CHAPTER 6

CONCLUSIONS AND FUTURE WORK

This chapter, the overall conclusions of this study are drawn and the future extensions of this study are proposed.

6.1. Conclusions

6.1.1. PZN-PZT based compositions

6.1.1.1. Effect of PZN content on PZN-PZT based ceramics

Ceramics in the $x\mathrm{Pb(Zn_{1/3}Nb_{2/3})O_3}$–$(1\text{-}x)\mathrm{Pb(Zr_{1/2}Ti_{1/2})O_3}$ [xPZN–(1-x)PZT] solid solution system are expected to display excellent dielectric, piezoelectric, and ferroelectric properties in compositions close to the morphotropic phase boundary (MPB). The electrical behavior of the ceramics with x = 0.1 – 0.5 has been characterized in order to identify the MPB compositions in this system. With combination of x-ray diffraction analysis, dielectric, piezoelectric and ferroelectric measurements, it was consistently shown that an MPB between tetragonal and rhombohedral phase exists around x = 0.2-0.3 in this binary system. In addition, the transition temperature decreased with increasing PZN content in the system. The variation of piezoelectric and dielectric properties showed similar increasing trend to density. The *P–E* and *s-E* loops demonstrated that PZN–PZT system changed gradually from tetragonal phase to the rhombohedral phase. These results clearly showed the significance of PZN in controlling the electrical responses of the PZN–PZT system.

6.1.1.2. Effect of Zr/Ti ratio on PZN-PZT based ceramics

Ceramic composition $0.2Pb(Zn_{1/3}Nb_{2/3})O_3$-$0.8Pb(Zr_xTi_{1-x})O_3$, [0.2PZN–0.8PZT] solid solution system is expected to display excellent dielectric, piezoelectric, and ferroelectric properties as it is close to the morphotropic phase boundary (MPB) of PZN-PZT system. The electrical behavior of the ceramics with x= 0.40, 0.45, 0.50, 0.52, 0.55 and 0.60 has been characterized in order to identify the MPB compositions in this system. With combination of x-ray diffraction analysis, dielectric, piezoelectric and ferroelectric measurements, it was consistently shown that an MPB between tetragonal and rhombohedral phase exists around x = 0.50 -0.52 in this binary system. In addition, the transition temperature decreases with increasing Zr content in the system. The *P–E* and *s-E* loops demonstrate that PZN–PZT system changes gradually from tetragonal to rhombohedral phase with changing Zr/Ti ratio. These results clearly show the significance of Zr/Ti ratio in optimizing the electrical properties of the PZN–PZT system.

6.1.2. Modified PZN-PZT compositions

6.1.2.1. Effect of MnO_2 addition on properties of PZN-PZT ceramics

In this section, the dielectric, piezoelectric and ferroelectric properties of MnO_2-doped $0.2Pb(Zn_{1/3}Nb_{2/3})O_3$-$0.8Pb(Zr_{1/2}Ti_{1/2})O_3$ ceramics, which is the morphotropic phase boundary composition of the PZN-PZT system, were investigated. Crystal structure changed to rhombohedral side when increasing MnO_2 content. With the addition of MnO_2, Curie temperature T_c, the piezoelectric constant

d_{33} and electromechanical coupling factor k_p were slightly decreased, but the mechanical quality factor Q_m was significantly increased. The *P–E* and *s-E* loops demonstrated decreased P_r and strain level but increased E_c with increasing amount of MnO_2. These results clearly show the significance of MnO_2 addition in the controlling electrical properties of the PZN–PZT system to exhibited ‘‘hard’’ characteristics.

