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Electric field induced fracture mechanism and aging of piezoelectric behavior in $Pb(MgNb)O_3$–$Pb(ZrTi)O_3$ multilayer ceramic actuators

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Abstract

Electrical aging and mechanical fracture behavior of $0.2(PbMg_{1/3}Nb_{2/3}O_3)$–$0.8(PbZr_{0.475}Ti_{0.525}O_3)$ multilayer ceramic actuators have been investigated by applying both bipolar and unipolar voltages for unpoled and poled actuator, respectively. After 8.7×10^7 cycles of 2 kV/mm bipolar voltage, unpoled multilayer ceramic actuator had distorted shape of *P*–*E* hysteresis loops. Effective electromechanical coupling coefficient k_{eff} of poled actuator was calculated by considering resonant frequency and anti-resonant frequency. Pseudo-peizoelectric constant, d_{33}, was also estimated through the strain versus electric field characteristics. From the SEM analysis, crack growth of multilayer ceramic actuator was clearly observed along the boundary between electrode and inactive region. The main reason for destruction of $0.2(PbMg_{1/3}Nb_{2/3}O_3)$–$0.8(PbZr_{0.475}Ti_{0.525}O_3)$ multilayer ceramic actuators was mechanical tensile stress coming from both actuation of, d_{33}, mode and motion due to Poisson ratio.

Keywords: A. Tape casting; D. PZT; Multilayer ceramic actuator (MCA); Aging; Piezoelectric constant; Electromechanical coupling coefficient

1. Introduction

Multilayer ceramic actuators are devices, which can be adjusted or moved by stimulation. Generally, ceramic actuators use electrical signal or mechanical stress for their stimuli to response. Piezoelectric materials, which convert mechanical energy into electrical energy and vice versa, have been widely employed for these actuators applications. Due to the rapid development of functional ceramics, many kinds of piezoelectric materials have been employed for ceramic actuators. Among them, $Pb(Zn_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ (PZN–PT) [1], $Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ (PMN–PT) [2], $Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbZrTiO_3$ (PMN–PZT) [3], and $Pb(Ni_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ [4] are representative materials for their high piezoelectric constant and electromechanical coupling coefficient. In many cases, piezoelectric properties have been associated with their morphotropic phase boundary (MPB), near MPB they show higher piezoelectric constant and electromechanical coupling coefficient. Especially, $0.2(PbMg_{1/3}Nb_{2/3}O_3)$–$0.8(PbZr_{0.475}Ti_{0.525}O_3)$ (hereafter PMN–PZT) materials show high piezoelectric constant, d_{33}, of 600–700 pC/N and electromechanical coupling coefficient k_p of 0.65. High Curie temperature of 350 °C makes this PMN–PZT material as a good candidate for actuator applications without any serious consideration of thermal degradation, since these types of devices create huge amount of heat during operational conditions.

Therefore, PMN–PZT materials can be regarded as advanced materials for actuators applications.

Tape casting, employed in this process, is a low-cost process to make high quality long film. During this process, green sheet have uniform thickness and smooth surface. In principle, more than 50 layers are stacked to have enough displacement with generating force. During the operation of multilayer ceramic actuator (hereafter MCA), applied high voltage of more than 50 V induces aging and destruction problems. Thus, these electrical and mechanical aging phenomenons should be carefully examined. By applying an electric field, domain walls in the piezoelectric material reorient along an electric field as much as they could. During this process, both electrical and mechanical aging was propagated. Although this aging is very important from the application point of view, very limited papers were reported

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on this effect. The aging should be considered in these two categories, i.e. depoling and destruction of actuators.

In this paper, we report on the aging and fracture behavior of PMN–PZT multilayer ceramic actuators.

