH_3OCH_2CH_2OH$) as the solvent using the sol-gel method [4].

The screen-printable pastes were prepared by kneading the ground PZT powder with 30wt% of organic vehicle (Ferro B75001) in a non-bubbling kneader (NBK-1, Kyoto Electro.). The Pt bottom electrodes were screen-printed on the high purity alumina substrate (15×15×1 mm^3). The PZT(40/60) paste was screen-printed on the substrates to form the first layer. These PZT(40/60) films were dried, and then the PZT(60/40) paste was screen-printed and dried on the PZT(40/60) films to form the second layer under the same conditions. This procedure was repeated 4 times. After removal of the solvents, the screen printed films were pressed at 0, 0.2, 0.4, 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 CuKaemission. The surface and cross-sectional microstructures of films were examined using scanning electron microscopy (SEM). The upper electrodes were fabricated by screen printing the 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 the X-ray diffraction patterns of the PZT heterolayered thick films printed on Pt/alumina substrate.

PZT thick films showed the typical XRD patterns of a perovskite polycrystalline structure without preferred orientation and no pyrochlore phase is observed.

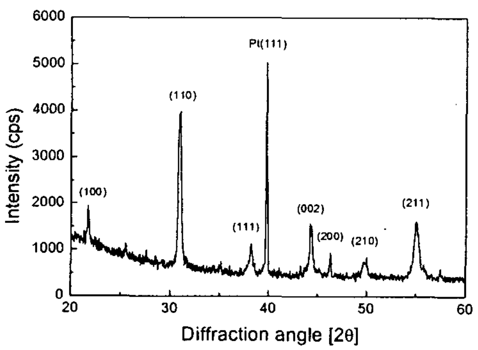

Fig. 1. X-ray diffraction patterns of the PZTheterolayered thick film.

Figure 2 shows the relative dielectric constant and the dielectric loss of PZT thick films with variation of applied pressure. The relative dielectric constant increased and the dielectric loss decreased with increasing applied pressure. These properties can be understood in terms of the effect of the densificationand the decreasing porosity. 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.

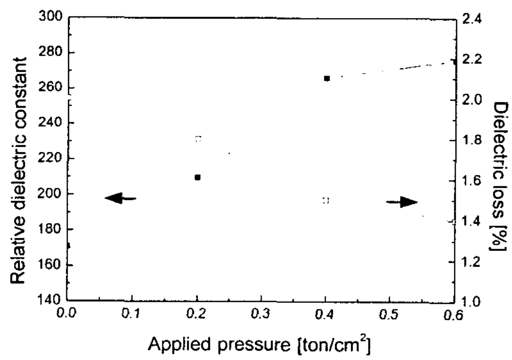

Fig. 2. Relative dielectric constant and dielectric loss of PZT heterolayered thick films as a function of applied pressure.

Figure 3 shows the P-E hysteresis loops of PZT thick films with variation of applied pressure. The remanent polarization and coercive field increased with increasing applied pressure and the values for the PZT thick films pressed at 0.6 ton/cm^2 were 17.04mC/cm^2, 78.09 kV/cm, respectively. Thus, the better the densification of the films, the higher is the ferroelectric properties.

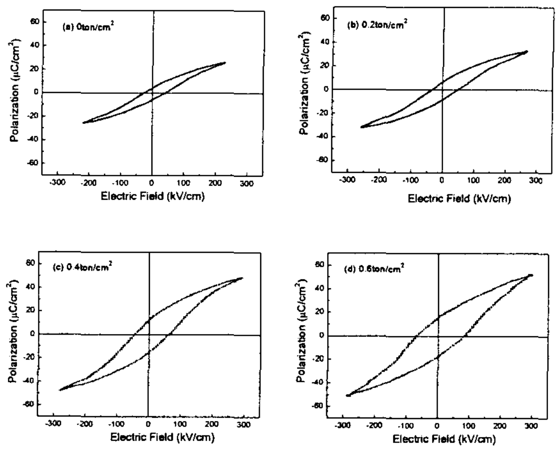

Fig. 3. P-E hysteresis loops of PZT thick films with variation of applied pressure.

4. Conclusion

In this research, PZT(40/60) and PZT(60/40) powders, prepared by using a sol-gel method, were mixed with an organic vehicle, and PZT heterolayered thick films were fabricated by screen-printing techniques by alternately using PZT(40/60) and PZT(60/40) pastes. The effect of mechanical pressure on the electrical properties of PZT heterolayered thick films has been demonstrated. PZT thick films showed the typical XRD patterns of a perovskite polycrystalline structure without preferred orientation and no pyrochlore phase is observed. The densification of the thick films was increased with increasing the applied pressure. 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.

Acknowledgments

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References

[1] J. F. Fernandez, E. Nieto, C. Moure, P. Duran, and R. E. Newnham: J. Mater. Sci. Vol. 30 (1995), pp. 5399

[2] T. Futakuchi, Y. Matsui, and M. Adachi: Jpn. J. Appl. Phys. Vol. 38(1999), pp. 5528.

[3] L. Simon, S. Le Dren, and P. Gonnard: J. Eur. Ceram. Soc. Vol. 21(2001), pp. 1441.

[4] S. G. Lee and Y. J. Shim: J. of KIEEME. Vol. 18 (2005). pp.550