Available online at www.sciencedirect.com
SciVerse ScienceDirect
Ceramics International 39 (2013) S541–S544
CERAMICS INTERNATIONAL
www.elsevier.com/locate/ceramint

Soft/hard PZT monolithic bi-layer composite actuator

Piyalak Ngernchuklin[a,*], Jungho Ryu[b], Chutima Eamchotchawalit[a], Dong-Soo Park[b]

[a]The department of Materials Innovation, Thailand Institute of Scientific and Technological Research (TISTR), 35 Mu 3 Klong Ha, Klong Luang, Pathum Thani, 12120, Thailand
[b]Functional Ceramics Group, Korea Institute of Materials Science, 797 Changwondaero, Seongsan-gu, Changwon, Gyeongnam 642-831, Republic of Korea

Available online 17 October 2012

Abstract

Lead Zirconate Titanate (PZT) is a well-known piezoelectric material which has been widely used for transducer, sensor and actuator applications. According to the compositional modifications near morphotropic phase boundary, PZTs can be classified into two types, soft PZT and hard PZT, which are distinguished by the piezoelectric properties. Therefore, the combination effect of the strain-E-field response of soft/hard PZT composite actuator has been much interested to be observed. In this study, two types of soft PZTs (PZT-610HD and PZT KP 60) and two types of hard PZTs (PZT-TISTR and PZT DPZ-HQ) were used. The ratio of the soft and hard PZT powders used to form bi-layer composite was designed for 1:1. The bi-layer composites were prepared by co-pressing and co-sintering to obtain the monolithic layer between the two layers. The displacement and electromechanical properties of all bi-layer composites actuators were investigated compared to the soft and hard PZT single layer counterparts. For an application of bi-polar E-field at 10 kV/cm, a maximum peak displacement (11 μm) was obtained from the bi-layer composite actuator prepared by 610HD-TISTR powders, while the displacement of soft and hard PZT single layer was shown at 1 μm and 0.4 μm, respectively. This can be discussed in terms of the sintering behavior as well as the ferroelectric properties of those soft and hard PZTs which affect the displacement response.

Keywords: B. Composites; E. Actuators; Soft PZT; Hard PZT

1. Introduction

Significant advances have been made with piezoelectric actuators in the past few decades. Piezoelectric actuators are used to replace the conventional actuators, including hydraulic, shape memory alloy, electromagnetic and linear induction actuators. This is due to their excellent electromechanical properties which make them suitable for the devices used for micropositioning, aerospace, communication, medication and fluid control, etc [1]. Several piezoelectric actuator designs have been proposed to meet the requirements including actuation performance and reliability. Those actuators are multilayer [2] bimorph [3] and unimorph [4]. To achieve both field-induced amplification and fatigue resistance associated with high electric field applied during operating the device, the functionally gradient materials (FGM) [5–7] and co-sintered (monolithic) ceramics [8,9] have been studied. Hall et al. [10] and Ngernchuklin et al. [11] presented the combination of piezoelectric (P) and electrostrictive (E) effect responses to the actuation performance and displacement hysteretic behavior. It was found that the displacement of the bi-layer P/E actuators was improved and showed narrow displacement hysteresis loop (reduced mechanical loss) due to non-hysteretic characteristics of its electrostrictive layer.

PZT is one of the most important piezoelectric materials due to its superior piezoelectric properties at tetragonal/rhombohedral MPB phases. Hence, PZT has been widely used for sensor and actuator applications. The modification of its properties by addition of acceptor and donor ions results in hard and soft PZTs, respectively [12]. Hard PZT's typically possess low permittivities and losses, small piezoelectric coefficients, high coercive field (E_c) resulting in difficult to pole and depole. Moreover, Hard PZTs such as PZT-8 show relatively narrow displacement hysteresis loop and small strain. Soft PZT's have higher permittivities, losses and piezoelectric coefficients, small coercive field (E_c) facilitating in switching of the dipoles under E-field. Although the

*Corresponding author. Tel.: +662 577 9439; fax: +662 577 9426.
E-mail address: piyalak@tistr.or.th (P. Ngernchuklin).

