# Degradation of 0.2PMN-0.8PZT Multilayer Ceramic Actuators

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Aging characteristics of 0.2PMN-0.8PZT multilayer ceramic actuators (MCA) has been investigated by applying both triangular wave function for unpoled and unipolar wave for poling. *P-E* hysteresis loops of the MCA had been distorted after about 90 million cycles running in triangular wave function. Effective electromechanical coupling coefficient was calculated in resonant and anti resonant frequencies. And pseudo-piezoelectric constant $d_{33}$ was also estimated from the strain versus electric field characteristics. The crack growth of MCA was clearly observed along to the boundary between electrode and inactive area. That results were thought due to the internal tensile stress came from both actuation of $d_{33}$ mode and motion of Poisson ratio.

*Keywords* : MCA, PMN-PZT, Aging, Depoling, Unipolar wave, Crack growth

## 1. INTRODUCTION

Actuators are devices, which can be used in on-off system, adjusted or moved by stimulation. So, actuators act by electrical signals or mechanical stress changes. Piezoelectric materials, which convert mechanical energy into electrical energy and vice versa, have been used for these applications. Due to the 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], and PMN-PZT[3] are representative materials. In many cases, piezoelectric responses have been associated with their morphotropic phase boundary(MPB), really, they show higher piezoelectric constant and electromechanical coupling coefficient in MPB. Especially, 0.2PMN-0.8PZT has 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 0.2PMN-0.8PZT as a good candidate for actuating material without any serious consideration of thermal degradation, since these types of devices create huge amount of heat during operation conditions. Therefore 0.2PMN-0.8PZT can be regarded as advanced materials for application as an actuator. In general, MCA have advantages of fast response, high generating force, and low power consumption over other types of actuators. Hence, MCA can be used for accurately controlled valve, inject nozzle, and pump.

Tape casting process is a low-cost process to make high quality long film. Using this process, we can get green sheet which has uniform thickness and smooth surface. In principle, more than 50 layers are stacked to have enough displacement with blocking force. To operate MCA devices properly, a high voltage of more than 50 V is usually applied to the devices. However, this high voltage makes device performance degraded with time. Thus, this aging phenomenon should be carefully examined; otherwise unexpected destruction of MCA devices can arise serious problems in the whole system. By applying alternative bias, domain walls in the piezoelectric material reorient to align along to an electric field as much as they could. During these processes, mechanical as well as electric aging make devices degraded state. Since piezoelectric MCA has outstanding mechanical characteristics such as high generating force and quick response for industrial applications, it was regarded as a good candidate for valves and injectors applications. However, it is very rare to fine reports related with aging effects depends on cycling.

So, we reported on the aging and failure mechanism of 0.2PMN-0.8PZT MCA.

## 2. EXPERIMENTALS

### 2.1 Preparation of MCA devices

Tape casting method was employed to fabricate MCA from the stoichiometric 0.2PMN-0.8PZT. 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, 50 layers green sheets with electrodes were laminated and then they were cut to desired pattern size of 5 × 5 × 5 $mm^3$. Finally sized MCA devices were fabricated by sintering process at 1100 °C. Thickness of ceramic layer and $Ag_{70}$-$Pd_{30}$ electrodes were measured by images of the SEM.

### 2.2 Measurement and analysis

Both poled and unpoled samples were prepared. The electric field of 2 kV/mm triangular wave function with 60 Hz was applied to the unpoled , and electric field of 1.8 kV/mm unipolar wave function with 910 Hz was applied to the poled MCA in order to be aged these actuators. The displacement as a function of electric field and leakage current and electric field was measured at the acceleration condition of 900 Hz. XRD patterns of the powders were analyzed by Philips P100 with Cu-$K_\alpha$ source. Polarization-Electric field (P-E) characteristics were carried out at 60 Hz using Sawyer-Tower. Dielectric and impedance properties were measured by HP 4194A impedance analyzer. Frequency dependent on dielectric permittivity and tan δ as well as impedance and phase were measured at room temperature. Cracked-regions of MCA were analyzed by SEM.

## 3. RESULTS AND DISCUSSION

MCA was evaluated by XRD. Figure 1 shows that unpoled 0.2PMN-0.8PZT is polycrystalline with small pyrochlore phase near 29 °. This phase may be come from sintering process, even though columbite method was employed to remove pyrochlore phase. The (001) and (h00) implies that unpoled 0.2PMN-0.8PZT has tetragonal at room temperature.

Figure 2 reveals XRD patterns of poled fresh 0.2PMN-0.8PZT, unpoled fresh 0.2PMN-0.8PZT, and poled aged 0.2PMN-0.8PZT, respectively. The intensity of 001-reflection of poled fresh MCA is bigger than those of unpoled fresh and poled aged MCA; the ratio of $I_{001}/I_{100}$ is 1.228 while it is 0.972 and 1.025 for unpoled fresh and poled aged states, respectively. By applying an electric field along to the <001> direction in poling process, large number of domains was aligned to the same direction with an electric field. Hence the intensity of 001-reflections was increased.