2. Experimental

2.1. Preparation of MCA devices

Tape casting method was employed to fabricate MCA from the stoichiometric $0.2(PbMg_{1/3}Nb_{2/3}O_3)$–$0.8(PbZr_{0.475}Ti_{0.525}O_3)$ powder. PbO, ZrO_2, TiO_2, MgO, and Nb_2O_5 powders, of high purity of 99.9%, were used as starting materials. The powders were mixed and calcined at 850 °C for 2 h. Ag_{70}–Pd_{30} electrode was applied to the 100 μm thick green sheets with screen-printing method, and then green sheets were cut to desired pattern size of 5 cm × 5 cm × 5 cm. Finally, MCA devices were fabricated by sintering process at 1100 °C.

2.2. Measurement and analysis

Both poled and unpoled specimens were prepared for experiments, electric field of 2 kV/mm bipolar bias with 60 Hz was applied to the unpoled actuator and electric field of 1.8 kV/mm unipolar bias with 910 Hz was applied to the poled actuators to make these actuators aged. Polarization versus electric field (*P*–*E*) characteristics was carried out employing Sawyer–Tower technique. To measure dielectric and impedance properties, *HP 4194 A* was employed. Frequency dependent dielectric permittivity and impedance were measured at room temperature. Scanning electron microscopy (SEM) was employed to examine cracked regions of MCA.

3. Result and discussion

Fig. 1 displays *P*–*E* hysteresis loops of unpoled PMN–PZT MCA before and after 8.7×10^7 cycle. Electric field of 2 kV/mm bipolar bias was applied to the actuator to observe aging behavior. After 8.7×10^7 cycles, the shape of *P*–*E* loop was distorted compared to the original *P*–*E* loop observed before aging. This implies that degradation of resistance in ceramic actuator is obvious, but it is negligible after 8.7×10^7 cycles. Suddenly, MCA was destroyed without any serious degradation of *P*–*E* hysteresis loop. This means that mechanical crack due to the d_{33} mode was more seriously damaged to the MCA than degradation of resistance.

Fig. 2 exhibits frequency dependent capacitance and tan δ of both unpoled fresh and aged PMN–PZT MCA measured from 100 Hz to 1 kHz at same temperature. It is very important to keep the same temperature, since PZT has pyroelectric properties. The MCA shows degraded value of capacitance and loss tan δ after cyclings. As shown in the fig-

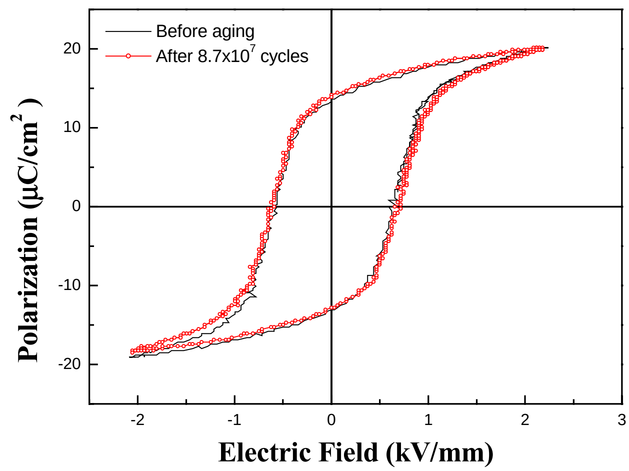

Fig. 1. Ferroelectric hysteresis loops of unpoled multilayer ceramic actuator before and after 8.7×10^7 cycles at 60 Hz.

ure, the capacitance was decreased and tan δ was increased as increasing the frequency. The capacitance was shrinked by 1.97 and 3.76% from 100 Hz to 1 kHz for unpoled fresh and unpoled aged MCA, respectively. The capacitance variance of aged MCA was more serious than fresh MCA. Aged MCA has higher tan δ than that of fresh MCA in all frequency ranges. This comes from degradation of resistance due to the continuous piezo-motion. Since tan δ has close relationship with resistance of MCA like $\tan\delta = 1/(\omega CR)$, degraded resistance raised tan δ.

There were reports that the electrode migration of MCA is a main reason for crack growth [5]. But we would like to point out the decreased capacitance after aging instead of increasing the capacitance. This means that Ag–Pd electrode did not migrate due to high electric field.

