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Microstructural and high temperature dielectric, ferroelectric and complex impedance spectroscopic properties of $BiFeO_3$ modified NBT-BT lead free ferroelectric ceramics

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ABSTRACT

Solid solutions of nano structured $Na_{0.5}Bi_{0.5}TiO_3$-$Ba_{0.925}Nd_{0.05}TiO_3$-$BiFeO_3$ (0.7NBT-0.2BT1-0.1BF & 0.7NBT-0.1BT1-0.2BF) compositions were prepared by the conventional sol-gel method and the effect of $BiFeO_3$ addition on microstructure, dielectric, ferroelectric and high temperature electrical properties of NBT-BT1-BF ceramics were investigated through XRD, SEM, dielectric and electrical characterization. X-ray diffraction patterns are well indexed and found that samples are crystallized in rhombohedral phase. SEM images have shown uniform distribution of grains and change in grain size with $BiFeO_3$ concentration. The samples exhibited relaxor behavior, which is accompanied by a shift in ε_{max} and Z" towards high temperature with increasing $BiFeO_3$ concentration. The grain and grain boundary response as well as the relaxation processes at different frequencies and temperatures were discussed. These observations suggest that $BiFeO_3$ addition to NBT-BT1 ceramics can be considered as a potential lead free ceramic system in multi-ferroic devices.

1. Introduction

In recent years, development of lead free ferroelectric ceramics has received significant attention from the view point of environmental protection. Sodium bismuth titanate (NBT) is considered as a promising candidate due to superior ferroelectric, piezoelectric and dielectric properties [1–4]. NBT based compounds such as NBT-BT, NBT-ST, NBT-BF, NBT-PZ, NBT-BS and NBT-KNN have been developed rapidly owing to their large piezoelectric properties [5–9]. Doping and the formation of solid solutions such as NBT-BT-KNN, NBT-BT-ST, NBT-BT-KBT, BNT-BT-$AgNbO_3$ were identified as effective ways to further enhance the ferroelectric and piezoelectric properties of NBT-BT based ceramic compositions [10,11].

Solid state synthesized powder methodology is the most commonly used technique to process the NBT-BT based solid solutions. However, this method leads to non-stoichiometry in the composition and introduces harmful impurities. It has been observed that trivalent dopants not only lowers the coercive field of NBT, but also enhances the piezoelectric properties. A previous study has shown that NBT and BF form a solid solution at room temperature and these two compounds exhibit ferroelectric behavior which is maintained within all solid solutions. Moreover, the coexistence of magnetization and electric polarization might allow an additional degree of freedom in the design of novel devices such as actuator, transducers and storage devices and also to construct multifunctional devices [12,13].

$BiFeO_3$ modified NBT based compositions are found to show better piezoelectric properties and ease of poling as compared with the pure NBT ceramics [14]. On one hand $BiFeO_3$, is anti-ferromagnetic until its Neel temperature at about T_N = 370 °C, while NBT do not present any magnetic properties so the solid solution may show interesting multiferroic behavior. Inspite of processing, excellent ferroelectric and piezoelectric properties, there are no reports about the effect of grain size on the ferroelectric properties and high temperature electrical properties, which are important for practical considerations. These results motivated the authors for investigation on $BiFeO_3$ doped NBT-BT1 system of ceramics.

Hence in the present study, to explore the new device heterostructures, which have potential applications as high density information storage, micro-sensors and micro-electromechanical systems, a new systems of $Na_{0.5}Bi_{0.5}TiO_3$-$Ba_{0.925}Nd_{0.05}TiO_3$-$BiFeO_3$ (0.7NBT-0.2BT1-0.1BF & 0.7NBT-0.1BT1-0.2BF) based nano particle compositions were synthesized by conventional sol-gel process and investigations were conducted. Since, high concentrations of $BiFeO_3$ may cause tremendous increase in defects that makes the use of the material

