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Effect of Nd_2O_3 adding on electrical properties of bismuth sodium titanate ceramics

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ABSTRACT

This research, fabrication and effect of neodymium oxide (Nd_2O_3) adding on electrical properties of bismuth sodium titanate (BNT) ceramics were studied. The sample powder, BNT/x Nd_2O_3 (x = 4, 8 and 12 wt %) was prepared via mixed oxide method from BNT and Nd_2O_3 starting powders. The starting powder were mixed by using zirconia grinding media in ethanol for 24 h and dried by an oven. The mixture powder was calcined at temperature of 800 °C and was then pressed and sintered at the temperature of 1075 °C. Phase formation of samples was examined by XRD technique. Moreover, the physical and electrical properties and microstructure of samples were determined. From the results, it was found that electrical properties of BNT ceramics can be improved by adding Nd_2O_3. Furthermore, grain size of BNT ceramics are decreasing with increasing of Nd_2O_3 quantity.

Keywords: BNT, Nd_2O_3, Electrical Properties.

INTRODUCTION

In the present day, bismuth sodium titanate, $Bi_{0.5}Na_{0.5}TiO_3$ (BNT) is widely used in many applications. Since BNT ceramics are strongly ferroelectrics and exhibit the outstanding advantages in a free control atmosphere and produce no lead pollution (Lam *et al*., 2005). Because health and environmental concerns with lead-containing materials, BNT is considered to be excellent candidate for use as lead-free piezoelectric ceramics to take place lead-containing piezoelectric ceramics (Lam *et al*., 2005; Nakamura and Nomura, 1966; Takanaka, 2001). Their crystal structures are the perovskite type with rhombohedral symmetry at room temperature. They show a strong ferroelectric property with a relative high Curie temperature of 320 °C. However, the application of pure BNT ceramics are limited by some its shortcomimgs in electric properties, such as low relative dielectric permittivity (ε_r), narrow sintering temperature range and high conductivity at room temperature (Takanaka, 2001). Therefore, it is necessary to improve BNT ceramics on dielectric properties for applications. From the previous works found that the rare-earth additives have effect to the piezoelectric and dielectric properties of PZT ceramics (Garg and Argrawal, 2001; Shannigrahi *et al*., 1999; Garg and Goel, 1999). Thereafter, it is very interesting to study the effect of adding Nd_2O_3 into non-lead based materials. In this work, the authors have studied the effects of rare earth additive (Nd_2O_3) on physical and electric properties of bismuth sodium titanate ceramics. Various weight percent of Nd_2O_3 were used for doping materials and adding to BNT ceramics *(like our previous work which adding Nd_2O_3 into PZT ceramics (Thamjaree, et al., 2007)*. The phase formation, physical properties, dielectric and piezoelectric properties were also studied. Moreover, the SEM micrograph was employ for microstructure determination.

METHODOLOGY

$(Bi_{0.5}Na_{0.5})TiO_3$ powder is prepared by a conventional one-step mixed oxide method. Commercially metal oxide and carbonate powder of high purity, for example Bi_2O_3 (98%), Na_2CO_3 (99.9%), TiO_2 (99%) and Nd_2O_3 (99%) were used as raw materials. The Nd_2O_3 was added to the mixture in various quantities (4, 8 and 12 percent by weight). Firstly, the precursors and dopants were weighed, mixed and milled by wet-milling in ethanol (Merck) using zirconia grinding media for 24 h. The dried powder was crushed and calcined in a closed alumina (Al_2O_3) crucible at temperatures of 800 °C for 2 h with a heating and cooling rate of 3 °C/min. The calcined powder was grinded in an agate mortar, pestle and sieved using a 100-mesh nylon sieve. The sieved powder was pressed to a cylindrical shape with diameter of 15 mm and thickness about 2 mm Polyvinyl alcohol (PVA, Fluka) 2 wt% was used as binder and added to the sieved powder. The green bodies were then sintered in a closed alumina crucible at temperature of 1075 °C for 2 h with heating/cooling rate of 5 °C/min. The sample was firstly heated at 5 °C /min to 500 °C for 1 h to burn out the binder. Then, the pellets were polished and electroded using silver paste (Acheson Electrodag 1415M) for dielectric and piezoelectric property measurements. Moreover, microstructures of the ceramic samples were examined by SEM technique.