Fig. 3(a) and (b) show frequency dependent impedance and phase of poled fresh PMN–PZT and poled aged PMN–PZT MCA, respectively. Frequency dependent impedance was measured from 1 kHz to 1 MHz frequency range. Since PMN–PZT MCA was poled, the resonance frequency (f_r) and anti-resonance frequency (f_a) of MCA were observed. In fresh PMN–PZT MCA case, at least four f_r and f_a resonant frequency were easily observed. However, in aged PMN–PZT MCA has only one ambiguous f_a and f_r as observed. By applying unipolar bias to the MCA, it seems that poling was continuously diminished.

From these f_a and f_r, the effective electromechanical coupling coefficient k_{eff} of MCA can be estimated. The equation can be express as follow [6]:

$$k_{eff} = \sqrt{\frac{f_a^2 - f_r^2}{f_a^2}}. \quad (1)$$

Calculated effective coupling coefficients are written in Table 1. As shown in the table, the calculated effective coupling coefficient decreased due to the aging effect. To clarify the piezoelectric motion of PMN–PZT MCA, strain versus applied electric field was measured by laser vibrometer us-

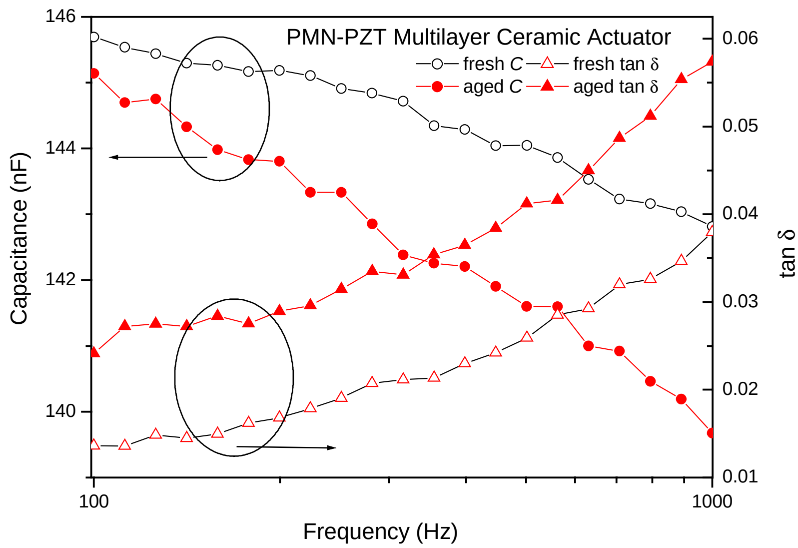

Fig. 2. Frequency dependent capacitance and tan δ of multilayer ceramic actuator measured from 100 Hz to 1 kHz at room temperature. Symbols (○) and (△) represent capacitance for unpoled fresh and aged actuators, respectively. Symbols (●) and (▲) represent tan δ for unpoled fresh and aged actuators, respectively.

ing Doppler effect. Fig. 4 exhibits strain dependent electric field relation from both poled fresh and poled aged MCA. If we assume that piezoelectric motion of MCA follows longitudinal mode, then piezoelectric equations corresponding to the longitudinal extension can be expressed as follow:

$$x_3 = s_{33}^E X_3 + d_{33} E_3, \qquad (2)$$

$$D_3 = d_{33} X_3 + \varepsilon_{33}^X E_3,$$

where E is electric field, X is the stress, x is the strain, D is the displacement, d is the piezoelectric constant, s_{33}^E is elastic compliance, and ε_{33}^X is the electromechanical coupling coefficient. By applying an electric field E_3, strain x_3 can be modulated. A pseudo-piezoelectric constant, d_{33}, is calculated from two point linear approximation using the strain at maximum and minimum electric field points. The slop of this figure corresponds to pseudo-piezoelectric constant of the actuator. As shown in the figure, pseudo-piezoelectric constant decreased due to aging. Calculated pseudo-piezoelectric constants are writ-

