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Device and Piezoelectric Characteristics of $Pb(Mn_{1/3}Sb_{2/3})O_3$-PZT Ceramics for Piezoelectric Transformer

Joon Ho Sohn, Soo Jeong Heo, Jeong Ho Sohn*, Joon Hyung Lee, Woo Hwan Jung, Dong Bum Kim*** and Sang Hee Cho**

Department of Inorganic Materials Engineering, Kyungpook National University, Taegu 702-701, Korea

**Department of Electronic Ceramics Engineering, Kaya University, Koryong, Kyungpuk 717-800, Korea*

***Department of Electronic Engineering, Howon University, Koonsan, Jeonpuk 573-718, Korea*

****LG C & D Ltd., R & D Lab., Osan, Kyungki 447-150, Korea*

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In the $(Pb_{1-x}M_x)[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_yTi_{0.95-y}]O_3$ system, where M=Ca and Sr, the piezoelectric properties were evaluated to examine the possibility of application to piezoelectric transformer. A Rosen-type piezoelectric transformer was formed, then the electrical properties of voltage step-up ratio, frequency characteristics etc. were analysed. The morphotropic phase boundary was determined to be y=0.475 in $Pb[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_yTi_{0.95-y}]O_3$ system, and the piezoelectric properties of this composition was k_p=0.59, Q_m=1600 and ε_r=1150. Moreover, when 1-2 mol% of Sr are substituted, enhanced piezoelectric properties of k_p=0.61, Q_m=1600 and ε_r=1400 were shown. The temperature rising ($\triangle T$) of a piezoelectric transformer with $Pb[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_{0.475}Ti_{0.175})]O_3$ composition was 10°C, and the voltage step-up ratio was 500 when the output voltage was 4000 V, whereas the $\triangle T$ was below 3°C and the resonant frequency variation ($\triangle f_r$) as a function of load resistance was below 5% when the output voltage was 2000 V. These characteristics are superior to the properties of materials, which were substituted by Ca or without substitution.

Key words : Piezoelectric Transformer, Voltage Step-up Ratio, Temperature Rising

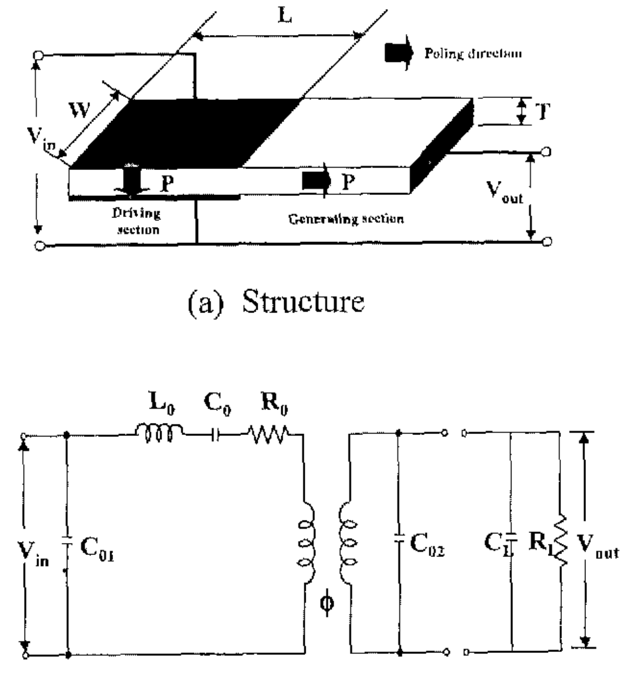
I. Introduction

Recently, piezoelectric ceramics have been widely used as elements not only low-power devices such as buzzer, filter, resonator, ignitor, speaker, *etc.* but also high-power devices such as transformer, ultrasonic motor, transducer, actuator, *etc.*[1-3] Among these various application areas, a study on piezoelectric transformer was originally reported by Rosen[4] at the end of 1950. In 1960's the application of piezoelectric transformer for horizontal deflection circuits of TV receivers was investigated. However, because of temperature rising from piezoelectric transformer and low efficiency, it could not put into practical use at that time.

