Improvement in Durability of Piezoelectric Stack Actuator

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1. Introduction
Piezoelectric actuators using lead zirconate titanate (PZT) have potential for use of automobiles,1) because they offer quick response, compactness and low power consumption. Because of these desirable characteristics, application of these actuators is expected to increase. Change in the properties of PZT stack actuators has not been completely understood, although few studies have dealt with their durability in co-fired multilayer actuators.2,3) Because these actuators were complex structures consisting of PZT pellets, electrodes and insulators. Therefore research pertaining to these properties with the goal of developing high-durability PZT actuators is needed. High-durability PZT actuator is necessary to have the potential of no deterioration of displacement and no crack occurrence over 10⁸ cycles driving such application as fuel injector.1)

In a previous study, a new composition with a high coercive field and mechanical quality factor (Qm) was developed5) and changes in the properties of PZT decomposed pellets from the stack were analyzed. That study suggested that to investigate high-durability actuator, it is necessary to research a new composition which have high coercive field (Ec) at high temperature and less heat generation. The initial characteristics of 4 types of PZT compositions are shown in Table 1. The relative dielectric constant (ε33T/ε0), kp and Qm were calculated by the resonant-antiresonant frequency method with the impedance analyzer (Model YHP4194A, Yokogawa-Hewlett-Packard, Tokyo, Japan). The Tc was measured using an impedance analyzer connected to a measurement fixture in a furnace. The Ec was calculated from D-E hysteresis loops obtained from Sawyer-Tower circuit at 10 Hz under a prestress of 20 MPa at 30°C. Sample A is the basic composition. Sample B exhibited improved bending strength of 40% by the introduction of the fine pulverizing process and oxygen pressure hot isostatic press

Table 1. Initial Properties and Compositions of Samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>ε33/ε0 (%)</th>
<th>kp</th>
<th>Qm</th>
<th>Tc (°C)</th>
<th>Ec (V/mm)</th>
<th>Bending strength (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>3700</td>
<td>72</td>
<td>60</td>
<td>230</td>
<td>1150</td>
<td>90</td>
</tr>
<tr>
<td>B</td>
<td>3100</td>
<td>65</td>
<td>65</td>
<td>242</td>
<td>1050</td>
<td>126</td>
</tr>
<tr>
<td>C</td>
<td>3460</td>
<td>73</td>
<td>61</td>
<td>246</td>
<td>1270</td>
<td>95</td>
</tr>
<tr>
<td>D</td>
<td>2690</td>
<td>68</td>
<td>112</td>
<td>282</td>
<td>1360</td>
<td>102</td>
</tr>
</tbody>
</table>

A: Pb0.88Sr0.10Ba0.02(Zr0.542Ti0.339Sb0.009Mn0.09In0.02)O3
B: Pb0.88Sr0.10Ba0.02(Zr0.542Ti0.339Sb0.009Mn0.09In0.02)O3
C: Pb0.88Sr0.10Ba0.02(Zr0.542Ti0.339Sb0.009Mn0.09In0.02)O3
D: Pb0.88Sr0.10Ba0.02(Zr0.542Ti0.339Sb0.009Mn0.09In0.02)O3

1) calculated from D-E hysteresis loops obtained from Sawyer-Tower circuit at 30°C under a prestress of 20 MPa
2) measured using a three-point-bending fixture with a 10 mm span
(O$_2$-HIP) treatment. Sample C exhibited improved $E_c$ of 10% by addition of 0.2 mol% acceptor element. Sample D exhibited improved $E_c$ of 10%, $Q_m$ of 100% and $T_c$ by addition of 1.1 mol% acceptor element. Figure 1 shows the relationship of relative potential for these samples.

The structure of the stacks is multilayer of poled pellets. Stainless steel inner electrodes were inserted between the PZT pellets. Sixty PZT pellets were then stacked, with two insulator pellets at each end, and the stainless steel inner electrodes were connected to leads to supply a driving voltage.

2.2 Displacement measurement and durability testing method

The stack actuator was inserted between flat plates, which were connected to a hydraulic pump. Displacement of the stack actuator was achieved under preloading conditions using two gap sensors with the examined resolution of 0.2 μm. The equipment was placed in an oven under a controlled temperature at 120°C. Displacement measurements were performed under a driving energy of 120 mJ having a 130 Hz pulse wave. Both the conductor and capacitor were inserted between power source and actuator. The circuit includes both voltmeter and ammeters to determine energy output from the power source and energy input to the actuator.