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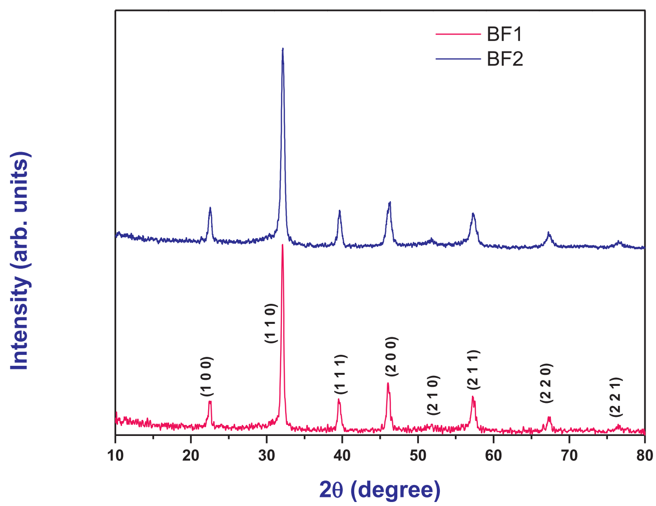

Fig. 1. XRD patterns of BF1 and BF2 ceramics.

difficult for technical applications, the concentration is limited to 0.2 in the NBT matrix. These compositions were processed to achieve stoichiometry in the final powders, high purity and uniform particle sizes. The processed powders were sintered and characterized for their structural, microstructural, ferroelectric and dielectric properties. Moreover, the effect of grain size on the electrical properties was also investigated using high temperature complex impedance spectroscopic analysis and discussed in detail.

2. Experimental

NBT-BT1-BF (NBT-$Na_{0.5}Bi_{0.5}TiO_3$, BT1-$Ba_{0.925}Nd_{0.05}TiO_3$, BF-$BiFeO_3$) based ceramics were prepared by sol-gel method using stoichiometric proportions of Nd_2O_3 (Indian Rare earths Ltd., purity 99%), Ti (100 Mesh, Aldrich 99.7%), $Ba(NO_3)_2$ (SD fine 99.5%), $NaNO_3$ (SD fine 99.5%), $Bi(NO_3)_3$ (Aldrich 99.5%), Fe_2O_3 (SD fine 99.5%), H_2O_2 (30%, SD fine) and ammonia solution (25% AR grade, SD fine) as starting materials. The prepared composition of $0.7Na_{0.5}Bi_{0.5}TiO_3 + 0.2Ba_{0.925}Nd_{0.05}TiO_3 + 0.1BiFeO_3$ is referred to as BF1 and composition of $0.7Na_{0.5}Bi_{0.5}TiO_3 + 0.1Ba_{0.925}Nd_{0.05}TiO_3 + 0.2BiFeO_3$ is referred to as BF2. Initially, stoichiometric amount of $Ba(NO_3)_2$ and $NaNO_3$ were dissolved in double distilled water and $Bi(NO_3)_3$. Nd_2O_3, Fe_2O_3 were dissolved in nitric acid and excess of concentrated nitric acid was removed by heat treatment. Then, the required amount of Ti metal powder was added to a

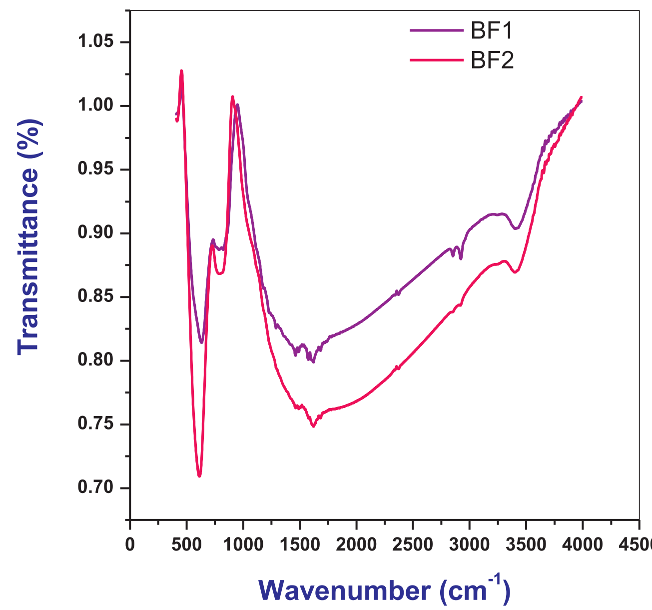

Fig. 3. FTIR spectra of BF1 and BF2 powders.