At the recent few years, the research on piezoelectric transformer has focused to increase the efficiency of the piezoelectric transformer through the development of high power piezoelectric ceramic materials, multi-layer transformer,[5-6] electrode pattern,[7-8] and driving circuit.[9] Finally, small but high power piezoelectric transformers, for example, inverters for the backlight of color LCD panels, are realized.[10] However, as the size of display panel increases, smaller and thinner piezoelectric transformer with high efficiency than the coil-type transformer is necessary.

Ceramic materials for manufacturing excellent piezoelectric transformers demand high electromechanical coupling factor (k_{31}, k_{33}) and high mechanical quality factor (Q_m). One of the most important concerns in the piezoelectric transformer is the voltage step-up ratio(γ=V_{in}/V_{out}), which is proportional to Q_m, k_{31}, k_{33}, and length/thickness ratio of transformer. Since the length/thickness ratio of the transformer is almost fixed by vibration mode and mechanical strength of the material, high Q_m, k_{31} and k_{33} are necessary. While, low tanδ is strongly preferred to reduce losses. Much work has been performed to improve the piezoelectric properties of materials. PZT-related system such as $Pb(Mg_{1/3}Nb_{2/3})$-PZT, $Pb(Sb_{1/2}Nb_{1/2})$-PZT, $Pb(Mn_{1/3}Nb_{2/3})$-$Pb(Mg_{1/3}Nb_{2/3})$-PZT, $Pb(Mn_{1/3}Sb_{2/3})O_3$-PZT, *etc.* are the materials that can be used for transformer. Among these materials, $Pb(Mn_{1/3}Sb_{2/3})O_3$-PZT system reported by Ohno et al.[11] is suitable for low temperature sintering, so this material is highly applicable for multilayer piezoelectric transformer.

In this study, $(Pb_{1-x}M_x)[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_yTi_{0.95-y}]O_3$ (M=Ca and Sr) system, a strong candidate material for piezoelectric transformer, was employed and the piezoelectric properties of the material with A-site substituents of Ca and Sr were examined. A Rosen-type piezoelectric transformer was formed and the voltage step-up ratio (γ),



(a) Structure

(b) Equivalent Circuits

Fig. 1. Structure and simplified equivalent circuits of ceramic transformer.

frequency characteristics, heat generation phenomena etc. were analysed.

II. Piezoelectric transformer

1. Principle and structure of piezoelectric transformer

The structure of transverse type piezoelectric transformer proposed by Rosen[4] and its equivalent circuit near resonant frequency are shown in Fig. 1. Combining the direct and inverse piezoelectric effects in the primary side of the transformer, an external voltage results in a mechanical strain and stress which propagate through the product, then the stress generates voltage (electric field) at the secondary section. This voltage generation occurs by the poisson coupling of transverse and longitudinal vibration of piezoelectric ceramics at driving and generating section, respectively. When the vibration frequency matches with the resonance frequency of secondary section of piezoelectric ceramics, which is dependent the dimension of piezoelectric ceramics, voltage generation occurs at the secondary section. Therefore, piezoelectric transformer properly operates near the resonant frequency only and this is a major difference with the coil-type transformer.

2. Equivalent circuits of piezoelectric transformer

In the equivalent circuits shown in Fig. 1(b), L_o, C_o and R_o represents equivalent mass, equivalent compliance, and equivalent mechanical resistance near resonant frequency, respectively. C_{o1} and C_{o2} is clamped capacitance of driving and generating section. Φ is turn ratio of ideal electromechanical transformer, and C^E is sound velocity in ceramics. These individual equivalent circuit constants can be calculated by Eq. (1).[12]

Table 1. Calculation Values of Equivalent Circuits Constants

Equivalent circuits constants	Calculation values
L_o	34.80 mH
C_o	91.35 pF
R_o	108.4 pF
C_{o1}	1787 pF
C_{o2}	11.56 pF
Φ	15.84

$$L_o = \frac{LT}{4C^{E2} d_{31}^2 Y_3^E W} \qquad C_{o1} = \frac{\varepsilon_{33}(1-k_{31}^2)WL}{T}$$

$$C_o = \frac{4Wd_{31}^2 Y_3^E L}{\pi^2 T} \qquad C_{o2} = \frac{\varepsilon_{33}(1-k_{33}^2)WT}{L}$$

$$R_o = \frac{\pi T}{4C^E Q_m Y_3^E d_{31}^2 W} \qquad \Phi = \frac{L}{T}\frac{k_{31}}{k_{33}} \tag{1}$$