RESULTS AND DISCUSSION

Figure 1 shows the typical of X-ray diffractogram of BNT powder which was calcined at temperature of 800 °C for 2 h. It can be seen that the perovskite-like BNT materials with orthorhombic structure were formed with the extra peak which mostly matched with neodymium oxide (Nd_2O_3). The series of peaks are matched with JCPDS no. 36-0340 (Powder Diffraction File, 2000). A weak reflection peak of unreact Nd_2O_3 phase increased with higher dopant quantities.

Table 1 shows physical and electrical properties of BNT samples. From the results, it could be found that the quantity of Nd_2O_3 have effects to physical and electrical properties of BNT ceramics. High density (93-97%) and shrinkage (~45-48%) of samples are increased with quantity of dopant. Furthermore, the particle and grain size of BNT powder and ceramics decrease with increasing of Nd_2O_3 quantity which is in the range of 0.28-0.24 μm and 1.25-0.43 μm, respectively. Morever, dielectric constant (670-830) and piezoelectric property (36-75 pC/N) of samples are improved by adding Nd_2O_3. It may be the effect of inhibiting grain growth of Nd_2O_3 on grain growth processing which effect to high density and high piezoelectric property. Besides, it can be observed that tendency of d_{33} value of 4 wt% and 8 wt% is not consistent with density. It may be due to the inhomogeneous mixing of Nd_2O_3 in BNT ceramics. However, adding Nd_2O_3 can promote the piezoelectric and dielectric properties of BNT ceramics which can develop to capacitor like PZT materials.

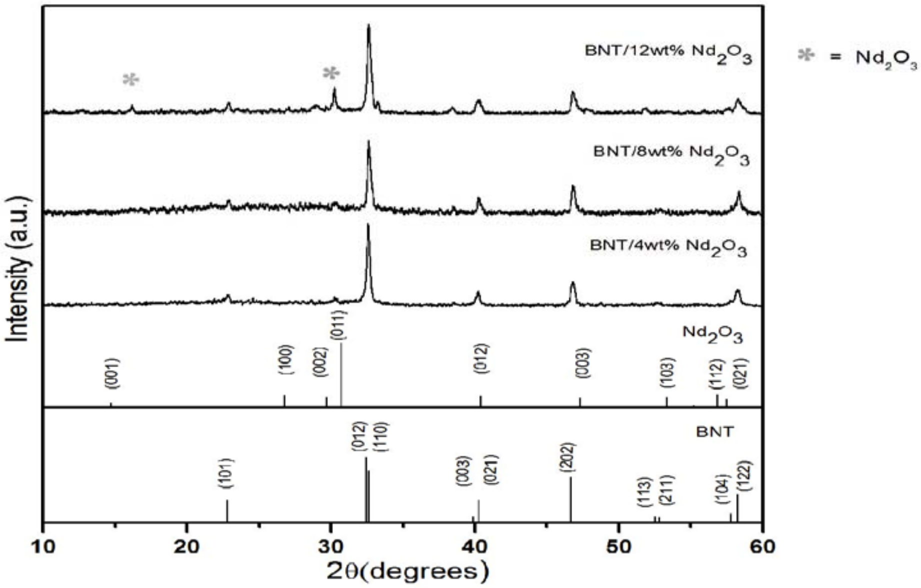

Figure 1 X-ray diffractogram of BNT powder doped with different quantities of Nd_2O_3 and calcined at temperature of 800 °C for 2 h.

Table 1 shows the physical and electrical properties of BNT samples.

Quantity of Nd_2O_3 (wt%)	Density (%)	Shrinkage (%)	Particle size (μm)	Grain size (μm)	ε_r	d_{33} (pC/N)
0 wt%	93.18	45.13	0.28	1.25	670	36
4 wt%	94.95	46.23	0.26	0.85	742	65
8 wt%	96.52	47.14	0.25	0.63	800	48
12 wt%	97.05	47.89	0.24	0.43	830	75

Figure 2 and Figure 3 show the SEM micrographs of BNT powder and ceramics, respectively. Figure 2 shows the morphology of BNT powder doped 4-12wt% Nd_2O_3. The agglomeration of irregular shape of BNT powder can be observed. Figure 3 shows SEM micrographs of BNT ceramics sintered at temperature of 1075 °C for 2 h. It can be seen that grain size of BNT ceramics decrease with increasing of Nd_2O_3 which corresponded with density of samples. The cube shape could be observed in all samples. The EDS was employed to determine the ratio of element in the sample which as shown in Figure 4. From the spectrum, it can be seen that all elements observed could be matched with the use starting powders.

