Cerium Oxide 첨가에 따른 압전트랜스포머용 $Pb(Mn_{1/3}Nb_{2/3})O_3-Pb(Zr,Ti)O_3$ 세라믹의 압전특성

論 文
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Piezoelectric Characteristics of $Pb(Mn_{1/3}Nb_{2/3})O_3-Pb(Zr,Ti)O_3$ Ceramics with CeO_2 Impurity for the Piezoelectric Transformer

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Abstract-Piezoelectric properties of PMN-PZT ceramics with CeO_2 impurity were investigated. Mechanical quality factor, Q_m of 1792, 1285 and the electromechanical coupling coefficient, k_p of 0.52, 0.54 were obtained from the specimen with 0.25 and 0.5 mole % CeO_2, respectively. Curie temperature was decreased with the addition of CeO_2 while the electric coercive field was proportional to the amount of impurity. Based on the system ceramics with 0.5 mole % cerium oxide, a Rosen type piezoelectric transformer was fabricated and tested. Voltage step-up ratios of 230 and 13 were obtained from the transformer at no load and 100 kΩ resistance, respectively. Experimental results showed a potential of the transformer for the practical use coupled with the expected strength increase by the grain size refinement.

Key Words : Mechanical quality factor(Q_m), Electromechanical coupling coefficient(k_p), Rosen type piezoelectric transformer, Voltage step-up ratios

1. Introduction

Piezoelectric transformers has been studied for the application of high voltage devices such as flyback transformers and DC power voltage sources[1]. Compared to the conventional electromagnetic transformers, piezoelectric transformers have several advantages including high efficiency, small size with low profile and inherent inflammable characteristics. However, before the piezoelectric transformers can be of practical use, major drawbacks, which are low step-up ratio and inadequate strength at higher power level must be addressed.

Recently, Rosen type transformer operated at resonance frequency has been applied to the inverter which drives cold cathode fluorescent lamp (CCFL) of notebook personal computer[2, 3]

In this experiment, we chose two system ceramics, $Pb(Zr,Ti)O_3$ and $Pb(Mn_{1/3}Nb_{2/3})O_3$ which have small amount of CeO_2 impurity, to investigate the variation in piezoelectric properties, mainly in electro - mechanical coupling coefficient and mechanical quality factor with the amount of CeO_2 addition.

The selection of CeO_2 is primarily based on the comparable ionic radii of Ce ions to those of the matrix atoms, which enables forming different types of substitutional solid solution, hence resulting in different piezoelectric properties.

In addition, using 0.5 mole % CeO_2 composition, which provides the optimum piezoelectrc characteristics, a Rosen type piezoelectric transformer was designed, fabricated and tested to examine the possibility of practical use.

2. Experimental

Ceramics with composition of $(Pb_{0.94}Ba_{0.06})(Zr_{0.52}Ti_{0.48})_{0.925}(Mn_{1/3}Nb_{2/3})_{0.075}O_3$ were produced from the reagent grade raw material oxide via conventional mixed -oxide process. The cerium oxide was mixed up to 2.0 mole % to the system ceramic. Raw materials of given composition were weighed and acetone -milled for 24 hours in a zirconia ball mill. Calcination at 850 ℃ for 2 hours in a covered alumina crucible was followed. The material was acetone ground for 15 hours in the mill and dried again. The 5 wt. % PVA solution was mixed up with the material in an alumina crucible and the material was sieved to -#100 mesh after drying. The powders were pressed into discs and sintered at 1260℃ for an hour. For the structure analysis, X-ray

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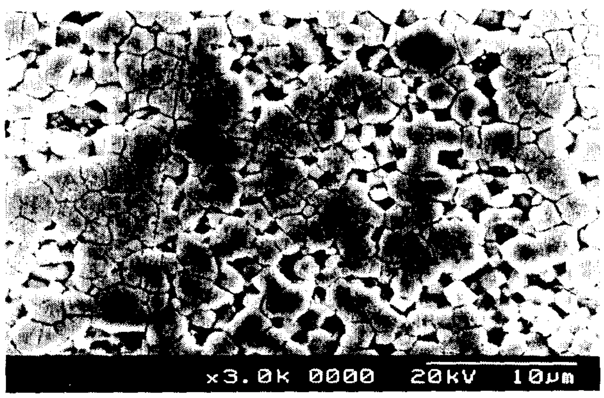
patterns for the sintered specimens were obtained. The microstructure was investigated using scanning electron microscopy and the grain size was determined from the SEM micrographs using linear intercept method. Dielectric properties of the specimens were measured from the capacitance with LCR meter using frequency of 1kHz and the relative dielectric constants were calculated from the values of capacitance. Electrical and piezoelectric properties of the specimens with poling treatment were determined through the resonance method using frequency data obtained from impedance analyzer HP4194A. The coercive field was obtained using sawyer-tower circuit.

