J. Korean Inst. Electr. Electron. Mater. Eng.
Vol. 36, No. 4, pp. 362-368 July 2023
DOI: https://doi.org/10.4313/JKEM.2023.36.4.6
ISSN 1226-7945(Print), 2288-3258(Online)

Regular Paper

Development of Textured 0.37PMN-0.29PIN-0.34PT Ceramics-Based Multilayered Actuator for Cost-Effective Replacement of Single Crystal-Based Actuators

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(Received May 30, 2023; Revised June 10, 2023; Accepted June 12, 2023)

Abstract: Multilayered actuators using $Pb(Mg_{1/3}Nb_{2/3})O_3$-$Pb(In_{1/2}Nb_{1/2})O_3$-$PbTiO_3$ (PMN-PIN-PT) crystals have demonstrated excellent properties, but are costly and lack mechanical strength. Textured PMN-PIN-PT ceramics exhibit robust mechanical strength and comparable properties to their single crystals form. However, the development of multilayered actuators using textured PMN-PIN-PT ceramics has not been achieved until now. This study presents the development of a multilayered actuator using textured 0.37PMN-0.29PIN-0.34PT ceramics with an $Ag_{0.9}/Pd_{0.1}$ inner electrode, co-fired at 950°C. A random 0.37PMN-0.29PIN-0.34PT ceramics multilayered actuator was also developed for comparison. The multilayered actuator consisted of 9 ceramic layers (36 μm thickness) with an overall actuator thickness of 0.401 mm. The textured and random 0.37PMN-0.29PIN-0.34PT ceramics-based multilayered actuators achieved displacements of 0.61 μm (0.15% strain) and 0.23 μm (0.057% strain) at a low applied peak voltage of 100 V. These results suggest that the developed multilayered actuator using high-performance textured 0.37PMN-0.29PIN-0.34PT ceramics has the potential to replace expensive single crystal-based actuators cost-effectively.

Keywords: Multilayered actuator, Textured ceramics, PMN-PIN-PT, Mechanical strength, Cost-effective

1. INTRODUCTION

Piezoelectric ceramic multilayered actuators are devices that convert electrical energy into mechanical energy through the inverse piezoelectric effect. They have been used in many applications such as precision positioners, motors, and vibration suppressors [1]. Mechanical deformation, also known as strain, under an applied electric field in piezoelectric ceramics, is a key property required for actuator applications [2]. Piezoelectric ceramics generally have much lower strain than single crystals of the same material system [3-6]. Despite their lower performance compared to single crystals, piezoelectric ceramics have been widely used in actuator

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applications where they are more practical than single crystals [7]. This is mainly because piezoelectric single crystals are expensive, have low mechanical strength, and their synthesis process is complex compared to piezoelectric ceramics. The reason for the inadequate strain response of piezoelectric ceramics is that the grains are randomly oriented, resulting in an average strain contribution [3-6].

It has been demonstrated that in piezoelectric ceramics, if randomly oriented grains are aligned in the same direction, particularly in the [001] direction, the strain response can be increased [8-17]. One of the effective approaches for developing aligned grains in piezoelectric ceramics is the templated grain growth (TGG) method [8-19]. This method involves using an anisotropic plate or needle-like single-crystal template, where a small volume fraction of the template crystals is uniformly distributed in the equiaxial fine matrix powder of the target material. The template crystals are then aligned in the desired crystallographic orientation, typically along the [001] direction. During the sintering process, the templates grow by consuming the target material [8-19]. One well-investigated textured piezoelectric ceramic is $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT), which exhibits improved strain compared to untextured (random) ceramic counterparts [15, 20-22]. The enhanced strain response of textured PMN-PT ceramics makes it ideal for use in actuator devices. However, this material system has a low Curie temperature (T_C) of 170℃, limiting its application to areas requiring high operating temperatures. It has been reported that the T_C of PMN-PT ceramics increases up to 300℃ when the $Pb(In_{1/2}Nb_{1/2})O_3$ (PIN) component is introduced to form the PMN-PIN-PT solid solution [23]. The relatively higher T_C of the PMN-PIN-PT material system makes it a good choice to meet the demand for high-operating temperature piezoelectric actuators. Due to its higher T_C and high strain, single-crystal PMN-PIN-PT has been used to develop multilayered actuator devices [24-26].