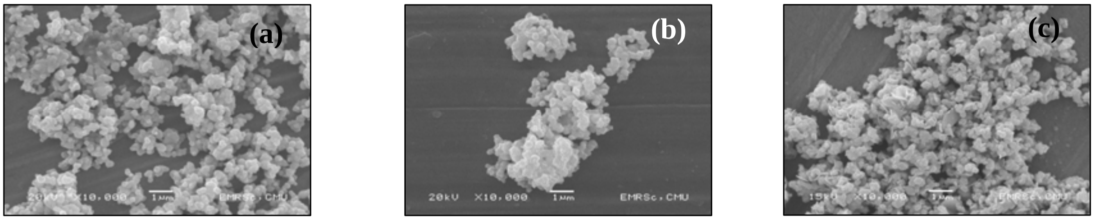

Figure 2 SEM micrographs of BNT powders (a) 4 wt% (b) 8 wt% and (c) 12 wt% of Nd_2O_3.

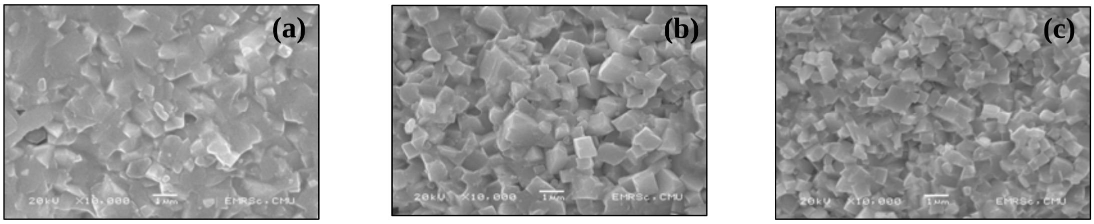

Figure 3 SEM micrographs of BNT ceramics (a) 4 wt% (b) 8 wt% and (c) 12 wt% of Nd_2O_3

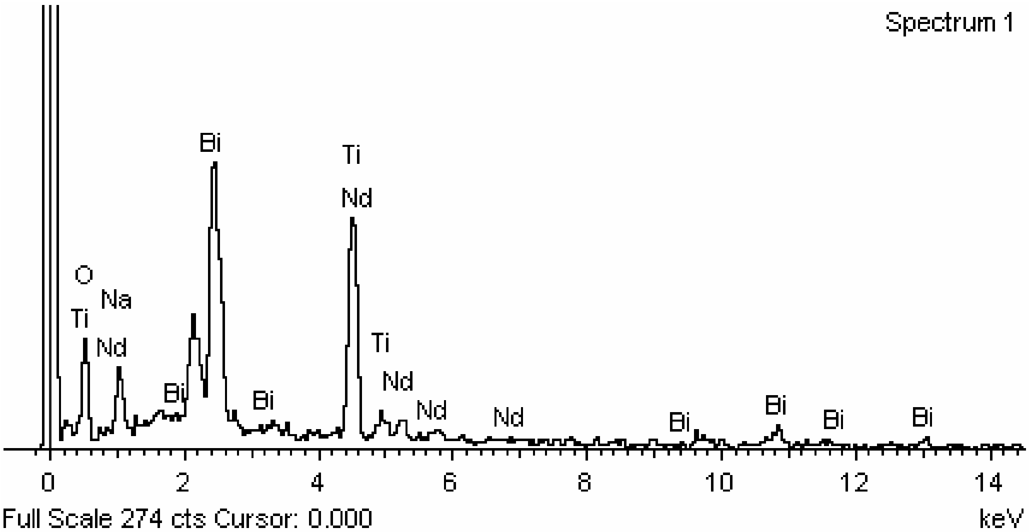

Figure 4 EDS spectrum of BNT ceramics.

CONCLUSION

The effect of adding various quantities of Nd_2O_3 on physical and electric properties of BNT ceramics were studied. From the result, it can be concluded that the density increased with the increasing of wt% of Nd_2O_3. The dielectric and piezoelectric properties of BNT ceramics are also improved with increasing of wt% of Nd_2O_3. The SEM micrographs show the grain growth with grain size in the range of 1.25-0.43 μm.

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