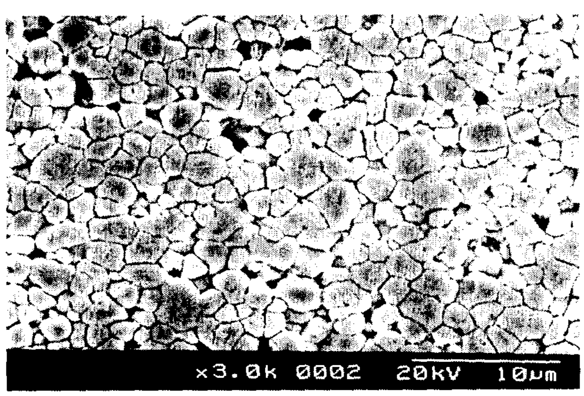
The impedance and admittance circles were determined using the impedance analyzer as well. The poling treatment of 25kv/cm and 15kv/cm, toward thickness and length direction, respectively, was performed to the Rosen type transformer with the size of $1\times8\times42$ mm.

3. Result and Discussion

As illustrated in Fig. 1, the grain size was suppressed as amount of CeO_2 increased.



(a) 0 mole %



(b) 0.5 mole%

Fig. 1 SEM micrographs of specimens with different CeO_2 addition

The micrographs clearly show the inhibition effect of impurities on the grain growth during solid state reaction at high temperature. The grain boundaries can be effectively pinned by the cerium oxide impurities concentrated at the boundary and the final grain size of the material can be determined by the amount of the precipitates at the grain boundaries.

As summarized in the Table1, the average grain size with no impurity and 2.0 mole % CeO_2 addition was 3.12 μm, and 1.74 μm, respectively, which implies that the ceramic with CeO_2 impurities can be effectively strengthened by the grain size hardening mechanism.

The X-ray diffraction patterns of all the composition materials revealed weak tetragonal phase as shown in the table as well.

Table 2 shows the effect of CeO_2 impurities on the Curie temperature and coercive field for the base composition of PMN-PZT. For reference, the variation in hysterisys loop is shown in Fig. 2.

As can be seen, Curie temperature was decreased as the amount of additive increased, which can be correlated with the variation in tetragonality as impurity increases. In contrast, the coercive field was proportional to CeO_2 mole %, indicating the restraining effect of grain size on the domain

Table 1 Grian size, lattice parameter and density of the specimens.

CeO_2 mole%	grain size (μm)	a(Å)	c(Å)	tetrago-nality (c/a)	density (g/cm^3)
0	3.12	4.0653	4.125	1.014	7.21
0.25	2.82	4.0647	4.117	1.013	7.35
0.5	2.57	4.0646	4.112	1.011	7.48
1.0	2.11	4.0824	4.009	1.004	7.47
2.0	1.74	4.0671	4.117	1.012	7.01

Table 2 Piezoelectric properties of PMN-PZT with CeO_2

CeO_2 mole%	E_c(kv/cm)	$\varepsilon_{33}^{t}/\varepsilon_0$	Q_m	$-d_{31}$ (10-12 C/N)	k_p	k_{31}	T_c (℃)
0	9.90	769	1251	77.5	0.55	0.29	329
0.25	9.99	767	1792	74.3	0.52	0.29	325
0.5	10.7	779	1285	82.5	0.54	0.31	322
1.0	10.5	886	1428	78.6	0.48	0.27	323
2.0	10.8	979	1327	77.5	0.47	0.26	319