Table 1 shows the equivalent circuit constants of a sample with y=0.475 calculated by Eq.(1). The secondary section of equivalent circuits is composed of load resistance (R_L) and stray capacitance (C_L, C_{o2}). Eq. (2) can be expressed when we assign the capacitance ratio of C_L/C_{o2} is a, mechanical characteristics impedance is Q_2, where C_2 is total capacitance of secondary section and fr is driving frequency.

$$C_2=C_{O2}+C_L=C_{O2}(1+a)$$
$$Q_2=2\pi f_r C_2 R_L \tag{2}$$

When R_L is ∞, γ_∞ is given in Eq. (3), and the γ of the equivalent circuit of Fig. 1(b) is expressed in Eq. (4).

$$\gamma_\infty = \frac{1}{\Phi f_r C_o^2 R_o} \tag{3}$$

$$\gamma = \frac{\gamma_\infty}{1+a}\frac{\sqrt{1+Q_2^2}}{1+Q_2^2+(1+a)^{-2}\gamma_\infty^{\,2}\frac{R_o}{R_L}} \tag{4}$$

3. Heat generation phenomena

Energy transformation from electrical to mechanical accompanies various losses.[13,14] One of them is heat generation from ceramic body caused by vibration. The temperature rising of piezoelectric ceramics by the heat generation restricts practical operation of transformer in some circumstances. Many studies have been carried out to reduce the temperature rising through the improvements in material characteristics and driving conditions. For the evaluation of heat generation, temperature rise ($\triangle T$) at ceramic surface is usually measured.

Ikushima[14] reported that the heat generation at large amplitude operation is attributed to the conversion from a part of elastic energy to heat energy by internal friction. Nagata et al.[15] reported a relationship between Q_m and temperature rising, and showed that the temperature rising was decreased as the Q_m increased in various piezoelectric transformers. On the other hand, Takahashi et al.[15] reported that the temperature rising is proportional to vibration loss. As reported earlier in many papers, the temperature rising of piezoelectric transformer became an important issue since it restricts the practical operation of piezoelectric transformer.

III. Experimental Procedure

High purity chemicals of PbO, ZrO_2, TiO_2, Mn_3O_4, Sb_2O_3, $SrCO_3$ and $CaCO_3$ were used for starting raw materials. Ca and Sr were substituted in the basic composition of $(Pb_{1-x}M_x)[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_yTi_{0.95-y}]O_3$, where M=Ca and Sr, $0.00 \leq x \leq 0.04$, $0.435 \leq y \leq 0.485$. The weighed powders were wet mixed for 24 hrs in a plastic jar with zirconia ball. After drying, the powders were calcined at 850°C for 2 hrs in alumina crucible, then ball milled again for crushing. The powder was mixed with aqueous PVA solution and sieved to form granules. Green pellets of disk and rectangular plate shape were formed at uniaxial pressure of 1.5 ton/cm². The samples were sintered in a covered alumina crucible at 1000°C~1250°C for 2 hrs with $PbZrO_3$+10 wt% ZrO_2 powder. Both surfaces of sintered body with a dimension of $10^{\phi} \times 0.6^{t}$ disk were covered with silver paste (dupont #7095), and heat treated at 600°C for 10 min. For the rectangular plate samples with a dimension of $40 \times 6.5 \times 0.65$ mm³, silver paste wes shaped to the Rosen type piezoelectric transformer. The samples were poled at DC 30 kV/cm, and the piezoelectric properties were analysed by IRE standards. X-ray diffractometer (Rigaku) and SEM (JEOL 4500) were used for phase identification and microstructure observation, respectively. Resonant and anti-resonant frequencies (f_r, f_a) were measured from room temperature to 250°C, and planar coupling factor (k_p) at each temperature was analysed. The properties of piezoelectric transformer was analysed with double rectifying circuits at λ(full-wave) mode, which has high γ. Thin thermocouple wire was positioned at the generate section of piezoelectric transformer to measure temperature rising from sample. Fig. 2 shows the schematic of measuring circuits.