The PMN-PIN-PT ceramic has been textured in various compositions and has shown T_C ranging from 200℃ to 230℃, exhibiting high strain compared to random PMN-PIN-PT ceramics and close to that of a single crystal [27-30] making them more cost-effective than using single crystals in multilayered actuators. However, to the best of our knowledge, textured PMN-PIN-PT ceramic multilayered actuators have not been developed yet. Therefore, in this work, a textured PMN-PIN-PT ceramic multilayered actuator is developed, and for comparison, a random PMN-PIN-PT ceramic multilayered actuator is also developed. The low-temperature sintered (950℃) PMN-PIN-PT textured ceramics, exhibiting a considerably high field-induced unipolar strain of 0.23% at 2 kV/mm and a T_C of 231℃, were developed through the TGG method using a $BaTiO_3$ (BT) plate-like template [31]. This textured material was reproduced to develop the multilayered textured ceramic actuator (MLTCA).

The processing method used to develop the MLTCA and multilayered random ceramic actuator (MLRCA) is co-firing, which involves firing the ceramic and inner electrode stack structure at the sintering temperature of the ceramics [32,33]. For the inner electrode to be stable at the firing temperature, its melting point should be higher than the firing temperature. Silver (Ag) is commonly used as an inner electrode in ceramic multilayered devices due to its good conductivity [34]. However, with a melting point of 960℃, it is too close to the sintering temperature of the PMN-PIN-PT ceramic, making it challenging to use as an inner electrode. Another commonly used inner electrode is a silver palladium alloy (Ag/Pd), but due to the high cost of palladium, only a small amount is typically used in the alloy. For this work, the selected inner electrode was $Ag_{0.9}/Pd_{0.1}$ by weight, which has a higher melting point of approximately 1,000℃ [34-38]. The microstructure and electromechanical properties of both the MLTCA and MLRCA were characterized, and the results were compared and discussed. The textured PMN-PIN-PT ceramic multilayered actuator developed in this work shows great potential for various applications requiring high-performance and cost-effective piezoelectric actuators operating at elevated temperatures.

2. EXPERIMENTAL

Detailed processing on the low-temperature sintering of 0.37PMN-0.29PIN-0.34PT textured ceramics is given in reference [31]. Experimental details on the development of co-fired MLTCA and MLRCA are shown in Fig. 1. First, the tape-cast green sheets with BT template for MLTCA and without BT for MLRCA were cut into 5×5 cm pieces. The two cut sheets were laminated by applying a uniaxial pressure of 10 MPa at 40℃ for 5 minutes. Then, the $Ag_{0.9}/Pd_{0.1}$ electrode was screen printed on the laminates, followed by drying the printed

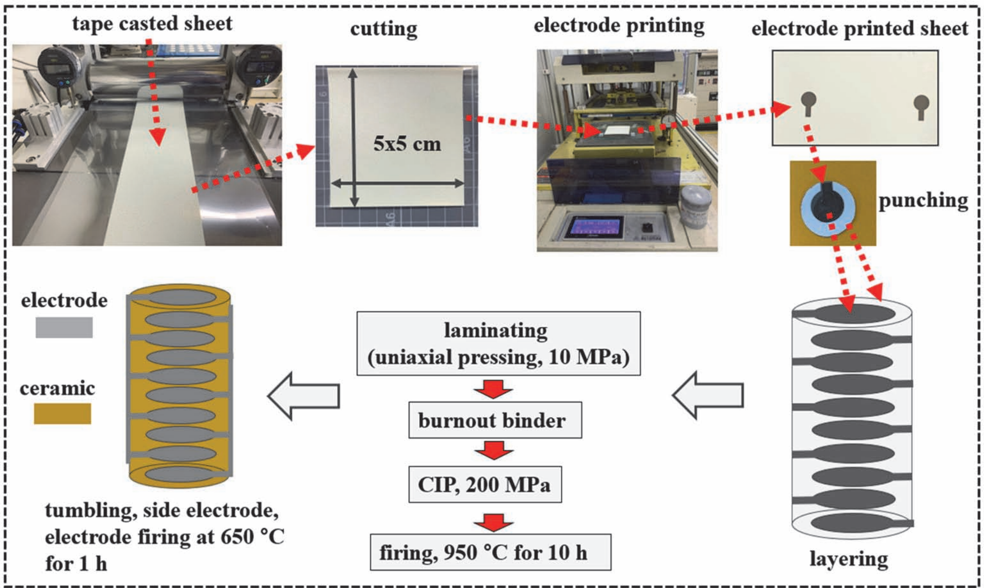

Fig. 1. Shows pictorial and schematic experimental details on the fabrication of co-fired multilayered textured and random 0.37PMN-0.29PIN-0.34PT ceramics actuators.

