Physical properties of inorganic PMW-PNN-PZT ceramics

Sang-Hoon Sin[1], Ju-hyun Yoo[1,3], Yong-Jin Kim[1], Sam-ki Baek[1], Jun-Soo Ha[1], Chung-Han No[1], Hyun-Seon Song[1] and Dong-Chan Shin[2]

[1]Semyung University, Sinwol-dong Jecheon Chungbuk 390-711, Republic of Korea
[2]Dong-IL Electronics Chungju 380-921, Republic of Korea

E-mail: Juhyun57@semyung.ac.kr

Abstract. In this work, inorganic $Pb(Mg_{1/2}W_{1/2})_{0.03}(Ni_{1/3}Nb_{2/3})_x(Zr_{0.5}Ti_{0.5})_{0.97-x}O_3$ (x= 0.02~ 0.12) composition ceramics were fabricated by the conventional solid state reaction method. And then their microstructure and ferroelectric properties were investigated according to the amount of PNN substitution. Small amounts of Li_2CO_3 and $CaCO_3$ were used in order to decrease the sintering temperature of the ceramics. The 0.10 mol PNN-substituted PMW-PNN-PZT ceramics sintered at 920℃ showed the excellent physical properties of piezoelectric constant (d_{33}), electromechanical coupling factor (k_p), mechanical quality coefficient (Q_m), and dielectric constant of 566 pC/N, 0.61, 73, and 2183, respectively.

1. Introduction

Recently, PZT-based ceramics have been widely used for applications such as transformer, and multilayer actuator [1-4]. Because PZT-based ceramics including $Pb(Ni_{1/3}Nb_{2/3})O_3$-$Pb(Zr,Ti)O_3$ and $Pb(Mg_{1/3}Nb_{2/3})O_3$-$Pb(Zr,Ti)O_3$ show an excellent piezoelectric properties at MPB(Morphotropic Phase Boundary), these three-component perovskite compounds have been actively studied [5]. However, a main problem encountered in manufacturing these ceramics is the volatilization of PbO during sintering process. High temperature sintering of PZT-based ceramics brings about serious problems such as environmental pollution and compositional fluctuations due to the volatilization of the lead oxide. Therefore, it is necessary to improve the physical properties of the ceramics using low temperature sintering. Many researchers have been tried to lower the sintering temperature of PZT-based ceramics without degrading the properties of the materials. In order to lower sintering temperature of the ceramics, sintering aids as additives are widely used [6-8]. Sintering aids used in this experiment were Li_2CO_3 (melting point=732℃) [9] and $CaCO_3$ (melting point=825℃) [10]. In this study, Pb $(Mg_{1/2}W_{1/2})_{0.03}(Ni_{1/3}Nb_{2/3})_x(Zr_{0.5}Ti_{0.5})_{0.97-x}O_3$ were prepared as a base composition . And also, in order to develop piezoelectric ceramics with excellent piezoelectric and dielectric properties, the amount of PNN substitution was changed.

[3] Address for correspondence: Ju-hyun Yoo, Semyung University, Sinwol-dong Jecheon Chungbuk 390-711, Republic of Korea. E-mail: Juhyun57@semyung.ac.kr.

IOP Conf. Series: Materials Science and Engineering **87** (2015) 012099 doi:10.1088/1757-899X/87/1/012099

2. Experimental procedure

The specimens were prepared by a conventional mixed oxide process. The compositions used in this study were as follows.

0.9925 $Pb(Mg_{1/2}W_{1/2})_{0.03}(Ni_{1/3}Nb_{2/3})_x(Zr_{0.5}Ti_{0.5})_{0.97-x}O_3$+0.0075$BiFeO_3$ (x= 0.02, 0.04, 0.06, 0.08, 0.10, 0.12)

The raw materials of PbO, MgO, WO_3, NiO, Nb_2O_5, ZrO_2 and TiO_2 were weighed by mole ratio and mixed together by ZrO_2 balls using the acetone as the medium. After drying, they were calcined at 850℃ for 2h. The calcined powder and $BiFeO_3$ was milled again with the additives, Li_2CO_3 (0.2 wt%) and $CaCO_3$ (0.25 wt%) for 24 h. The powders were pressed into disk of diameter of 14Φ and 2mm in thickness under pressure of 1,000 kg/cm^2. Burned out at 600℃ for 3h, and then sintered at 920℃ for 1 h 30 min. For measuring the piezoelectric properties, after sintering the piezoelectric ceramics were lapped into 1 mm thickness and then metallized with Ag paste at 600℃ for 15 min. Specimens were poled under a dc field of 3.5 kv/mm in silicon oil at 120℃ for 30 min. All electrical measurements were performed about 24h after poling. The crystal structure and microstructure of specimens were analyzed through scanning electron microscopy (SEM) and X-ray diffraction (XRD), respectively. For measuring the dielectric properties, capacitance was measured at 1 kHz using an LCR meter (ANDO AG 4304) and dielectric constant was calculated. For piezoelectric properties, the resonant and anti-resonant frequencies were measured by an Impedance analyzer (Agilent 4294) in conformity to IEEE standard and then the electromechanical coupling factor and mechanical quality factor were calculated. Piezoelectric constants were obtained using d_{33} meter (APC YE2730A).