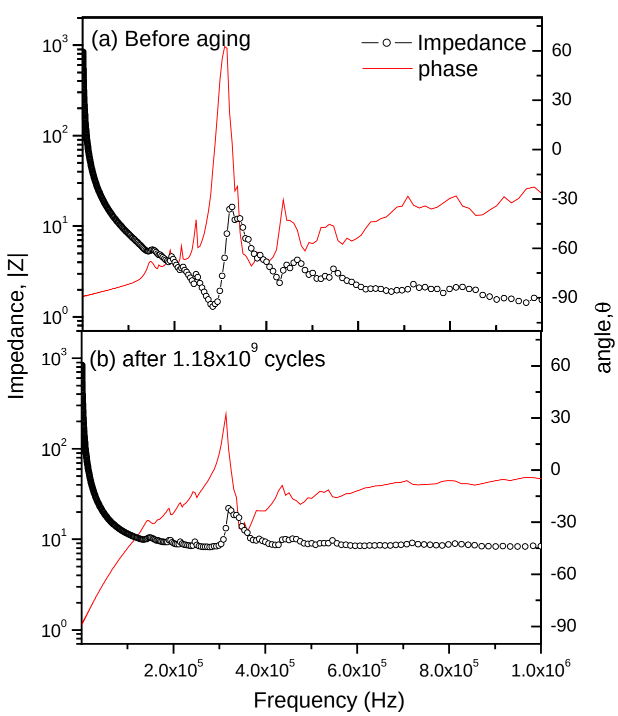

Fig. 3. Frequency dependent impedance and phase of multilayer ceramic actuator measured from 1 kHz to 1 MHz at room temperature; (a) poled fresh multilayer ceramic actuator, and (b) poled aged multilayer ceramic actuator. Symbols (–○–○–) and solid line stand for the impedance and phase of multilayer ceramic actuator, respectively.

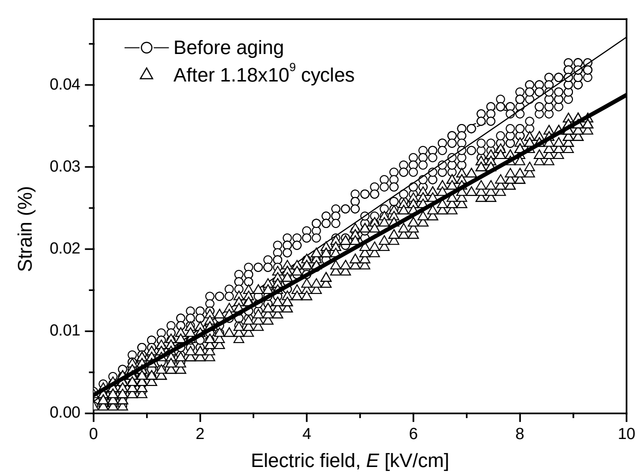

Fig. 4. Strain versus electric field characteristics of the multilayer ceramic actuator measured by laser vibrator. Symbols ○ and △ represent poled fresh multilayer ceramic actuator and poled aged actuator, respectively.

Table 1
Resonance frequency, effective electromechanical coupling coefficient, k_p, and pseudo-piezoelectric constant, d_{33}, of poled $0.2(PbMg_{1/3}Nb_{2/3}O_3)$–$0.8(PbZr_{0.475}Ti_{0.525}O_3)$ multilayer ceramic actuators

	Cycling number			
	0	5.4×10^8	7.03×10^8	1.18×10^9
Resonant frequency f_r (kHz)	286	303	303	303
Anti-resonance frequency f_a (kHz)	321	325	319	319
Effective k_p	0.454	0.361	0.312	0.312
Pseudo-piezoelectric constant d_{33} (pm/V)	444	383	373	366