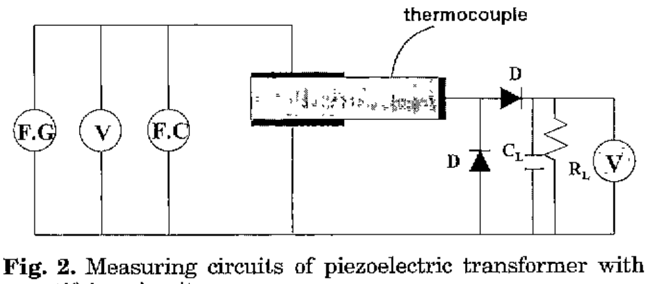


Fig. 2. Measuring circuits of piezoelectric transformer with a rectifying circuit.

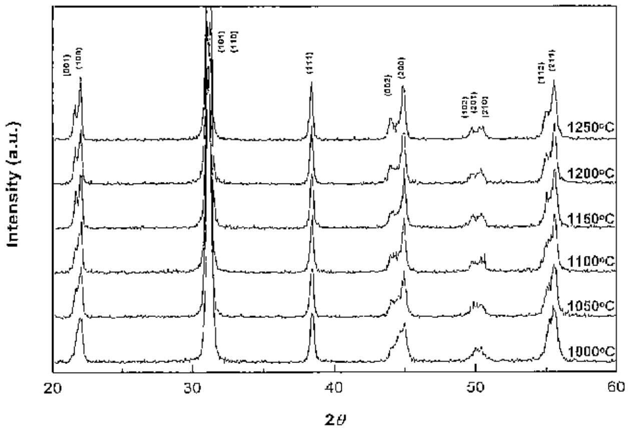


Fig. 3. X-ray diffraction patterns of $Pb[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_{0.475}Ti_{0.475}]O_3$ samples sintered at various temperatures for 2 h.

IV. Results and Discussion

1. Material Characteristics

1.1 $Pb[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_yTi_{0.95-y})]O_3$ system

Fig. 3 shows powder X-ray diffraction pattern of samples with a composition of Zr=0.475 as a function of sintering temperature. All of diffraction peaks were identical with the perovskite crystalline structure, and the crystal symmetry was changed from tetragonal to rhombohedral as the content of Zr increased. It is generally known that 100% of perovskite PZT can hardly be produced from PbO-ZrO_2-TiO_2 mixture even though it is calcined at relatively high temperature of 850°C. The X-ray diffraction results were not shown here though, every powder calcined at 850°C revealed 100% perovskite. This result signifies that the $Pb(Mn_{1/3}Sb_{2/3})O_3$ somehow helped low temperature synthesis, and the low temperature sintering was thought to be available at 1000°C as shown in Fig. 3. This result can be extended for the application to multilayer piezoelectric transformer.

Fig. 4 shows microstructure of samples sintered at 1175°C as a function of Zr content. As the content of Zr increased, the average grain size was decreased from 2.7 to 2.1 μm. This result is in a good agreement with other study that $PbTiO_3$ enhances grain growth. Fine and uniformally distributed grain size of microstructure is necessary to the proper operation of piezoelectric transformer, and the resultant microstructure of sintered samples shown in Fig. 4 meets the needs of piezoelectric transformer.

Fig. 5 represents k_p, Q_m and dielectric constant (ε_r) of samples sintered at 1175°C as a function of Zr content. At y=0.475, where is believed the morphotropic phase boundary, a good piezoelectric characteristics of k_p=0.59, Q_m=1600 and ε_r=1150 were shown.

