Using of electrical properties of modified-PZT ceramics materials to design water transfer tubes that help purify water and preserve the environment

(1) loubna ben amor *Departement Chimie, FMSM, Université Kasdi Merbah-Ouargla*
(2) omar bentouila *Departement Chimie, FMSM, Université Kasdi Merbah-Ouargla*
(3) amar khelassi *Departemen Geni civil et hydrolyque, FAS, Université Kasdi Merbah-Ouargla*
(4) kribaa oum keltoum *Departement Chimie FSSNL, Université* Mohammed Khider *-Biskra*
(5) Boutarfaia Ahmed *Departement Chimie, FSSNL, Université* Mohammed Khider *-Biskra*
E-Mails: loubnabenamor@yahoo.fr

Abstract— After many years of use of metal water transfer pipelines the revolution turns to the design of canals by renovated materials that have experienced a boom in recent years. The materials in contact with drinking water suffers from the failure and the consequences associated with the deterioration of the networks by the corrosion which marks a very important environmental stake. In this study proposing the use of doped PZT type materials, they are resistant to salts, these materials are better exploited when combined with other materials, in order to improve the lifespan of the channel. 0.05 $Pb[Fe_{1/2}\ Nb_{1/2}]O_3$-0.05 $Pb[Ni_{1/3}\ Nb_{2/3}]O_3$-0.90 $Pb[Zr_xTi_{(1-x)}]O_3$ [PFN-PNN-PZT] quaternary piezoelectric ceramics with varying Zr/Ti ratios located near the morphotropic phase boundary (MPB) were prepared by a conventional mixed-oxide route. The effect of Zr/Ti ratio and sintering temperature on the dielectric properties of our simples were investigated. The new MPB in this quaternary system with optimum piezoelectric properties was found at x = 0.51-0.53. The dependence of the dielectric constant (ε) and the loss tangent on the Zr/Ti ratio shows a pronounced maximum of at Zr/Ti : 51/49 and a minimum tanδ . As the Zr/Ti ratio increases, the *Tc* of PFN-PNN-PZT ceramics decreases and consequently the peak in the dielectric spectrum corresponding to the *Tc* moves towards room temperature. A *Tc* of 360 °C is obtained when Zr/Ti: 51/49.

Key-Words— PFN-PNN-PZT; Sintering temperature; Morphotropic phase boundary; Piezoelectric properties; Dielectric properties.

INTRODUCTION

The complex compound, Pb(ZrTi)O3 (referred as to PZT), is a solid solution of ferroelectric PbTiO3 (*TC* 490 ^{0}C) and antiferroelectric PbZrO3 (*TC* = 230 °C) in different Zr/Ti ratios [1], are known as an important piezoelectric materials [2].

PZT ceramics have been used in several technological applications such as ultrasonic sensors, high energy capacitors, piezoelectric actuators, and photoelectric devices [3,4]order to enhance its piezoelectric properties, both of the effects of different processing conditions [5,6]and substitutions of various dopants [7,8]of different ionic sizes and valences have been studied. Doping with different element changes the physical and chemical properties of PZT ceramics. Due the broad range of possible isomorphism in the perovskite structure of ABO_3, PZT ceramics can be accept dopants with different valences into both A-site (Pb-site) and B-site (Zr/Ti –site) of the lattice [9]. Based on aliovalent substituents in the compound, dopants can be classified into two types: donors (higher valence ions) and acceptors (lower valence ions.

Doping PZT with Nb^{5+} increases the electric permittivity and piezoelectric coefficients [10], but

PZT doped with iron presents lower dielectric constant and loss constant [11].

Furthermore, the morphotropic phase boundary (MPB) [12] is an essential parameter to be considered because in this region, tetragonal and rhombohedral phases coexist, and consequently the properties of PZT are improved (dielectric and piezoelectric properties).

The aim of the present work is to study the dielectric properties for PFN-PNN-PZT quaternary ceramics. An effort has been made to determine the MPB phase contents with variations in the Zr/Ti ratio**.** The effects of Zr/Ti ratio on the properties of sintered PFN-PNN-PZT quaternary system ceramics were investigated systematically.

Experimental

Samples with general formula : 0.05 $Pb[Fe_{1/2} Nb_{1/2}]O_3$-0.05 $Pb[Ni_{1/3} Nb_{2/3}]O_3$-0.90 $Pb[Zr_xTi_{(1-x)}]O_3$ ($0.48 \leq x \leq 0.55$) were synthesized from starting materials Pb_3O_4 (99.90%), ZrO_2 (99.90%), TiO_2 (99.90%), Fe_2O_3 (98%), NiO (99.90%) and Nb_2O_5 (99.6%). Each mixture of the starting powders was mixed in a centrifugal mill with absolute alcohol using an agate ball for 3h. The powders were then calcined at 800 °C for 2h at heating and cooling rates of 2°C.min^{-1}. The powders were molded by the pressure of 150 MPa in 12 mm in diameter and about 1 mm in thickness. The pressed disks were covered with alumina crucible and then sintered at 1180 °C for 2 h. To limit PbO loss from the pellets, a PbO-rich atmosphere was maintained by placing a $PbZrO_3$ inside the crucible.