The pulse wave of output voltage from the power source was applied and the output voltage introduced current wave. The output energy ($E$) from the power source was calculated from Eq. (1), using the voltage ($V$), current ($I$) and flowing time ($t$). The current increased with increasing voltage. However, smaller current was observed after the voltage reached a maximum.

$$E = V \int I \, dt$$

Using this circuit, the heat generation caused by the displacement hysteresis is prevented by keeping to the constant energy. The dynamic stack durability test was used on the displacement measurement system. Test conditions consisted of 20 MPa prestress at 120°C under a driving energy of 120 mJ with a 100 Hz pulse wave. After testing, the stack displacement was measured and $k_p$ was measured at the decomposed pellets. The X-ray intensity was expected at the proportion of $I_{002} = I_{002}/(I_{002} + I_{000})$.

2.3 Stack temperature and current–voltage properties

The differential stack temperature was defined as the difference in temperature between different places in the stack actuator. The temperature of the outer edge of the pellets at the center (pellet No. 30) and at the upper-end (pellet No. 1) of the stack were measured by an infrared thermometer for starting temperature. The current–voltage properties of power circuit were measured and concluded the relationship between heat generation.

The influence of the driving method was measured by using stack B. Two driving methods were performed, one was a voltage-control from −400 to 1200 V/mm and another was energy-control of 120 mJ having a 130 Hz pulse wave. The influence of $Q_m$ and $T_c$ values was measured by comparison with stacks C and D under a driving of energy-control.

3. Results and discussion

3.1 Dynamic durability tests of stack actuator

Figure 2 shows changes in displacement for the stack actuator after dynamic durability tests. Stack A exhibited 28% deterioration in displacement after 10⁸ cycles which was caused by self-heating. Stack B exhibited 16% deterioration in displacement after 10⁸ cycles. It is rather effective for inhibition of displacement decrease to the change of driving method from voltage-control to energy-control for the same stack actuator. Stack B exhibited 30% deterioration in displacement after 10⁸ cycles by voltage-control driving in a previous study Ref. 4). On the other hand, there was small self-heating, therefore, 6–2% deterioration in displacement were observed in stacks C and D.

To identify the reason for these results, the stacks were decomposed to pellets and the properties of each pellet were measured. Figure 3 shows the dependence of $k_p$ and polarization ($P_r$) on number of repetition. Both $k_p$ and $P_r$ decreased with the increase of the cycles. The bending strength of pellets decreased with the increase of cycles as shown in Fig. 4. Then, it is considered that the internal microcrack occurred due to the lattice mismatch introduced by repeated domain rotation. Figure 5(a) shows the image of optical microscopy for the surface of positive electrode decomposed pellet after 10⁸ cycles of stack B. The similar crack growth was observed on all of the decomposed pellets after 10⁸ cycles of stack B. The white lines are cracks growing from the edge of circle because of maximum tensile stress caused by the 20 MPa prestress to flat surface direction. No cracks were observed on the surface of negative electrode. It is considered that the crack occurred at the edge of positive side of pellet due to maximum tensile stress.
at the edge of pellet and grown to the center of diameter concluding from the crack structure. Figure 5 (b) shows the scanning electron microscope (SEM) image of cross section of the edge of positive electrode. The crack occurred at the edge of positive side of PZT pellet and grown to the center of diameter and negative side. The crack depth from the positive surface was 150 μm. It is considered very slow crack growth because of crack length of 150 μm during 10^8 cycles. This does not mean brittle fracture for ceramics. Cyclic domain rotation at high temperature is considered the reason of this slow crack growth. Figure 6 shows the X-ray diffraction patterns of the surface of (002) and (200) of PZT pellets without Ag electrode by chemical method. It was observed that same peak angles of the deteriorated positive surface changed slightly toward the high angle of the poling sample. It was meant the tensile stress direction to the sample surface. It was considered that the tensile stress decided by the final domain direction as c-axis orientation. It was observed that same peak angles of the deteriorated negative surface changed slightly toward the low angle of the poling sample. It was meant the compressive stress direction to the sample surface. It was considered that the compressive stress decided by the final domain direction as a-axis orientation. At the poled sample, the same pattern was observed both positive and negative surfaces, however at the tested sample, the intensities of each surface were observed large difference. Positive surface was shown c-axis direction and negative surface was shown sintered pattern. Then the relative ratio of I_c expressed X-ray intensity shown in Fig. 7. c-axis direction on the deteriorated sample means 180° domain rotation parallel to the poling direction considered with several examples of deteriorated PZT specimens. It does not mean the repoling considered from the decrease of k_p and P_r as shown in Fig. 3.