0272-8842/$ - see front matter
http://dx.doi.org/10.1016/j.ceramint.2012.10.131

field induced strain (0.1%) of soft PZT is much higher than that of hard PZT, it represents higher electrical loss and a wider displacement hysteresis loop due to domain motion [12,13]. From the electrical properties and the actuation characteristic of those soft and hard PZTs, the combination effect of the strain-E-field response of soft/hard PZT composite actuator has been much interested to be observed.

In this study, the bi-layer soft/hard PZT composite actuators prepared from two types of soft PZTs (PZT 5H and PZT 5A) and hard PZTs (PZT4 and PZT8) were fabricated. Soft PZT powders used in this experiment were 610-HD (TRS technologies) and KP 60 (Kyongwon Ferrite) represented as PZT-5H and PZT-5A, respectively. For hard PZT powders used were PZT-TISTR (TISTR) and DPZ-HQ (Dai Nippon toryo) as PZT 4 and PZT 8 respectively. The ratio of the soft and hard PZT powders used to form bi-layer composite was designed for 1:1. The bi-layer composites were prepared by co-pressing and co-sintering to obtain the monolithic layer between the two layers. The displacement and electromechanical properties of all bi-layer composites actuators were evaluated compared to the soft and hard PZT single layer.

2. Experimental procedure

2.1. Fabrication of soft/hard PZT monolithic bi-layer composite actuator

In this study, commercially available soft PZT 610-HD from TRS technologies Inc. USA, KP 60 from Kyongwon Ferrite Korea, hard PZT from TISTR (Thailand) and DPZ-HQ from Dai Nippon toryo, Japan were used. All powders were mixed with binder (6 wt% of the polyethylene glycol (PEG) solution), heated and then sieved through 135 mesh. The single layer samples of all four powders were pressed at 58 MPa and sintered at 1100 and 1250 °C to examine the firing shrinkage. The four types of bi-layer composite as deigned in Table 1 with soft and hard PZT volume ratio of 1:1 were made by pressing soft PZT and hard PZT materials sequentially in a single pressing step at 58 MPa. The pressed powders were heated at 550 °C for 1 h to remove binder and then were sintered in a closed alumina crucible with PbO source at 1100 and 1250 °C. The dome height was defined as the distance from the base to the center of the inner dome surface. The top and bottom surfaces of the bi-layer composite were electroded with fire-on silver paste and heat treated at 700 °C for 15 min. Then, all samples were poled under 30 kV/cm electric field strength in a heated silicone oil bath for 15 min at 120 °C. The electromechanical properties of the single layer samples and bi-layer composites were measured.

Table 1
The designation of soft/hard PZT bi-layer composite and sintering temperatures.

Soft/hard PZT powder for bi-layer composite	Sintering temperature (°C)
KP 60—TISTR	1100
KP 60—DPZHQ	1100
610HD—TISTR	1250
610HD—DPZHQ	1250

2.2. Characterization techniques

1. The piezoelectric charge coefficients, d_{33} and d_{33}^{eff}, were measured with a Berlincourt piezometer (Channel Products, Inc.) at 100 Hz.
2. The impedance analyzer was used to measure the frequency of maximum and minimum impedance (f_m and f_n) of the composites in order to compute Q_m according to the IEEE standards [14].
3. The bipolar axial displacements of the single layer ceramic and bi-layer composite actuators were measured by a laser probe (LK-G10, Keyence) in conjunction with a 10 kV (DC) power supply (Trek 609E-6) and a special displacement fixture. The oscilloscope (TDS 2012B, Tektronix) was used to display the displacement vs. E-field. The applied electric field was parallel to the poling direction in all measurements.

3. Results and discussions

Table 2 presents % planar shrinkage of single layer samples of four compositions at 1100 and 1250 °C. At 1100 °C, the highest and lowest sintering shrinkages were observed in PZT-DPZHQ (16.88%) and TISTR (4%), respectively. These results had influenced on the geometries of the bi-layer composites after sintering.