The bulk densities of sintered ceramics were determined by the Archimedes method in water.

For measuring the piezoelectric characteristics, the specimens were polished to 2 mm thickness and then electrodeposited with Ag paste. The pellets are carried out at 110 °C in a silicone oil bath by applying fields of 2.5kV cm^{-1} for 45 min. All samples were aged for 24 h prior to measuring the piezoelectric and dielectric properties.

The dielectric properties (from a room temperature to 450°C) of the poled ceramics were investigated using an automatic (LCR) meter at 1 kHz.

Results and discussion

Morphological characterization of the PFN-PNN-PZT

Figure 1 and Figure 2 shows the variation of bulk density and porosity with the compositions at variation sintering temperature (1100, 1150, and 1180° C). The porosity decreases when the sintering temperature increases. Meanwhile, the density of specimens increased, and shows the maximum value of 7.4 at 51/49 with 1180° C, so the optimum temperature of sintering is 1180 °C. The quality of the material increases with increasing density and it increases with increasing the sintering temperature [13].

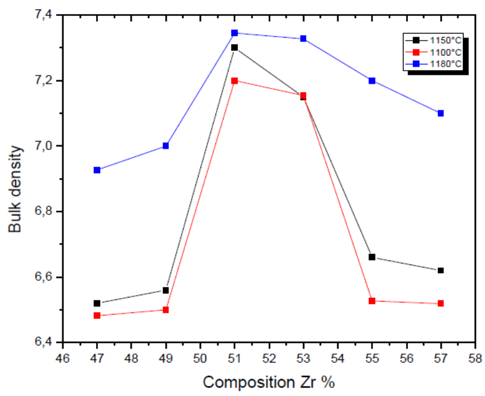

Fig1: *Change in bulk density as functions of Zr in the composition and sintering temperature*

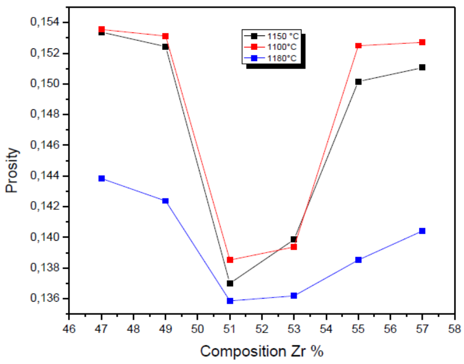

Fig2 :*Change in porosity as functions of Zr in the composition and sintering temperature*

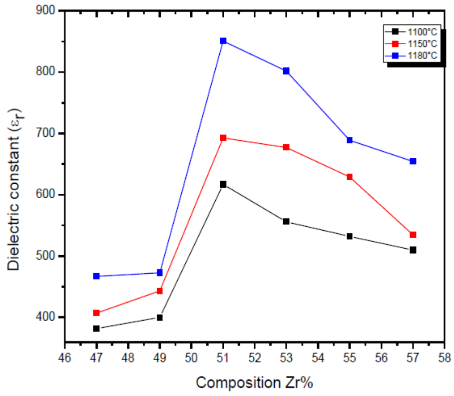

Fig3 : *Dielectric Constant (ε) as a function of the composition (Zr%)*

Dielectric characterization of the PFN-PNN-PZT

Figure 3 and Figure 4 shows the temperature and composition dependence of the dielectric constant of PFN-PNN-PZT ceramics with different sintering temperatures (1100, 1150 and 1180 °C) at 1 kHz. For the three temperatures of sintering 1100, 1150 and 1180 °C, we can observed that the permittivity increases gradually with the increase in the composition and takes a maximum for the sample with Zr/Ti = 51/49 included in the morphotropic phase boundary (MPB) at the temperature 1180 °C and then decreases. Consequently the peak in the dielectric spectrum corresponding to the Tc moves towards room temperature, as shown in Figure 5. The highest dielectric constant peak is achieved in the sample with Zr = 51, which gives a Tc of 360°C.