Then we can consider the crack growth model on low E_r sample as shown in Fig. 8. The crack occurred at the edge of positive side of PZT pellet due to tensile stress at the edge of its and grown to the center of diameter and negative side due to markedly domain rotation on positive side caused by changing voltage compare with small change on negative side as earth.

3.2 Property of decomposed pellets from stacks C and D

Figure 9 shows the k_p of decomposed pellets from stacks C and D after dynamic stack durability tests of 10^8 cycles. k_p decreased by 15% at the pellets of stack C. In previous study, the marked k_p decrease was observed by an increase of driving temperature. It is considered that the reason of k_p decrease was the temperature increase during the durability tests. The decrease of k_p was accompanied by depoling. The deterioration in the displacement of stack C appeared to be the result of decreased displacement in the pellets. On the other hand, the k_p decreased by 8% at the pellets near the center of the stack D. It appears that the Q_m and T_c values have a strong influence on durability in addition of E_r and bending strength treated in Refs. 4, 9 and 11.

3.3 Change of stack temperature

Figure 10 shows the time dependence of stack temperature caused by a different driving method of stack C. In voltage-control case, the increase of temperature...
reached a maximum after 5 min. The temperature of pellets placed at the center of the stack increased markedly. The increase in temperature was 57°C and was higher at the center of the stack than at the upper-end. However, in energy-control case, the increase of temperature was 44°C, a result of lower self-heating. It is considered that the low self-heating is caused by a small increase of current during driving. Figure 11 shows the time dependence of stack temperature caused by energy-control driving for stacks C and D. The temperature of stack center was lowered 14°C compare with sample D and C. Using this conductor-capacitor circuit, the heat generation caused by the displacement hysteresis is suppressed by keeping to the constant energy. The capacitance of the stack actuator increased with temperature showing that it was affected by the self-heating of the stack actuator, which in turn depended on the displacement hysteresis. However, the displacement hysteresis was held to a minimum by controlling the energy.

3.4 Change of stack temperature and current–voltage properties for driving

Figure 12 shows the properties of voltage and current at stack C. $I_{max}$ is the current value, which flow to conductor–capacitor circuit from power source. $I_t$ is the time of current flowing to this circuit. These were increased since driving start. This tendency is the same for the increase of stack temperature shown in Fig. 11. The increase of $I_{max}$ and $I_t$ are considered because of the repeated domain rotation or displacement hysteresis loop in the low Qm and Tc sample. The increase of $I_{max}$ and $I_t$ are considered because of the increase of stack capacitance, once the temperature increase. In the Eq. (1), the voltage will be decreased in the increase of $I_{max}$ and $I_t$ if Adt, if the energy kept to constant. Therefore the displacement of actuator will be decreased under driven at high temperature and frequency. The stack increased $I_{max}$ and $I_t$ considerably was considered difficult of prevention the decrease of displacement, although driv-
ing by conductor–capacitor circuit.

On the other hand, Fig. 13 shows the stack D properties of voltage and current. The decrease of total voltage $V_{pp}$ was small at 60 V compared with 80 V in stack C, because of the least possible increase in $I_{max}$ and $I_{t}$. Therefore, in the PZT at high $Q_m$ and $T_c$, the small decrease of displacement driven at high temperature and frequency was indicated by the current–voltage properties flowing in the conductor–capacitor circuit.

4. Conclusions

A study of durability test using a piezoelectric stack actuator having various values of $Q_m$, $T_c$, $E_c$ and bending strength led to the following conclusions.

(1) The high bending strength but low $E_c$ stack showed a decrease of displacement and appeared cracks growing from the edge of positive side. The crack occurred at the edge of positive side of PZT pellet and grown to the center of diameter and negative side. It is considered very slow crack growth because of crack length of 150 µm during $10^8$ cycles. Cyclic domain rotation at high temperature is considered the reason of this growth, never mean brittle fracture for ceramics.

(2) The decomposed pellets from the high $E_c$ but low $Q_m$ stack appeared deterioration of $k_p$. The increase of temperature of low $Q_m$ stack was larger than that of high $Q_m$ stack for a starting temperature of 120°C with 130 Hz drive. It is considered that the low self-heating is caused by a small domain rotation introduced by more addition of the acceptor element. The result of this small self-heating, 2% deterioration in displacement was observed in high $Q_m$ and $T_c$ stack.

(3) The change of current–voltage properties of the high $Q_m$ and $T_c$ stack appeared to be small compared with the low $Q_m$ and $T_c$ stack. It appears that $Q_m$ and $T_c$ values have a strong influence on durability in addition of $E_c$ and bending strength.

References