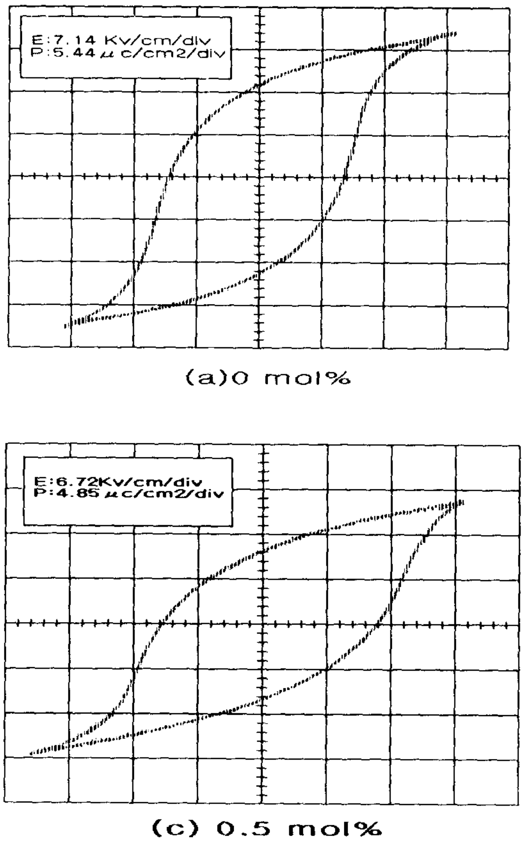


Fig. 2 Variation of hysteresis loop as CeO_2 impurity addition

switching. Table 2 also shows the variation in electromechanical coupling coefficient, k_p and the mechanical quality factor, Q_m. As the speculation in other PZT system ceramics with MnO[4)] impurity, Ce ions can have different valencies of Ce^{3+} and Ce^{4+}. Since the ionic radii of Ce^{3+} and Ce^{4+} are similar to those of Pb^{2+}(1.20Å) and Ba^{2+}(1.34Å), as of 1.07 Å and 0.9Å, respectively, Ce ions are capable of occupying A site in the ABO_3 perovskite lattice regardless of valencies. When the A site in the lattice is replaced with Ce ions, vacancies can be caused, leading to the increase in dielectric constant and electromechanical coupling coefficient. Thus, the increased dielectric constant and electromechanical coupling coefficient at 0.5 mole % CeO_2 addition can be attributed to the substitution of Ce ions to the A site in the perovskite lattice. At the 0.25 mole % CeO_2, however, decrease in dielectric constants, increase in mechanical quality factor and lowered electromechanical coupling coefficient were observed. Such variations in dielectric properties could be speculated on the basis of the behavior of Ce ions in the lattice as well. On the contrary to the case of 0.5 mole % impurity addition, at 0.25 mole % CeO_2, it is believed that only the Ce^{4+} ions with smaller ionic radius are substituted to B site rather than A atom position in the perovskite lattice, resulting in the generation of oxygen vacancies in the structure[5]. The oxygen vacancies directly contribute to the shrinkage of the lattice and hence, dielectric properties are lowered as a result of decrease in dipole moment.

Based on the small grain size, higher electromechanical coupling coefficient and mechanical quality factor at 0.5 mole% CeO_2, as summarized in Table 2, a Rosen type transformer with 0.5 mole % impurity addition was fabricated and tested.

Table 3 shows the dependence of voltage gain and efficiency of the transformer on the load resistance and frequency. The voltage gain was increased and the efficiency of about 80% was obtained as load resistance increased.

Fig 3 shows the equivalent circuit of the piezoelectric transformer. The constants of equivalent circuit were calculated by the admittance circle as shown in Fig. 4[6] As can be seen in Fig. 5, significant increase in voltage step-up ratios of 230 and 13 were obtained from the transformer at no load and 100 kΩ resistance, respectively. Resonant frequencies of the transformer were 81.4 KHz and 80 KHz, at each load.

Table 3 Resonant frequency and maximum voltage gain and efficiency according to load resistance

R [kΩ]	theoretical			measured		
	voltage gain	effi-ciency	resonant frequency	voltage gain	effi-ciency	resonant frequency
50	14.5	87	80.2	11.4	80.3	80.2
100	15.9	91	80.6	13.4	81	80.7
150	20.2	92	80.6	15.5	81	80.9
200	23.4	92	80.8	18.5	77.5	81