The electromechanical properties of soft and hard PZTs are shown in Table 3. The results of d_{33}, Q_m and E_c of all four samples were comparable to the standard type of PZT-5A for KP60, PZT-5H for 610HD, PZT-4 for TISTR and PZT-8 for DPZHQ. Also, the displacements at 1 kV/mm of all samples were in a range of 0.4–1.2 μm. Fig. 1, the butterfly-type loop was observed in 610HD (soft PZT) resulting from domain switching when E-field applied was higher than E_c, while the others showed linear displacement when E-field applied was comparable to or lower than E_c (Table 2).

Table 4 exhibits the geometries and electromechanical properties of soft/hard bi-layer composite actuators. All composites had a dome-shaped structure with various dome heights corresponding to the planar shrinkage

Table 2
% Planar shrinkage of soft and hard PZTs.

Materials	**% Planar shrinkage @1100 °C**	**% Planar shrinkage @1250 °C**
KP 60	16.04 ± 0.22	–
610 HD	13.88 ± 0.2	17.46 ± 0.30
TISTR	4 ± 0.17	15.65 ± 0.23
DPZHQ	16.88 ± 0.14	16.48 ± 0.23

Table 3
Electromechanical properties of soft and hard PZT single layers sintered at 1100 and 1250 °C.

Material	Standard piezo-type	d_{33} (pC/N)	Q_m	E_c (kV/mm)	Displacement (μm) @1 kV/mm	Displacement loop @1 kV/mm
KP 60—1100 °C	PZT-5A	220	90	1.26	0.8	Near linear
TISTR—1100 °C	–	Not densify samples, no measurement				
DPZHQ—1100 °C	–	330	970	–	0.45	–
610HD—1250 °C	PZT-5H	540	50	0.87	1.2	Butterfly
TISTR—1250 °C	PZT-4	320	305	1.05	0.5	Linear
DPZHQ—1250 °C	PZT-8	310	820	1.0	0.55	Linear

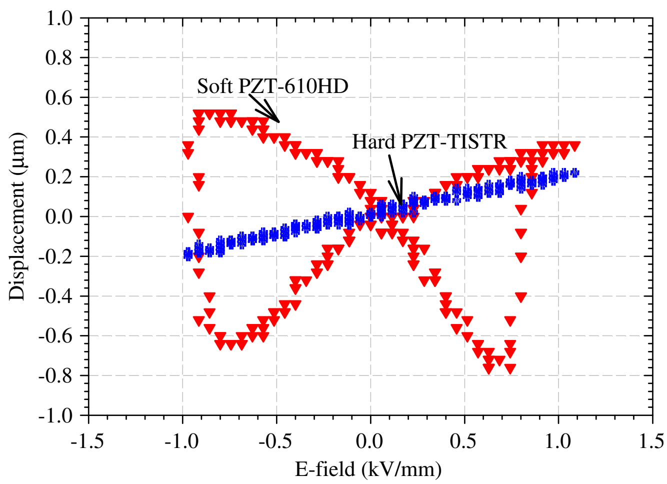


Fig. 1. Bipolar field dependence of displacement of Soft PZT-610HD and Hard PZT-TISTR.

difference of the two materials. In bi-layer KP60-TISTR had the highest dome height because of a large difference in planar shrinkage of KP60 (≈16%) and TISTR (≈4%) at 1100 °C. For bi-layer KP60-DPZHQ, almost the same planar shrinkage was observed in KP60 (≈16.04%) and DPZHQ (≈16.88%) resulting in a small curve of the composite with DPZHQ layer inside of the curve.

The bi-layer 610HD-TISTR showed the curved shape with 610HD layer inside of the curve. This was because 610HD was having higher shrinkage starting from 1100 °C. In case of the bi-layer 610HD-DPZHQ, the sintering temperature at 1250 °C, 610 HD ceramic (17.46%) had higher shrinkage than that of DPZHQ ceramic (16.48%). However, composite with DPZHQ layer inside of the curve was obtained. This was because the DPZHQ ceramic had higher sintering shrinkage at low temperature (1100 °C).