1.2 $(Pb_{1-x}M_x)[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_yTi_{0.95-y}]O_3$ (M=Ca and Sr) system

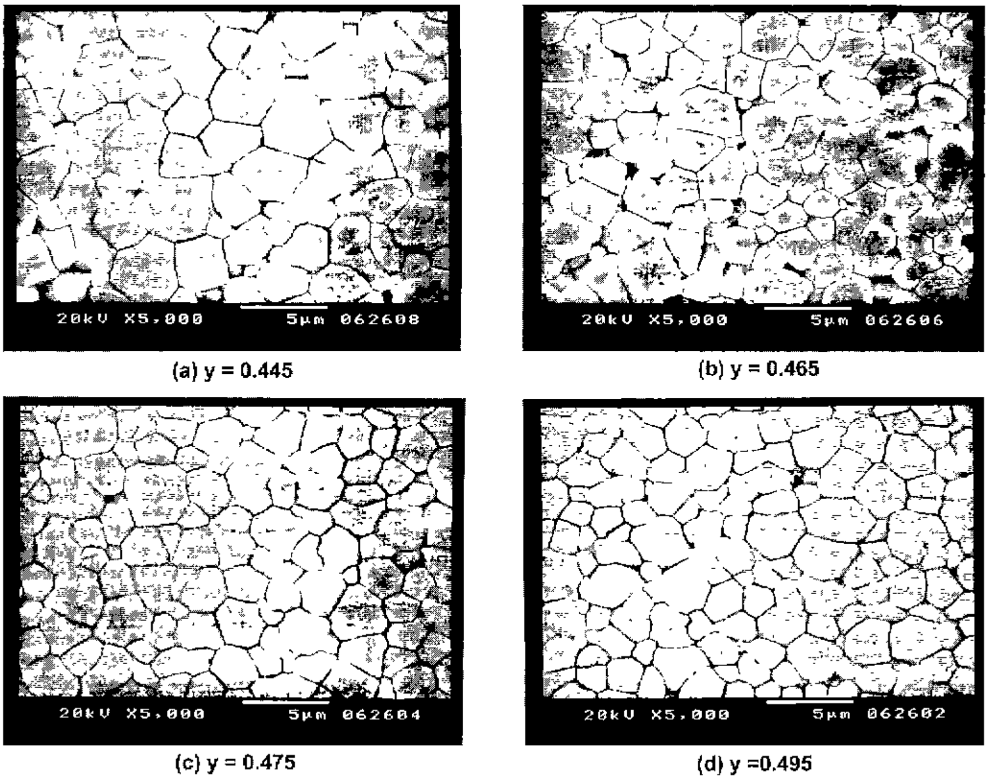


Fig. 4. Microstructures of $Pb[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_yTi_{0.95-y}]O_3$ samples as a function of Zr content. The samples were sintered at 1175°C for 2 h.

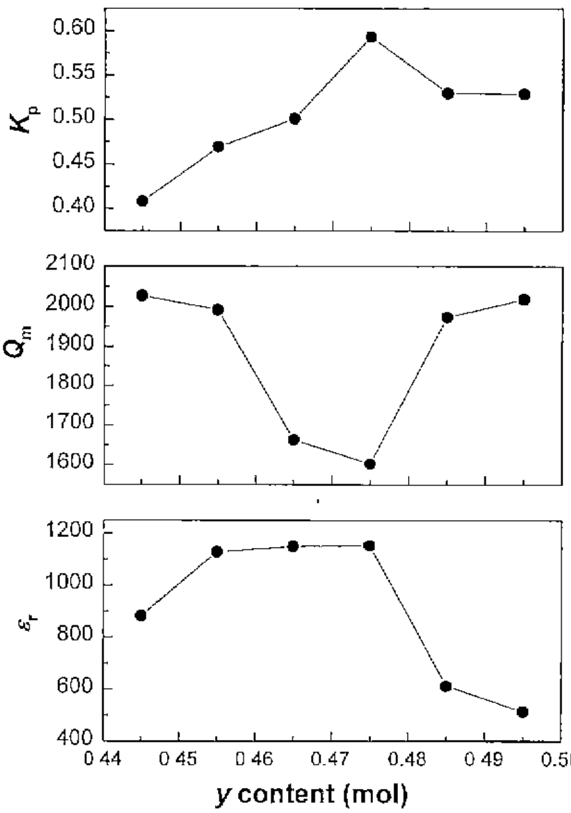


Fig. 5. Piezoelectric properties of $Pb[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_yTi_{0.95-y})]O_3$ samples as a function of Zr content. The samples were sintered at 1175°C for 2 h.