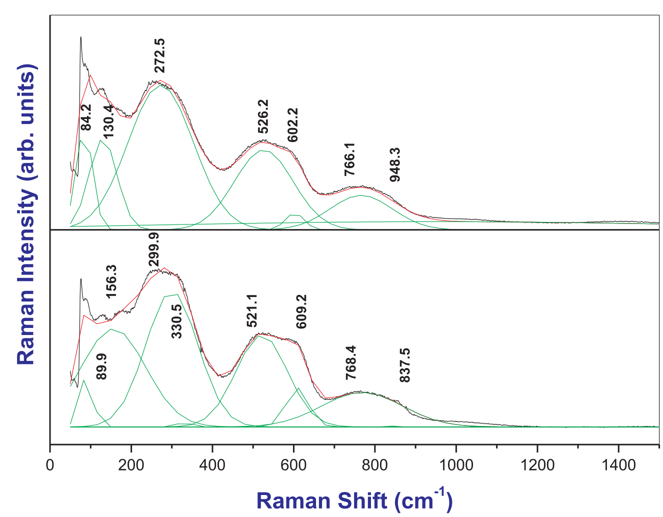

Fig. 4. Raman Spectra of BF1 and BF2 powders.

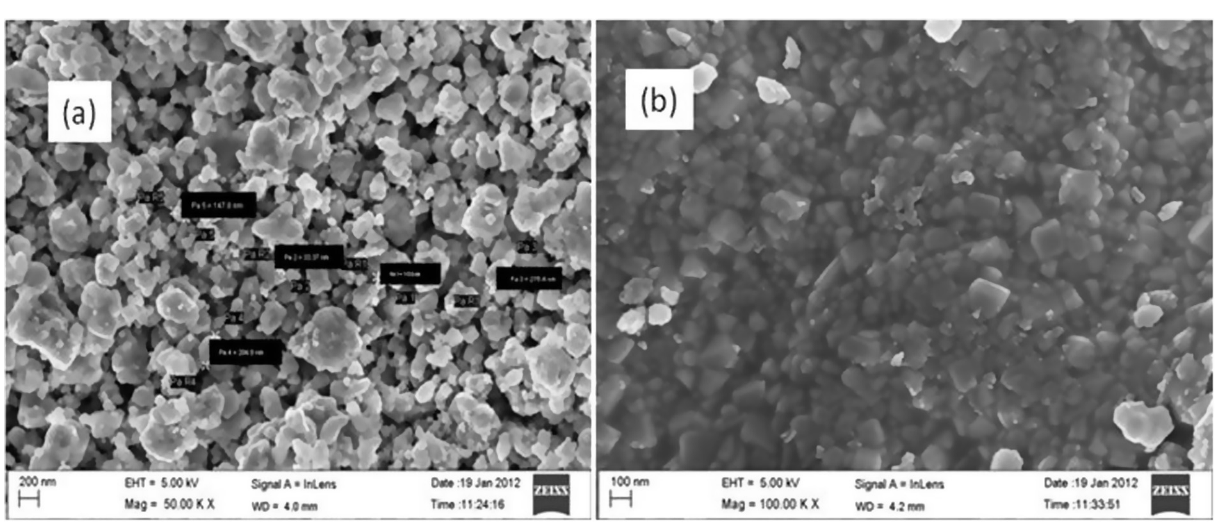

Fig. 2. Powder morphology of (a) BF1 (b) BF2 powders.