6.1.2.2. Effect of Fe_2O_3 addition on properties PZN-PZT ceramics

In this section, the dielectric, piezoelectric and ferroelectric properties of Fe_2O_3-doped $0.2Pb(Zn_{1/3}Nb_{2/3})O_3$-$0.8Pb(Zr_{1/2}Ti_{1/2})O_3$ ceramics, which is the morphotropic phase boundary composition of the PZN-PZT system, were investigated. Crystal structure changes to tetragonal side with increasing Fe_2O_3 content. With the addition of Fe_2O_3, Curie temperature T_c, the piezoelectric constant d_{33} and electromechanical coupling factor k_p were slightly decreased, but the mechanical quality factor Q_m was significantly increased. The *P–E* and *s-E* loops show that decreased P_r and strain level but increased E_c when addition amount of Fe_2O_3. These results clearly showed the significance of Fe_2O_3 addition in the electrical properties of the PZN–PZT system with ‘‘hard’’ characteristics.

6.1.2.3. Comparison between Fe_2O_3 and MnO_2 addition on properties of PZN-PZT ceramics

In this study, the dielectric piezoelectric and ferroelectric properties of Fe_2O_3 and MnO_2-doped $0.2Pb(Zn_{1/3}Nb_{2/3})O_3$-$0.8Pb(Zr_{1/2}Ti_{1/2})O_3$ were investigated. In case of addition of Fe_2O_3, the crystal structure was shifted to tetragonal phase. On the other hand, addition of MnO_2 shifted the crystal structure to rhombohedral. With addition of Fe_2O_3 or MnO_2, Curie temperature T_c, the piezoelectric constant d_{33} and electromechanical coupling factor k_p were slightly decreased, but the mechanical quality factor Q_m was significantly increased. Polarization and strain characteristics were shown with increasing E_c, decreasing of P_r, P_s and strain indicating hardening effect with Fe_2O_3 and MnO_2 addition.

6.2. Future work

This study is developing $Pb(Zr_{0.50}Ti_{0.50})O_3$-$Pb(Zn_{1/3}Nb_{2/3})O_3$ based ceramics with improved electromechanical coupling factors and reasonable mechanical quality factor Q_m. These compositions are known as “soft” materials originally with large electromechanical coupling factors and piezoelectric constants.

For a piezoelectric material suitable for AC-driven application such as piezoelectric transformer and ultrasonic motor, it is essential to satisfy both the requirements for high power actuator and receiver. This implies that the material should have a large mechanical coupling factor (*k*) and a piezoelectric constant (*d*), as well as a high mechanical quality factor (Q_m). Further, a high dielectric constant is required in order to draw large power with low dielectric loss in order to suppress heat generation. Consequently, designing a high power composition involves generation of ‘‘hard’’ and ‘‘soft’’ properties at the same time. Previous investigations have revealed that high soft properties are obtained around the morphotropic phase boundary (MPB) compositions. Modification of the MPB with acceptorions in the form of oxide or carbonates or as a relaxor-based compound such as

$Pb(Mn_{1/3}Nb_{2/3})O_3$ and $Pb(Ni_{1/3}Nb_{2/3})O_3$ yields a combinatory material with hard and soft characteristics.

Finally, most high power devices have a multilayer ceramic component to achieve specific characteristics. Multilayering of the piezoelectric composition requires good sinterability and compatibility with an electrode material. Since the sintering temperature of a PZT-based high-power composition is high, approximately 1200 ^oC, a Ag/Pd alloy is commonly used as the electrode because a pure Ag metal diffuses easily into a piezoelectric ceramic at high temperatures. However, a Pd metal is expensive. Consequently, a reduction in the sintering temperature of piezoelectric ceramics is required for the fabrication of cost-effective multilayer piezoelectric devices. Low-temperature sintering offers the advantage of using noble metal electrodes instead of expensive Ag–Pd alloys and reduces the burden on the electric consumption and furnace lining. Methods for reducing the internal electrode cost are classified into use of silver (Ag)—Pd alloy electrodes having a high Ag content (more than 70%) to achieve low temperature sintering of the dielectrics and use of base metals such as nickel (Ni) and copper (Cu) as internal electrodes by using a nonreducible dielectric that can be fired in a reducing atmosphere

Therefore, in future work it is of interest to develop PZN-PZT based ceramics by

1. addition of relaxor ferroelectric which have high *k*, *d* properties suitable for application at high driving condition.
2. developing compositions based on low temperature sinterable to achieve specific characteristics.