Ions of alkali-earth metals, Ca^{2+} and Sr^{2-}, which have ionic radii of 1.06, 1.27 and 1.43 Å, respectively, are frequently used to substitute Pb^{2+}(with an ionic radius of 1.32 Å). Using element substitution, one can change properties of the piezoelectric ceramic, but still maintain its perovskite structure.

Fig. 6 shows piezoelectric properties of samples as a function of substitution content. In the case of Sr and Ca substitution, k_p was decreased and this is believed to be caused by decreased tolerance factor. Especially, greater decrease of k_p was observed in Ca substitution. This result can also be explained by the viewpoint of tolerance factor. When 1 mol% of Sr was substituted, ε_r and k_p were slightly increased. This result was previously reported by Kulcsar[17] and was explained by definite fluxing effect. Great increase of Q_m at 3 mol% of Ca substitution is believed due to decreased dielectric constant. However, the overall electrical properties of k_p, Q_m and ε_r in specific substitutional content are quite good for application of piezoelectric transformer.

Fig. 7 shows the variation of k_p ($\triangle k_p$) of poled samples as a function of temperature. Decreasing of k_p by temperature increasing is one of the most important factor to be considered in piezoelectric transformer, which is operating at high electrical power. When the temperature of piezoelectric transformer is increased, the aligned domains randomly reorient, which is so called depoling effect. Pure composition without substituents showed slow de-

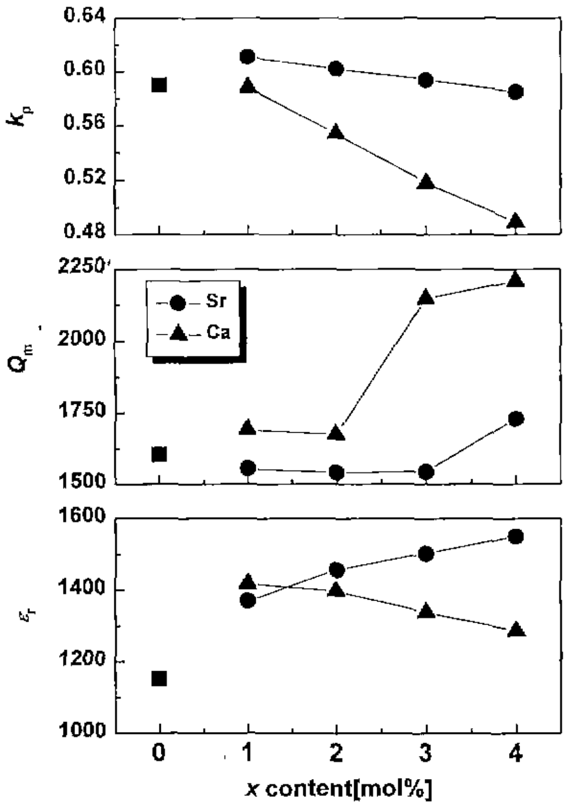


Fig. 6. Piezoelectric properties of $(Pb_{1-x}M_x)[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_{0.475}Ti_{0.475}]O_3$ samples (M=Ca and Sr) as a function of substituents content. The samples were sintered at 1175°C for 2 h.

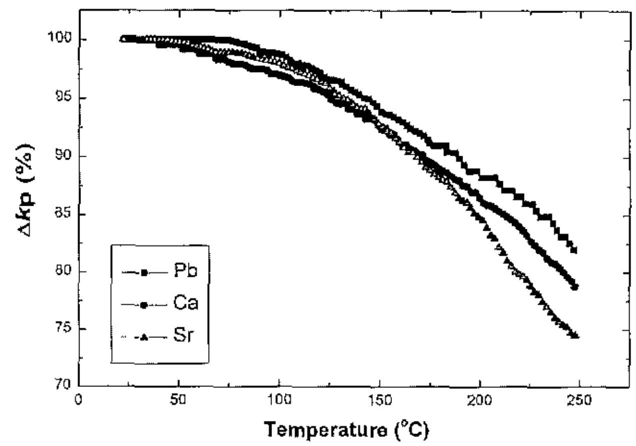


Fig. 7. Temperature dependence of k_p of $(Pb_{0.99}M_{0.01})[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_{0.475}Ti_{0.475}]O_3$ samples with M=Pb, Ca and Sr. Samples were sintered at 1175°C for 2 h.