electrode at 80℃ for 5 minutes in the oven. The electrode-printed tape-cast sheets were punched with a 10 mm diameter circular cutter, followed by layering them alternately based on the side extended electrode direction. The layered structure was then laminated with a uniaxial pressure of 10 MPa at room temperature. Next, the binder burnout was carried out under similar heating conditions as in reference [18]. After binder burnout, the samples were subjected to cold isostatic pressing (CIP) at 200 MPa. Then, the samples were fired at 950℃ for 10 hours in a box furnace using a double crucible sealed with $PbZrO_3$ powder, which helps minimize lead loss [18]. The sintered samples were then tumbled to expose the extended side electrode of each layer and connect them by printing the electrode on the side of the tumbled stack. Finally, the side electrode firing was carried out at 650℃ for 1 hour. Before electrical property measurement, the sintered multilayered structure was poled by applying a 3 kV/mm electric field in silicone oil at 120℃ for 30 minutes. The microstructure of the multilayered was examined using a backscattered electron image (BSE, JEOL JSM-7900F). Energy-dispersive X-ray spectroscopy (EDS, JEOL JSM-6610LV) was used to investigate the stability of the $Ag_{0.9}/Pd_{0.1}$ electrode. The field-induced strains and displacements of the MLTCA and MLRCA were measured using a laser displacement sensor (aixACCT Systems GmbH 2013, Germany).

3. RESULT AND DISCUSSION

The impact of inner electrode thickness on the piezoelectric strain response of MLTCA is demonstrated in Fig. 2(a). Thicker electrodes have a suppressive effect on the strain response, as observed with a 10 μm thick electrode [see Fig. 2(b)] resulting in only 0.1% strain at 2 kV/mm. Reducing the inner electrode thickness to 7 μm [see Fig. 2(c)] increases the strain response to 0.14%. However, further reduction to 4 μm leads to a decreased strain response of 0.04%, likely due to the formation of voids caused by electrode discontinuation, as shown in Fig. 2(d). Therefore, an inner electrode thickness of 7 μm appears to be the optimal choice. This optimization of the inner electrode thickness was solely performed on the textured ceramics, and this optimized thickness was then utilized for both the MLTCA and MLRCA designs.

The BSE cross-sectional microstructure in Fig. 3(a) provides a clear view of the layered structure of the MLTCA, with the single-layer thickness measurement of 36 μm textured 0.37PMN-0.29PIN-0.34PT ceramics sandwiched between the $Ag_{0.9}/Pd_{0.1}$ electrodes. The line scan EDS elemental analysis in Fig. 3(b) confirms the presence of Ag and Pd in the inner electrode layer. The color-coded representation of Ag electrode (red), and Pd electrode (purple) in Fig. 3(c) and (d) further illustrate the distinct layers in the

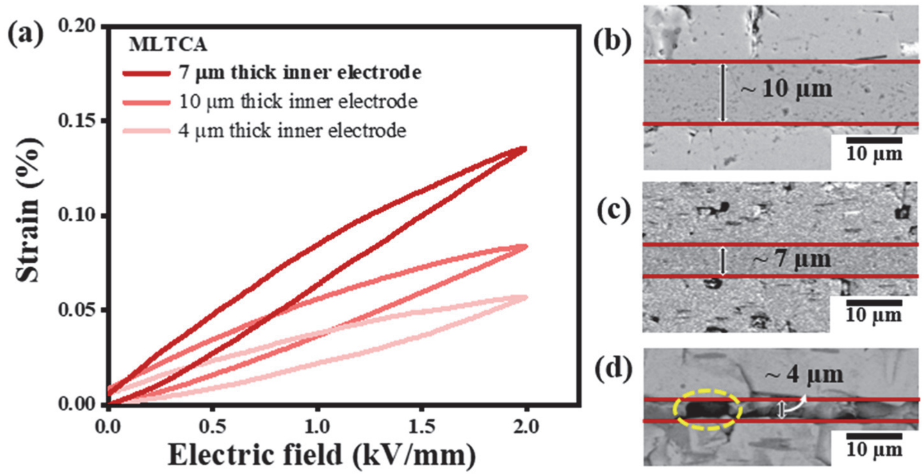

Fig. 2. The effect of electrode thickness on the strain response of MLTCA. (a) Strain versus electric field curves for different electrode thicknesses. BSE cross-sectional images of the MLTCA structure with inner electrode thicknesses of (b) 10 μm, (c) 7 μm, and (d) 4 μm.