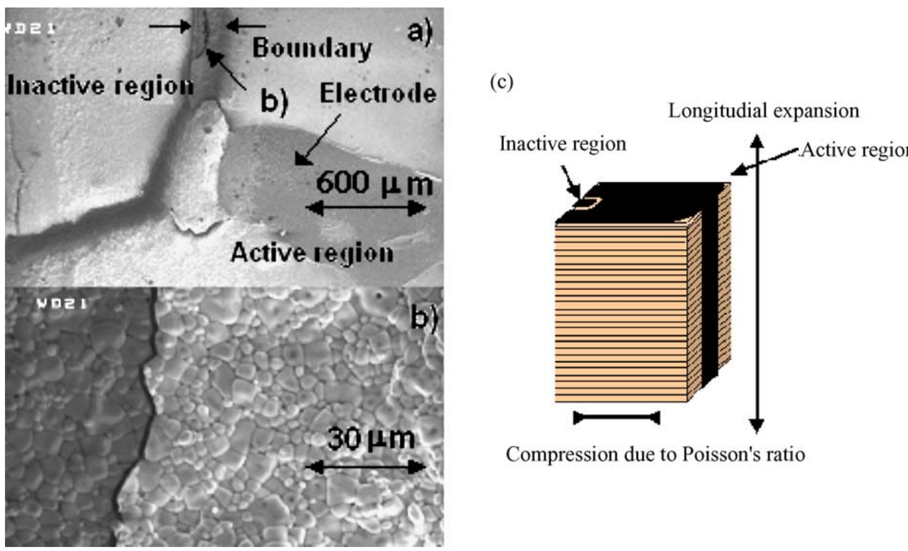

Fig. 5. Scanning electron microscopy images showing cracked surface of (a) $Pb(Mg_{1/3}Nb_{2/}3)O_3$–$PbZrTiO_3$ actuator, (b) crack line of destroyed actuator, and (c) illustration of tensile stress generated within the multilayer ceramic actuators.

ten in Table 1. As shown in Figs. 3 and 4, MCA loose its piezoelectric response due to degradation of MCA and the depoling effect. Pseudo-peizoelectric constant decreased 17.5% after 1.18×10^9 unipolar cycles of unipolar bias.

Fig. 5(a) reveals the SEM images of destroyed surface of PMN–PZT actuators. After 8.7×10^7 cycles of 2 kV/mm bipolar voltage, finally tested MCA was destroyed. Small piece of electrode was observed in the surface of destroyed PMN–PZT MCA. Since MCA have d_{33} mode for its actuation, very strong periodic tensile stress is applied to the boundary between active region of electrode and inactive region along to the electric field. At the same time another tensile stress applied to the boundary between active and inactive region normal to an electric field direction comes from the Poisson's ratio to compensate volume expansion parallel to the electric field. Due to this accumulation of tensile stress, crack develops between the active and inactive region. Small crack in Fig. 5(a) is magnified in (b). Boundary between active and inactive region is clearly observed. The crack was propagated along to the boundary between the active and inactive region regardless of the grain boundary. Fig. 5(c) illustrate expansion direction and tensile stress result in crack of MCA.

4. Conclusions

Aging and destruction behavior of $0.2(PbMg_{1/3}Nb_{2/3}O_3)$–$0.8(PbZr_{0.475}Ti_{0.525}O_3)$ multilayer ceramic actuators has been investigated by applying both bipolar and unipolar wave function for unpoled and poled MCA, respectively. *P–E* hysteresis loops were traced with a triangular shape electric field of 2 kV/mm to unpoled MCA. After 8.7×10^7 cycles of triangular wave function, *P–E* hysteresis loops of MCA devices have been distorted. Pseudo-peizoelectric constant, d_{33}, was calculated from the strain versus electric field characteristics. From the frequency dependent capacitance and loss tangent, it is clearly observed that aged MCA has higher loss tangent due to degraded resistance. We found that in mechanical aging, the tensile stress come from both vertical, d_{33}, expansion mode and horizontal motion of Poisson ratio, mainly influenced by the performance of multilayer ceramic actuators.

Acknowledgements

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