3. Results and discussion

Figure 1 shows density of specimens as a function of PNN substitution. With increasing PNN substitution, the density was gradually increased. The maximum value of 7.85 g/cm^3 at x=0.12 was appeared. It is considered that the densification of density can be improved by sintering aids.

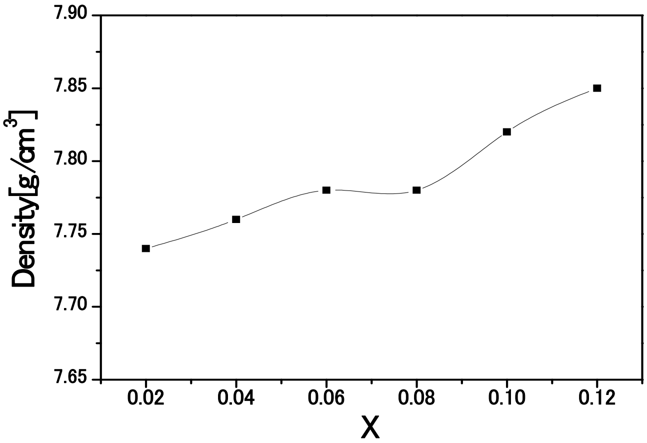

Figure 1. Density of specimens as a function of PNN substitution.

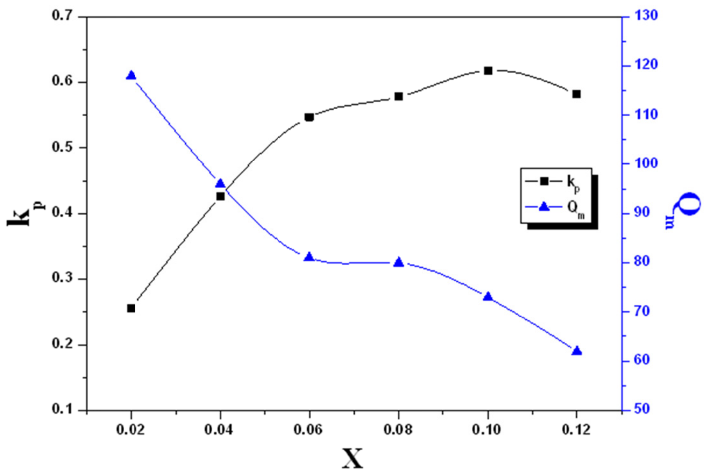

Figure 2. k_p and Q_m with the amount of PNN substitution.

Figure 2 shows electromechanical coupling factor (k_p) and mechanical quality factor (Q_m) with the amount of PNN substitution. k_p increases with the increase of PNN substitution, and reaches the maximum value of 0.618 at x=0.10. Subsequently, k_p was decreased at x=0.12. This phenomenon is due to the excess substitution of PNN. The variation of Q_m showed the opposite properties to piezoelectric coefficient (d_{33}), electromechanical coupling factor (k_p) and dielectric constant (ε_r). When the PNN substitution is x=0.10, the Q_m was reached 73 and then decreased with the increase of PNN substitution. Generally, PZT -based ceramics show the decreased Q_m value at MPB region.

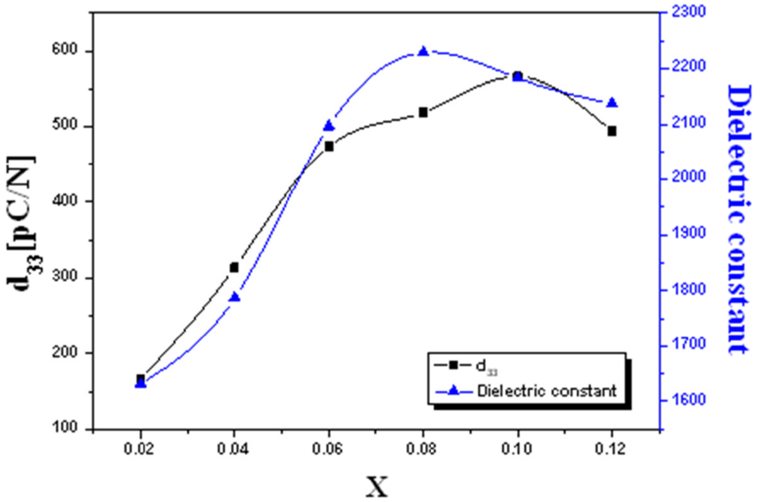

Figure 3. Piezoelectric coefficient (d_{33}) and dielectric constant (ε_r) of specimens as a function of PNN substitution.