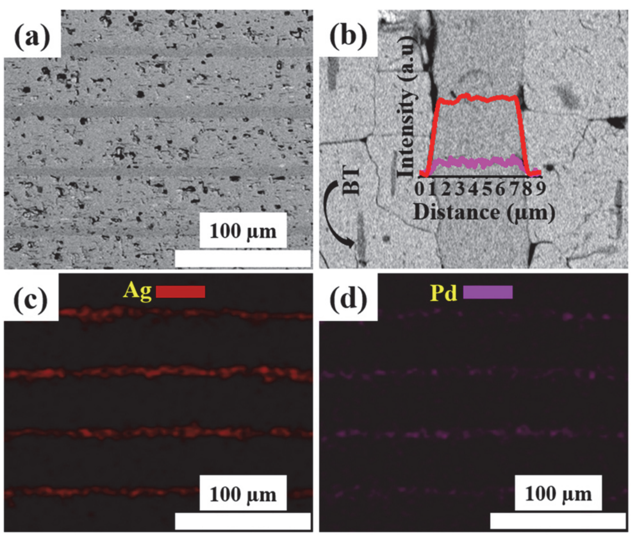

Fig. 3. (a) Cross-sectional BSE image of MLCTA, (b) line scan EDS elemental analysis, the BT represents $BaTiO_3$ template, (c) the red color represents the Ag electrode, and (d) the purple color represents the Pd electrode.

MLTCA. The absence of interdiffusion between the electrode and ceramic layers in the BSE image indicates that the $Ag_{0.9}/Pd_{0.1}$ inner electrode layer maintains its stability even after sintering at a temperature of 950℃ for 10 h which is a crucial characteristic for the reliability and performance of the MLTCA.

Figure 4 presents the results of the field-induced unipolar strain and displacement of the MLTCA. The unipolar strain of the multilayered actuator is defined as the deformation in response to an electric field in one direction, and it is calculated by dividing the total strain obtained from the multilayered by the total number of ceramic layers. At an applied electric field of 3 kV/mm, the active layer of the MLTCA produced a high unipolar strain of 0.19%. Of particular importance, the strain response showed nonlinearity of only 11.3%, which in general ranges between 10% to 15% for piezoelectric ceramics making this device ideal for precision positioning applications. The displacement results of the MLTCA with applied peak voltage were also presented in Fig. 4(b). The displacement is a measure of the deformation of the MLTCA in response to an applied voltage. For instance, at an applied peak voltage of 150 V, the MLTCA produced a displacement of 0.9 μm which is 0.225% of the total thickness of the actuator (0.401 mm) which is more than twice larger than that of typical PZT-based multilayered actuators [39-41]. The higher displacement of the MLTCA is directly related to the higher unipolar strain produced by the textured ceramic layers under the applied field. Overall, the results in Fig. 4 demonstrate that the MLTCA design is effective in producing higher unipolar strain and displacement, which are essential properties for actuation applications.

Figure 5 presents a comparison between the MLTCA and MLRCA in terms of their unipolar strain, displacement, and bipolar strain and displacement. In the first subplot [Fig. 5(a)],

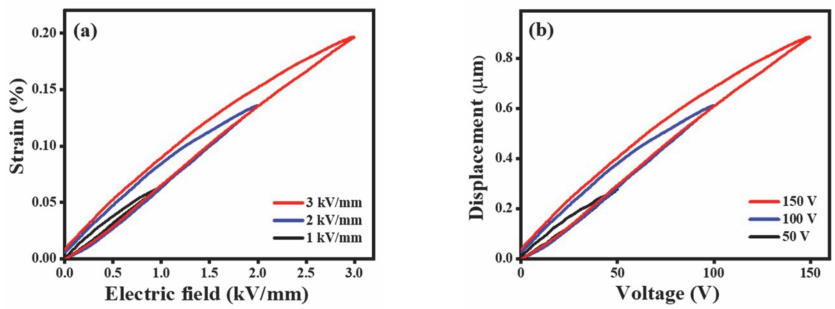

Fig. 4. (a) Variation of the field-induced unipolar strain of the MLTCA and (b) variation of the displacement of the MLTCA with various peak voltage.