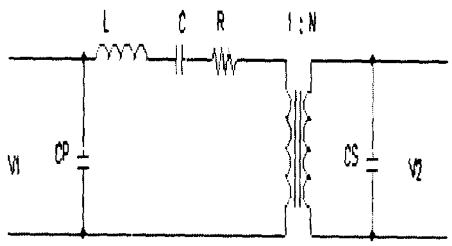
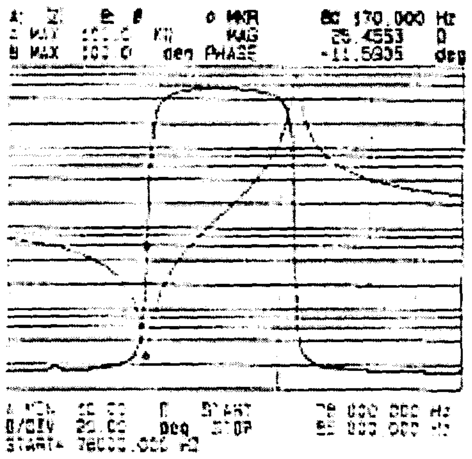
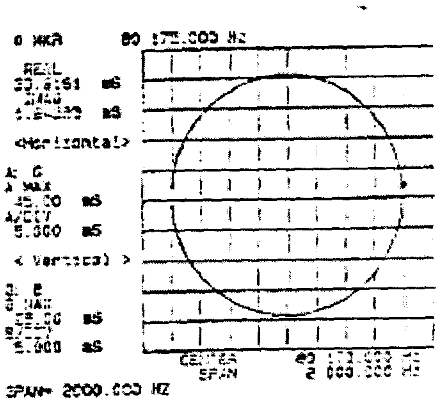


Fig. 3 Lumped constant equivalent circuit of PT(R=25.1, Cp=3.86nF, L=53.3mH, C=74.02pF, Cs=12.65pF)

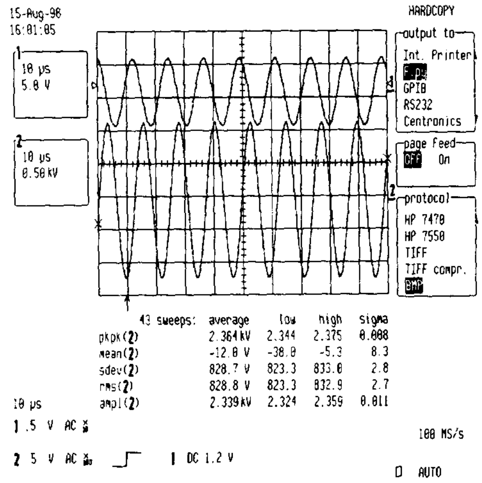


(a) Impedance

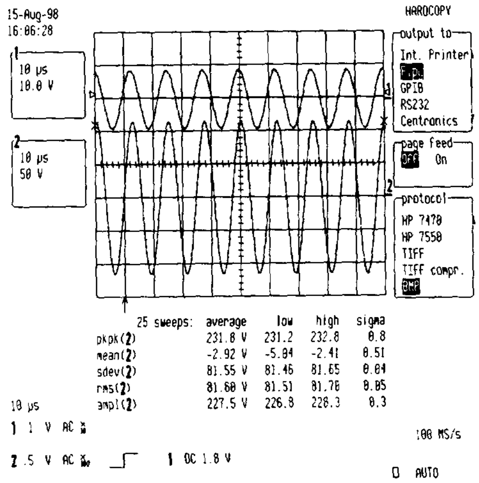


(b) Admittance circle

Fig. 4 Impedance and admittance circle with short circuiting secondary terminal



(a) no load



(b) 100kΩ

Fig. 5 Input and output of PT at no load and 100kΩ resistance(1: input, 2: output)

4. Conclusion

In order to develop the composition ceramics for the piezoelectric transformer, we investigated the dielectric, structural and piezoelectric effects of CeO_2 addition on the base composition of $(Pb_{0.94}Ba_{0.06})(Zr_{0.52}Ti_{0.48})_{0.925}(Mn_{1/3}Nb_{2/3})_{0.075}O_3$.The experimental results obtained from the investigation can be summarized as follows.

The grain size was decreased with the addition of CeO_2. Mechanical quality factor, Q_m of 1792, 1285 and the electromechanical coupling coefficient, k_p of 0.52, 0.54 were obtained from the specimen with 0.25 and 0.5 mole % CeO_2, respectively. Curie temperature was decreased with the addition of impurity while the electric coercive field was increased as the amount of cerium oxide. A Rosen type transformer fabricated in our laboratory, with 0.5 mole % CeO_2 showed voltage step up ratios of 230 and 13 at no load and 100 kΩ resistance, respectively, which indicates a potential of the composition for the practical use.

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