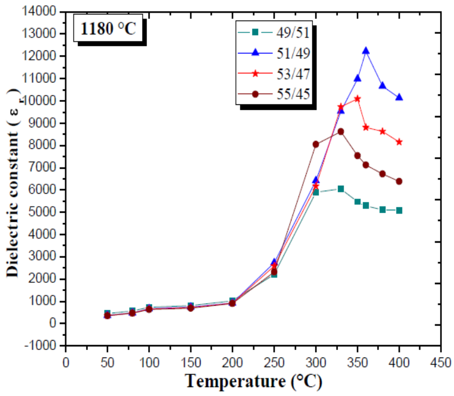

Fig4 : *Dielectric Constant (ε) as a function of the temperature.*

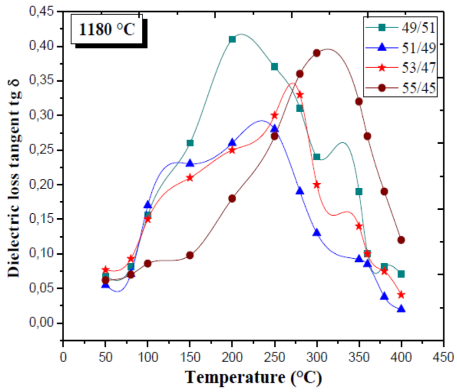

Fig6 : *Dielectric loss tangent (tg δ) as a function of the temperature.*

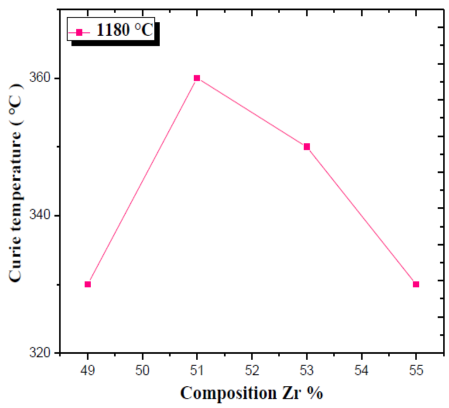

Fig5 : *Variation of Curie temperature as functions of composion (Zr%) sintered at 1180°C.*

Figure 6 shows temperature dependence of the dielectric loss tangent for all specimen. All the specimen shows a similar trend, a minimum value of tan δ obtained when Zr=51, in contrast to the dielectric Constant.

Conclusion

In this study, 0.05 $Pb[Fe_{1/2}Nb_{1/2}]O_3$-0.05 $Pb[Ni_{1/3}Nb_{2/3}]O_3$-0.90 $Pb[Zr_xTi_{(1-x)}]O_3$ ceramics ($0.48 \leq x \leq 0.55$) were successfully prepared using a solid-state mixed oxide technique at different sintering temperature. The ratio of Zr/Ti and the sintering temperature strongly affects the electrical properties of PFN-PNN-PZT ceramics. A transition from tetragonal to rhombohedral phase and density were observed as Zr/Ti ratio increased, and greatly important improvement in electrical properties when sintering temperature and ratio of Zr/Ti increase. The MPB region ($0.51\leq x \leq 0.53$). The dielectric properties (ε =13108, tan δ= 0.012) were observed for the composition sintered at 1180 °C that contains Zr/Ti ratio of 51/49, which could be suitable for high-power applications.

References

[1] K. Uchino, Adv. Mater. Res. 55–57 (2008) 1.

[2]Sriram. S.; Bhaskaran. M.; Mitchell A **2010**. Ed. Ernesto Suaste Gómez.

[3] Izyumskaya, N., Y.-I. Alivov, S.-J. Cho, H. Morkoç, H. Lee and Y.-S. Kang **2007** Processing, Structure, Properties, and Applications of PZT Thin

Films. Critical Reviews in Solid State and Materials Sciences. 32: 111-202.
[4]Chauhan, A., S. Patel, R. Vaish, C.R. Bowen **2015** Anti-Ferroelectric Ceramics for High Energy Density Capacitors. Materials. 8:8009–8031.
[5]Praveen, J. P., T. Karthik, A.R. James, E. Chandrakala, S. Asthana, D. Das **2015**Effect of poling process on piezoelectric properties of sol–gel derived BZT–BCT ceramics. J Eur Ceram Soc. 35(6):1785-1798.
[6]Wongmaneerung, R, R. Tipakontitikul, P. Jantaratana, A. Bootchanont, J. Jutimoosik, R. Yimnirun, S. Ananta **2016** Structure and phase formation behavior and dielectric and magnetic properties of lead iron tantalatelead zirconate titanate multiferroic ceramics. Materials Research Bulletin. 75:91-99.
[7]Fu, S.-L., S.-Y. Cheng, C.-C. Wei **1986** Effects of doping pairs on the preparation and dielectricity of PLZT ceramics. Ferroelectrics. 67: 93-102.
[8]Tadon, R. P., V. Singh **1994** Properties of low temperature sintered neodymium doped lead zirconate titanate ceramics, J. Mat. Sci. Lett., 13:810-812.
[9]Nguyen, M. D., T. Q. Trinh, M. Dekkers, E. P. Houwman, H. N. Vu, G. Rijnders **2014** Effect of dopants on ferroelectric and piezoelectric properties of lead zirconate titanate thin films on Si substrates. Ceramics International. 40:1013–1018.
[10]Bornand, V., D. Granier, P. Papet, E. Philippot **2001** Elaboration and characterization of Nb-doped PZT ceramics. Annales de Chimie Science des Materiaux. 26: 135-139.
[11]Raí, R., S. Sharma, R. N. P. Choudhary **2005** Dielectric and piezoelectric studies of Fe doped PLZT ceramics, Materials Letters .59 (29-30) 3921-3925.
[12]Wilkinson. A. P. ; Pattanaik. J. Xu.; Billinge. S, S. J. L.; **1998**. Chem. Mat. *10,* 3611–3619
[13] Boutarfaia, A. **2000** Investigation of Co-Existence Region in Lead Zirconate-Titanate Solid Solutions: X-Ray Diffraction Studies. Ceramic International. 26(6): 583-587.