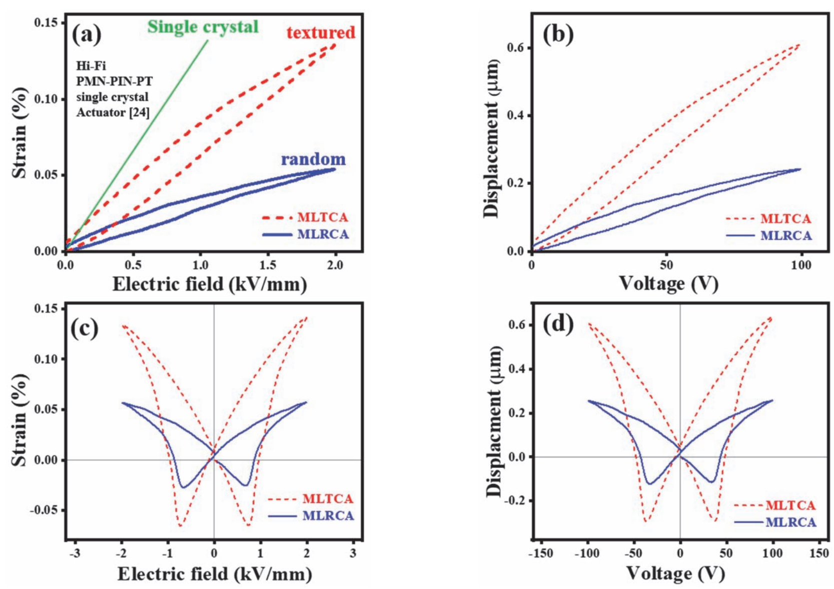

Fig. 5. (a) Comparison of the unipolar strain, (b) displacement, (c) bipolar strain, and (d) bipolar displacement of MLTCA and MLRCA [The strain and displacement of the MLTCA are re-drawn from Fig. 4 for ease of comparison. For comparison, the strain level of the multilayered actuator developed using single crystal PMN-PIN-PT [24] is also included in (a)].

the unipolar strain of MLTCA, MLRCA, and single crystal [24] multilayered actuator of the same material system is compared. The unipolar strain of MLTCA is shown to be higher than that of MLRCA, with the former exhibiting a strain of 0.064%, 0.14%, and 0.19% at an applied electric field of 1 kV/mm, 2 kV/mm, and 3 kV/mm, respectively, while the latter has a strain of 0.055% at 2 kV/mm with nonlinearity of 14%. These results show that the use of textured ceramics in multilayered actuators narrowed the performance gap between the multilayered actuator formed from random and single crystals of the same materials system. In the second subplot [Fig. 5(b)], the displacement of MLTCA and MLRCA is compared. The MLTCA is shown to exhibit longer displacement than the MLRCA at the same applied peak voltage, with MLTCA achieving displacements of 0.28 μm, 0.61 μm, and 0.9 μm at 50 V, 100 V, and 150 V, respectively, while MLRCA has a displacement of 0.25 μm at 100 V. In the third and fourth subplots [Fig. 5(c) and (d)], the bipolar strain and displacement of both actuators are compared. The subplots show that both actuators exhibit symmetric switching characteristics, with MLTCA exhibiting more than 2 times higher bipolar strain and longer displacement than MLRCA at the same applied field and peak voltage. In summary, the results suggest that textured ceramics can be a promising alternative to low-performance random ceramics and expensive single crystal-based actuators. The developed multilayered actuator has the potential for use in various applications, such as precision positioners, motors, and vibration suppressors.

4. CONCLUSION

The development of a high-performance multilayered actuator using textured ceramics is presented in this study. Multilayered actuators are crucial components in various applications such as precision positioners, motors, and vibration suppressors, as they convert electrical energy into mechanical energy. In this study, a multilayered actuator was developed using textured 0.37PMN-0.29PIN-0.34PT ceramics with an $Ag_{0.9}/Pd_{0.1}$ inner electrode through co-firing at 950°C for the first time. The resulting actuator showed 0.19% strain at an applied electric field of 3 kV/mm with low nonlinearity of 11.3% and 0.9 μm displacement at 150 volts. Compared to the multilayered actuator made using random ceramics, the actuator made using textured ceramics exhibited more than 2 fold higher strain and displacement at the same applied field and peak voltage, respectively. These findings demonstrate that the developed multilayered actuator using textured ceramics can be a promising alternative to expensive single crystal-based actuators. By using the TGG method, the actuator's performance can be improved, leading to a wide range of potential applications in various industries.

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ACKNOWLEDGEMENTS

This research was supported by Korea Electrotechnology Research Institute (KERI) Primary research program through the National Research Council of Science & Technology (NST) funded by the Ministry of Science and ICT (MSIT) (No.23A01032) and UST Young Scientist Research Program 2021 (2021YS28) through the University of Science and Technology of the Republic of Korea.

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