Figure 3 shows piezoelectric coefficient (d_{33}) and dielectric constant (ε_r) of specimens as a function of PNN substitution, measured at room temperature. As results in figure 2, piezoelectric coefficient (d_{33}) increases with the increase of PNN substitution. The variation of d_{33} showed the maximum value of 566pC/N at x=0.10 PNN substitution. In addition, these results are because of appearance of MPB at x=0.10 and the feasibility of domain wall movement can be increased owing to increased polarization efficiency at MPB. Dielectric constant (ε_r) of specimens was increased with the increase of PNN substitution, reached the maximum value of 2,229 at x=0.08, and then decreased with the further increase of PNN substitution. This result can be explained from the improvement of sinterability and increase of density.

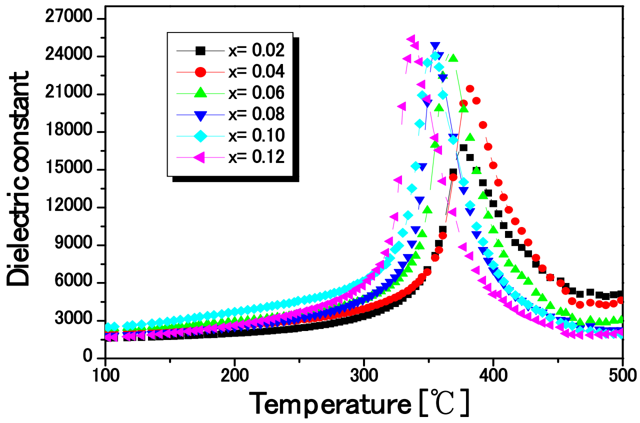

Figure 4. Temperature dependence of dielectric constant as a function of PNN substitution.

Figure 4 shows the temperature dependence of dielectric constant as a function of PNN substitution. Curie temperature (T_c) was decreased with the increase of PNN substitution. It shifted from 377℃ to 337℃. In addition, when the PNN substitution amount was x= 0.10, Curie temperature (T_c) was 355℃.

4. Conclusions

In this study, in order to develop the ceramics with excellent piezoelectric properties, $0.9925[Pb(Mg_{1/2}W_{1/2})+(Ni_{1/3}Nb_{2/3})_x(Zr_{0.5}Ti_{0.5})_{0.97-x}]O_3 + 0.0075\ BiFeO_3$ ceramics were investigated as a function of PNN substitution. The results obtained from the experiment are as follows:

- When x= 0.10, the maximum values of k_p= 0.61, ε_r= 2183, and d_{33}= 566 pC/N were shown.
- As amount of PNN substitution increased, Curie temperature (Tc) shifted to low temperature range. When the x = 0.10, Curie temperature(Tc) was 355℃.

References

[1] Jeong Y H, Yoo K G and Lee J H 2009 *J Electroceram* **23** 387-91
[2] Chao X, Yang Z, Dong M and Zhang Y 2011 *J Magn Magn Mater* **323** 2012-6
[3] Shin T H, Ha J Y, Song H C, Yoon S J, Park H H and Choi J W 2013 *Ceram Int* **39** 1327-33
[4] Ha J Y, Choi J W, Kang C Y, Choi D J, Kim H J and Yoon S J 2005 *Mater Chem Phys* **90** 396-400
[5] Yoon K H, Kim K S and Choi B S 2001 *Journal of the Korean Institute of Electrical and Electronic Material Engineers* **14** 20-5
[6] Yoo J H, Kim K J, Jeong Y H and Lee S H 2007 *Journal of the Korean Institute of Electrical and Electronic Material Engineers* **20** 766-70
[7] Lee M W, Kim S J, Yoon M S, Ryu S L and Kweon S Y 2008 *Journal of the Korean Institute of Electrical and Electronic Material Engineers* **21** 1077-82
[8] Lee K S, Lee L H, Yoo J H and Ryu S L 2007 *Journal of the Korean Institute of Electrical and Electronic Material Engineers* **20** 1034-8
[9] Hu T, Jantunen H, Deleniv A, Leppavuori S and Gevorgian S 2004 *J. Am. Ceram. Soc* **87** 578-83
[10] Licht S, Wu H J, Hettige C, Wang B, Asercion J and Stuart J 2012 *Electronic Supplementary Information (Chem. Comm.)* **1** 1-20