The measured effective d_{33} (d_{33}^{eff}) values were lower than the calculated effective d_{33} (Cal. d_{33}^{eff}) for all bi-layer composites (Table 4).

This attributed from the difference in the relative permittivity of those soft and hard PZT and the residue stress generated in the composite, which impeded domain alignment under E-field during poling.

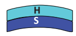
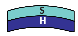
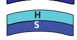
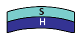
The displacements of all bi-layer composites were investigated at 1 kV/mm as presented in Table 4. The highest displacement was observed in the bi-layer 610HD-TISTR of 11 μm with less hysteretic loop (Fig. 2) compared to the butterfly loop (Fig. 1). The soft PZT used in this composite exhibited the lowest E_c (0.87 kV/mm) and the soft PZT layer was inside the curve that could facilitate for contraction of the composite under E-field applied, while the lowest displacement of 0.1 μm was found in bi-layer KP60-TISTR owing to the TISTR layer was not fully dense at 1100 °C. Bi-layer KP60-DPZHQ and 610HD-DPZHQ possessed less displacement of 0.3 and 1.3 μm, respectively. This could be explained as E-field applied (1 kV/mm) was lower than the E_c of bi-layer KP60-DPZHQ (Ec=1.25 and 1.0 kV/mm) then it was not enough to switch the domains. Like bi-layer 610HD-DPZHQ (E_c=0.87 and 1 kV/mm), the PZTs had the same E_c values as bi-layer 610HD-TISTR (E_c=0.87 and 1.05 kV/mm), the difference of both samples was that the bi-layer 610HD-DPZHQ presented of hard PZT layer inside of the curve. At 1 kV/mm, domains of 610HD layer in the bi-layer 610HD-DPZHQ was reorientation; however, 610HD layer was interfacing to DPZHQ layer which did not contract at this field. Thus the DPZHQ layer would constrain the displacement of 610HD layer. This resulted in a small displacement in the bi-layer 610HD-DPZHQ (1.3 μm). In contrast, the 610HD layer of the bi-layer 610HD-TISTR was inside the curve, at 1 kV/mm E-field, the 610HD layer was contracted. This geometry facilitated for higher displacement of 11 μm.

Fig. 2 presents the displacement of the bi-layer 610HD-TISTR at 1 and 2 kV/mm. At E-field 2 kV/mm the composite exhibited higher displacement and larger loop than that of 1 kV/mm. This was because E-field at 2 kV/mm was much higher than the E_c of 610HD (E_c=0.87 kV/mm) and TISTR (E_c=1.05 kV/mm). As a result 90° domain reorientation at high E-field application was occurred in the composite.

4. Conclusions

The soft/hard bi-layer monolithic composite actuators were prepared from co-pressing and co-sintering. The PZT materials with higher sintering shrinkage at low temperature (1100 °C) would be inside of the curved bi-layer composite. From the viewpoint of geometry, the dome-shaped actuator that the soft PZT layer was inside of the curve promoted the displacement performance (bi-layer 610HD-TISTR of 11 μm @ 1 kV/mm). In view of ferroelectric property, the PZT material with lower E_c was a

Table 4
Geometries and electromechanical properties of soft/hard bi-layer composite actuators.

Bi-layer composite	Curved height (mm)	Calculated d_{33}^{eff} (pC/N)	Measured d_{33}^{eff} (pC/N)	Q_m	Displacement (μm) @ 1 kV/mm
KP 60—TISTR (1100 °C)	1.38 	277	120	107	0.1 ± 0
KP60—DPZHQ (1100 °C)	0.25 	276	230	135	0.3 ± 0
610HD—TISTR (1250 °C)	0.52 	389	240	129	11 ± 0
610HD—DPZHQ (1250 °C)	0.76 	406	260	108	1.3 ± 0.28