However, the ratio of $I_{001}/I_{100}$ of poled aged MCA become similar with unpoled fresh stated by loosing poling properties. From the XRD θ-2θ scan, lattice parameters of 0.2PMN-0.8PZT were calculated by employing Nelson-Rilley extrapolation function with least mean square methods. The calculated values are written in the Table 1. As shown in the Table 1, the ratios of lattice parameter, c/a, are similar with in three case. We can argue that lattice parameters are hardly changed during poling and aging process.

Figure 3 showed P-E hysteresis loops of unpoled 0.2PMN-0.8PZT MCA before and after aging. Electric field of 2 kV/mm triangular wave function was applied to the actuator to observe aging behavior. After 8.7×$10^7$ cycles, the shape of P-E loop was shifted than the original loop observed before aging. This implies that

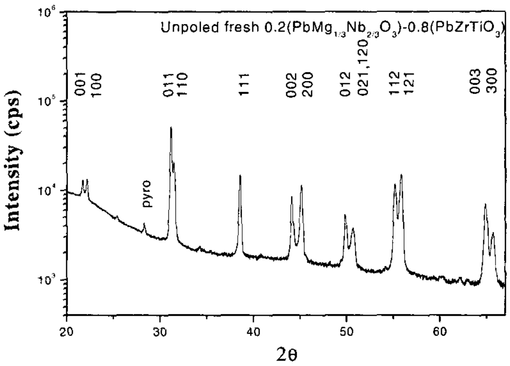


Fig. 1. XRD patterns (Cu $K_\alpha$) of 0.2PMN-0.8PZT actuators.

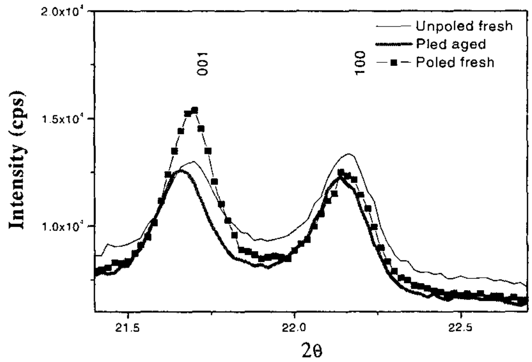


Fig. 2. Magnified XRD patterns (Cu $K_\alpha$) of poled fresh, unpoled fresh, and poled aged 0.2PMN-0.8PZT actuators.

degradation of resistance in ceramic actuator is obvious, but negligible after $8.7\times10^7$ 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 propagated and more seriously damaged to the MCA than degradation of resistance.

Figure 4 exhibits capacitance and tanδ of both unpoled fresh and aged 0.2PMN-0.8PZT MCA dependent on fre-

Table 1. Lattice parameters *c* and *a* of 0.2PMN-0.8PZT MCA.

| | Unpoled fresh | Poled fresh | Poled Aged |
|---|---|---|---|
| Lattice parameter, *c* | 4.377 Å | 4.385 Å | 4.377 Å |
| Lattice parameter, *a* | 4.334 Å | 4.352 Å | 4.347 Å |

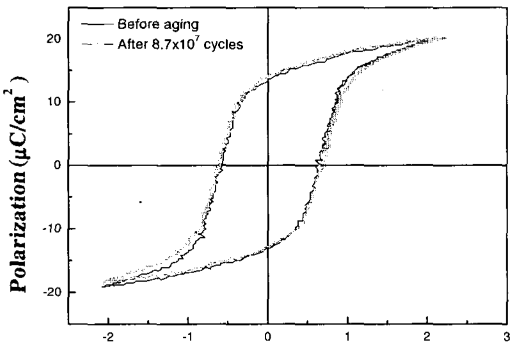


Fig. 3. Ferroelectric hysteresis of unpoled actuator before and after $8.7\times10^7$ cycles running at 60 Hz.

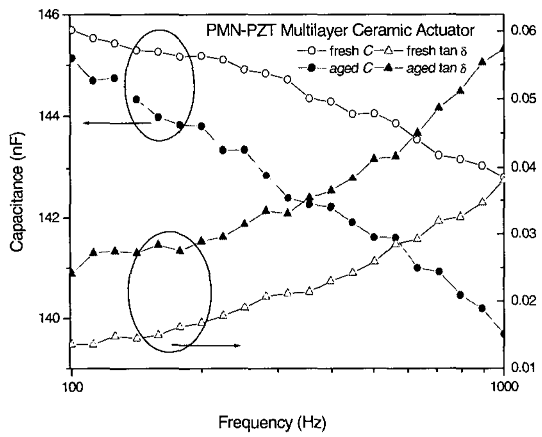


Fig. 4. Frequency dependent capacitance and tan δ of the actuator measured from 100 Hz to 1 kHz at room temperature. Symbol ○ and ● represent capacitance for unpoled fresh and aged actuators, respectively. Symbol △ and ▲ represent tanδ for unpoled fresh and aged actuators, respectively.