crease of k_p from 70°C and temperature dependence of k_p in the whole temperature region was smaller than that of A-site substituted composition. This result is closely related to the temperature rising of piezoelectric transformer during operation and it provides a substantial data for the selection of material which is available for the piezoelectric transformer.

2. *Properties of piezoelectric transformer*

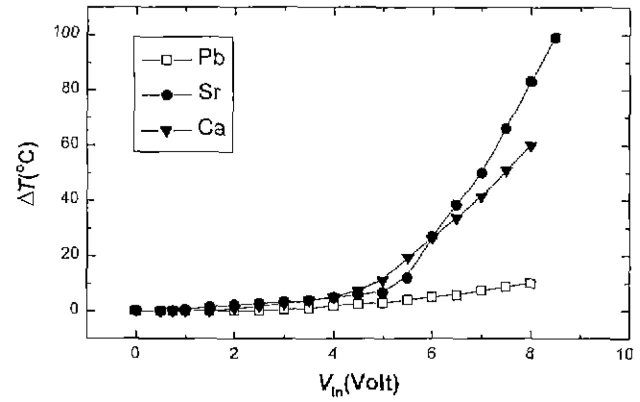


Fig. 8. Temperature rise($\triangle T$) of $(Pb_{0.99}M_{0.01})[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_{0.475}Ti_{0.475}]O_3$ samples (M=Ca and Sr) as a function of input voltage (V_{in}).

Fig. 8 shows the input voltage (V_{in}) dependence of heat generation at the surface of samples. In the case of pure composition, temperature rise($\triangle T$) up to 8 V of V_{in} was negligible. However, a drastic heat generation from 5 V was observed at the surface of samples substituted by Ca and Sr. This result seem to have close relationship with the variation of k_p as a function of temperature shown in Fig. 7.

Takahashi et al.[13,18] reported that heat generation occurs due to the increase of vibration level in finite-amplitude (large-amplitude) piezoelectric vibration, and a great heat generation occurs at a certain vibration level. Temperature rising is proportional to the vibration loss, which is dependent to the composition of materials, and the vibration loss is proportional to the square of vibration velocity. Therefore, when the isovalent ions are substituted in this experiment, the vibration loss was changed and the temperature abruptly increased at critical vibration velocity (5 V of V_{in} in this study). The decrease of Q_m also seem to be caused due to the temperature rising of the ceramics.

Fig. 9 shows the voltage step-up ratio (γ) as a function of R_L. In the relationship between load resistance (R_L) of piezoelectric transformer and γ, Wada et al.12) devide it into 3 areas. In this study, the γ values calculated from equivalent circuit constants and Eq. (4) were shown in Fig. 9, and the result is identical to the experimental results. When R_L was below 10 MΩ, γ revealed almost the same result of k_p and Q_m as shown in Fig. 6. While over 10 MΩ, γ of the composition with isovalent substitution was half of that of the composition without substitution. This result can be explained by the $\triangle k_p$ and $\triangle T$ shown in Fig. 7 and Fig. 8, respectively. That is, γ was decreased by the deteriorated piezoelectric properties due to the temperature rising at the critical value.