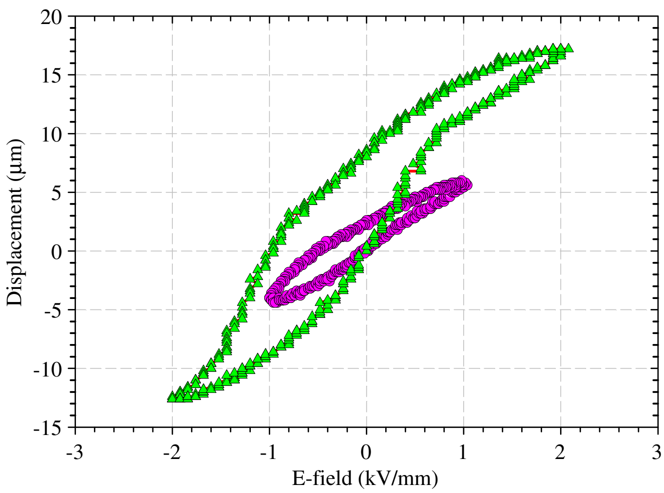


Fig. 2. Bipolar field dependence of displacement of bi-layer 610HD-TISTR at 1 and 2 kV/mm.

preference for actuator applications thanks to easier domain switching under low E-field applied.

References

[1] K. Uchino, Ferroelectric Devices, Marcel dekker Inc., New York, 2000.

[2] S.R. Winzer, N. Shankar, A.P. Ritter, Designing cofired multilayer electrostrictive actuators for reliability, Journal of the American Ceramic Society 72 (12) (1989) 2246–2257.

[3] M.R. Steel, F. Harrison, P.G. Harper, The piezoelectric bi-morph: an experimental and theoretical study of its quasistatic response, Journal of Applied Physics D 11 (6) (1978) 979–989.

[4] K.M. Mossi, G.V. Selby, R.G. Bryant, Thin-layer composite unimorph ferroelectric driver and sensor properties, Materials Letters 35 (1–2) (1998) 39–49.

[5] J. Qui, J. Tani, T. Ueno, T. Morita, H. Takahashi, H. Du, Fabrication and high durability of functionally graded piezoelectric bending actuators, Smart Materials and Structures 12 (1) (2003) 115–121.

[6] P.W. Alexander, D. Brei, J.W. Halloran, The fabrication and material characterization of PZT based functionally graded piezoceramics, in: Proceedings of SPIE, 5764, 2005, pp. 57–70.

[7] Y.H. Chen, T. Li, J. Ma, Development of piezoelectric monomorph actuator using electrophoretic deposition, Journal of Materials Science 41 (24) (2006) 8079–8085.

[8] J.A. Juuti, H. Jantunen, V. Moilanen, S. Leppavuori, Manufacturing of prestressed piezoelectric unimorphs using a postfired biasing layer, IEEE Transactions on Ultrasonics, Ferroelectronics and Frequency Control 53 (5) (2006) 838–846.

[9] H. Zhang, J. Li, B. Zhang, Sintering and piezoelectric properties of co-fired lead zirconate titanate/Ag composites, Journal of the American Ceramic Society 89 (4) (2006) 1300–1307.

[10] A. Hall, M. Allahverdi, E.K. Akdogan, A. Safari, Development and electromechanical properties of multimaterial piezoelectric and electrostrictive PMN-PT monomorph actuators, Journal of Electroceramics 15 (2) (2005) 143–150.

[11] P. Ngernchuklin, E.K. Akdogan, B. Jadidian, A. Safari, Electromechanical displacement of piezoelectric-electrostrictive monolithic bilayer composite, Journal of Applied Physics 105 (2009).

[12] S.L. Swartz, Topics of electronic ceramics, IEEE Transactions on Electrical Insulation 25 (5) (1990) 937–956.

[13] S.E. Park, T.R. Shrout, Ultrahigh strain and piezoelectric behavior in relaxor based ferroelectric single crystals, Journal of Applied Physics 82 (4) (1997) 1804–1811.