quency in the frequency ranges from 100 Hz to 1 kHz at room temperature in order to remove pyro-effect. The MCA showed degraded value of capacitance and loss tanδ after cyclings. As shown in the Fig. 4, the capacitance was decreased and tanδ was increased according to increase frequency. The capacitance was changed with 1.97 % and 3.76 % in the frequency range from 100 Hz to 1 kHz for unpoled fresh and aged MCA, respectively. This probably come from degradation of resistance and increased non-contact region due to the continuous piezo-motion. Since tanδ has close relation to the resistance of MCA, degraded resistance raised tanδ according to tanδ = 1/(wCR). There are reports on that the electrode migration of MCA is a main reason for crack growth[4]. But we would like to point out the decreased capacitance after aging instead of increased capacitance. This means that Ag-Pd electrode did not migrate due to high electric field.

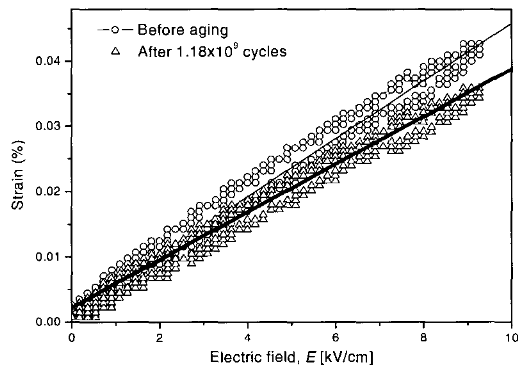


Fig. 5. Strain versus electric field characteristics of the MCA measured by laser vibrator. Symbol ○ and △ represent poled fresh and poled aged MCA, respectively.

Table 2. Resonance frequency, effective electromechanical coupling coefficient $k_p$, and pseudo-piezoelectric constant $d_{33}$ of poled 0.2PMN-0.8PZT MCA.

| Cycling number | 0 | $5.4\times10^8$ | $7.03\times10^8$ | $1.18\times10^9$ |
|---|---|---|---|---|
| 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 |

Figure 5 exhibits strain dependent on electric field of both poled fresh and poled aged MCA. If we assume piezoelectric motion of MCA follow longitudinal mode, then piezoelectric equations corresponding to the longitudinal extension are can be expressed as follow.

$$x_3 = s_{33}{}^{E} X_3 + d_{33} E_3 \qquad (1)$$

$$D_3 = d_{33} X_3 + \varepsilon_{33}{}^{X} E_3 \qquad (2)$$

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 $\varepsilon_{33}{}^{X}$ 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 MCA. As shown in the Fig. 5, pseudo-piezoelectric constant become decreased due to aging. Calculated pseudo-piezoelectric constants are written in Table 2. As shown in the Fig. 6, MCA loose its piezoelectric response due to degradation of MCA and depoling effect. This result well coincides with XRD data in Fig. 2. Pseudo-peizoelectric constant decreased 17.5 % after $1.18\times10^{9}$ unipolar cycles.

Figure 6(a) reveals the SEM images of destroyed surface of 0.2PMN-0.8PZT MCA. Small piece of electrode was observed in the surface of destroyed 0.2PMN-0.8PZT MCA. Since multilayer actuators have $d_{33}$ mode for their 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, this come from the Poisson's ratio to compensate volume expansion parallel to the electric field. Due to this accumulation of tensile stress effect, crack develops between the active and inactive region. Small crack in the Fig. 6(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. Figure 6(c) illustrates expansion direction and tensile stress result in crack of MCA.

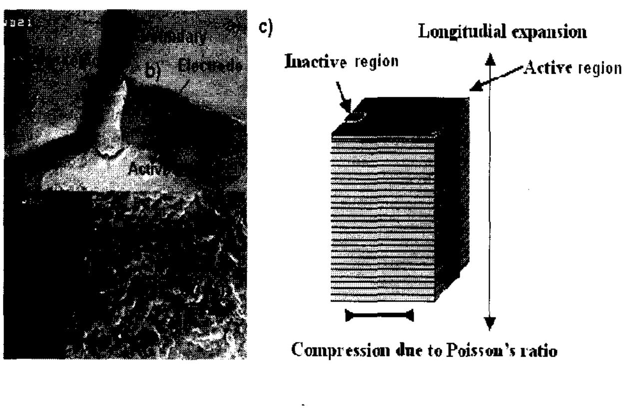


Fig. 6. Images of SEM, which show a) cracked surface of PMN-PZT MCA, b) crack line of destroyed MCA, and c) illustration of tensile stress generated within the MCA.

## 4. CONCLUSION

Aging behavior of 0.2PMN-0.8PZT MCA has been investigated by applying both triangular wave function for unpoled and unipolar wave function for poled MCA. *P-E* hysteresis loops were traced with a triangular shape electric field of 2 kV/mm to unpoled MCA. After $8.7\times10^{7}$ 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. We found that mechanical aging, the tensile stress come from both vertical $d_{33}$ expansion mode, and horizontal motion of Poisson ratio were mainly influenced on the performance of MCA.

## ACKNOWLEGMENT

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