Another important characteristics of piezoelectric transformer is that the V_{out} depends on the operational frequency of V_{in}. Fig. 10 shows the frequency dependence of γ as a function of R_L in the sample of Zr=0.475. If the

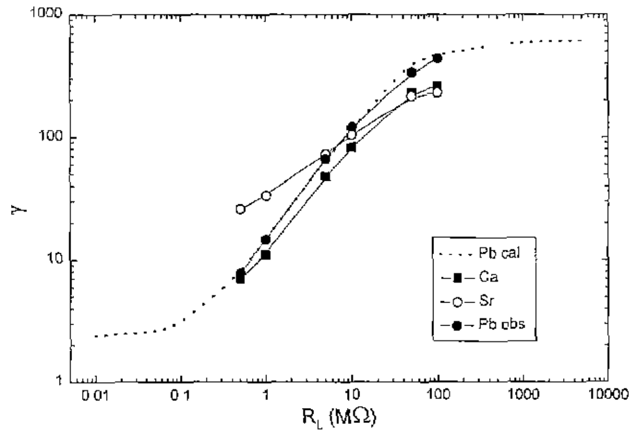


Fig. 9. The step-up voltage of $(Pb_{0.99}M_{0.01})[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_{0.475}Ti_{0.475}]O_3$ samples (M=Ca and Sr) as a function of load resistance.

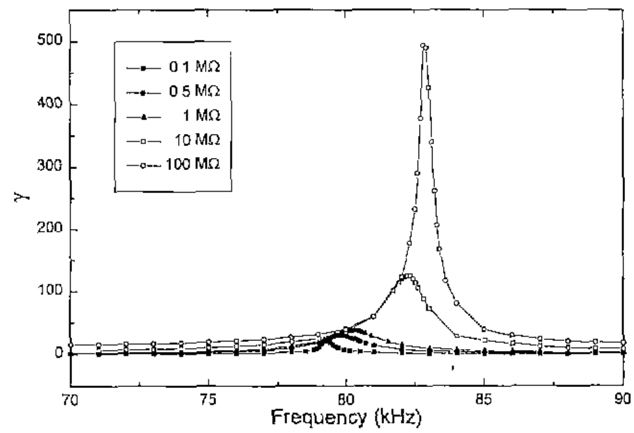


Fig. 10. Frequency dependence of $Pb[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_{0.475}Ti_{0.475})]O_3$ system as a function of load resistance.

dimension of ceramics and piezoelectric constants are fixed, f_r depends on R_L and C_L, and maximum output voltage appears at f_r. Besides, output voltage increased greatly as R_L increased. As the RL increased the broad resonance curve changed to sharp and the resonant frequency was increased. This phenomena shows good agreement with a literature of Wada et al.,[12] which was caused by elastic vibration of piezoelectric transformer.[13] In the case of $R_L=\infty$, γ will abruptly increased, and vibration velocity also will increased drastically by the influence of γ. Consequently self-failure of transformer might be occur by great internal stress. In the composition of Zr=0.475, $\triangle T$ was 3°C and $\triangle f_r$ was below 5% when V_{out} was 2000 V.

V. Conclusions

Systematic examination on the piezoelectric properties of $(Pb_{1-x}M_x)[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_yTi_{0.95-y}]O_3$ (M=Ca and Sr) system was carried out, and the application to the piezoelectric transformer was considered. A Rosen-type piezoelectric transformer was formed and the voltage step-up ratio, frequency characteristics, and temperature rising phenomena were analysed.

1. In $Pb[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_yTi_{0.95-y})]O_3$ system, the composition of morphotropic phase boundary was $Pb[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_{0.475}Ti_{0.475})]O_3$ where Zr=0.475. The piezoelectric characteristics of k_p, Q_m and ε_r showed 0.59, 1600 and 1150, respectively. This composition can be a strong candidate material for multilayer piezoelectric transformer since only 100% of perovskite phase was observed at 1000°C, which enables low temperature sintering.

2. The piezoelectric properties of $(Pb_{1-x}M_x)[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_{0.475}Ti_{0.475}]O_3$ (M=Ca and Sr) system, where 1-2 mol% of Sr is substituted, the piezoelectric properties of k_p, Q_m and ε_r were 0.61, 1600 and 1400, respectively. Those values are quite good characteristics to be used for piezoelectric transformer.

3. In the composition of $Pb[(Mn_{1/3}Sb_{2/3})_{0.05}Zr_{0.475}Ti_{0.475})]O_3$, temperature rising ($\triangle T$) was below 10°C and the voltage step-up ratio was 500 when the output voltage was 4000 V. At the 2000 V of output voltage, $\triangle T$ was 3°C and $\triangle f_r$ was below 5%.

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