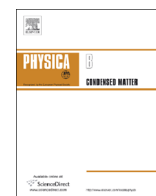




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Dielectric characterization of bismuth layered (Bi_2O_3) ($\text{Na}_x\text{Fe}_{1-x}\text{O}_3$) ceramics

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ABSTRACT

Bismuth layered (Bi_2O_3)($\text{Na}_x\text{Fe}_{1-x}\text{O}_3$) ($0.2 \leq x \leq 0.8$) ceramics have been prepared by the conventional solid-state reaction method. Powder XRD analysis reveals the polycrystalline structure of the samples. Dielectric properties, viz. dielectric constant, dielectric loss and ac electrical conductivity of the ceramics sintered at various temperatures have been studied in the frequency range 10^{-1} – 10^7 Hz. The frequency and temperature dependent conductivity investigations have been carried out by using impedance spectroscopy. It was observed that the samples exhibit bulk conduction which increases with increase in temperature and with the increase in Na^+ ion concentration. The conductivity mechanism at room temperature shows a frequency dependence which can be ascribed to the presence of oxygen vacancies. Combined impedance and modulus plots were used to analyze the sample behavior as a function of frequency and temperature. The effect of temperature on the scaling of dielectric modulus indicates that the conductivity relaxation mechanism is temperature independent. The overlapping of the normalized peaks corresponding to impedance and electrical modulus suggests the single mechanism for the dynamic processes occurring in the present ceramics. The dielectric and conductivity properties of the studied ferroelectric ceramics make it a promising material for fatigue resistance in device applications. Scanning electron microscope (SEM) and energy dispersive X-ray analysis (EDX) confirms the formation of solid solution of BNFO ceramic as major phase.

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1. Introduction

Layered ferroelectric ceramics have received a lot of attention and are widely used in the field of non-volatile random access memories and ceramic multilayer capacitors [1]. In 1950s, Aurivillius synthesized a new series of layered ceramics, which contained bismuth layer as a structural constraint [2,3]. Before 1950s lead based zirconia ceramics were used as dielectric materials but these bismuth layered ceramics have advantage over lead based zirconia ceramics as the later undergo degradation in charge storage properties with the application of electric field. Some layered structure oxides of the Aurivillius family with a general formula $\text{Bi}_2\text{A}_{n-1}\text{B}_n\text{O}_{3n+3}$ consist of regular intergrowth of $\text{A}_{n-1}\text{B}_n\text{O}_{3n+1}$ perovskite-like slabs and Bi_2O_2 sheets, where A is mono-, di-, or trivalent elements (Bi, Na, K, Ca, Sr, Ba, or Pb) allowing dodecahedral coordination and B is trivalent transition elements (Fe, Ti, Nb, Ta, Mo or W) allowing octahedral coordination and n is number of perovskite-like slabs [4]. A large number of different compounds have been obtained from a wide variety of combinations of A and B cations in the thick

perovskite slab of n layers, but the $[\text{Bi}_2\text{O}_2]^{2+}$ sheets have been considered impossible to modify without breaking the structure. The double-sided Bi_2O_2 sheets are composed of square pyramidal BiO_4 groups sharing their basal edge. In these compounds the Bi^{3+} cations combine with trivalent cations Fe^{3+} to maintain the valence state equilibrium [5] and it is assumed that some of the Fe^{3+} ions should go to the A site. Generally, the crystal structure, microstructure and physical properties of materials are influenced by the composition fluctuation and experimental condition [6,7]. Nowadays, much research work has been performed on improvement of dielectric and ferroelectric properties of bismuth layered perovskites [8,9]. It is interesting to note that by doping of various metal oxides in the above bismuth compounds, physical properties of the material can be improved [10]. The major contribution to the ferroelectric polarization of bismuth layered perovskites is due to the displacement of A-site cations along the 'a' axis of the perovskite unit accompanied by octahedral rotations around 'a' and 'c' axes. Because of the presence of Fe ions, these materials not only show ferroelectric properties but also have magnetic ordering [11]. Bismuth layered structure ferroelectric (BLSF) ceramics generally have high curie temperature, coercivities and dielectric strength apart from low dielectric dissipation factor, good mechanical and thermal stability [12]. The impedance spectroscopic method is very effective tool and

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is widely used to characterize dielectric behavior of the polycrystalline ceramic materials. The data may be analyzed in terms of four possible formalisms, the impedance (Z^*), the electric modulus (M^*), the permittivity (ϵ^*), and the admittance (Y^*). These are interrelated as

$$\begin{aligned} M^* &= j\omega C_0 Z^* \\ \epsilon^* &= (M^*)^{-1} \\ Y^* &= (Z^*)^{-1}, \\ Y^* &= j\omega C_0 \epsilon^* \end{aligned} \quad (1)$$

where ω is the angular frequency, C_0 is the vacuum capacitance between the measuring cell and electrodes with an air gap in place of the sample. $C_0 = \epsilon_0/k$, where ϵ_0 is the permittivity of free space $\{8.854 \times 10^{-14} \text{ F/cm}\}$, and $k = l/A$, the cell constant where l is the thickness and A is the area of the specimen, respectively. In general, the dielectric properties of ferroelectric materials arise due to intra-grain, inter-grain and other electrode effects. Complex impedance analysis has been emerged as a very powerful tool for separating out the grain, grain boundary, and electrode contributions in the electrical properties of samples. In the present work, to elicit more information about the mechanism of electrical transport in the titled ceramics, a detailed and systematic investigation on impedance and dielectric measurements have been carried out at different frequencies and temperatures.

2. Experimental

The polycrystalline fine powders of $(\text{Bi}_2\text{O}_3)(\text{Na}_x\text{Fe}_{1-x}\text{O}_3)$ ceramics ($0.2 \leq x \leq 0.8$, x is in step of 0.2) were prepared from solid-state reaction technique by using analytical grade Bi_2O_3 , Na_2CO_3 , Fe_2O_3 in stoichiometric ratios. The exact composition along with sample code of all the prepared ceramics is given in Table 1. The reactants were mixed and grinded mechanically. The ceramic samples were sintered in air atmosphere at optimized temperatures and time, i.e., at 780, 700, 650 and 625 °C for 6 h (to avoid melting) with a uniform increase in temperature at a rate of 1 °C per minute in a programmable electric furnace. The formation of single-phase compounds was checked with an X-ray diffraction (XRD) technique on powdered samples using Rigaku (Miniflex) X-ray diffractometer. Requisite amount of polyvinyl alcohol (PVA) was added as a binder to make the pellets of the powdered samples. The pellets (diameter ~ 13 mm and thickness ~ 1 –3 mm) were prepared using a hydraulic press at 15 T of pressure. After this the pellets were heated at 500 °C for one hour to remove the PVA. For dielectric measurements both the parallel surfaces of the sintered pellets were coated with high purity air-drying silver paint and then dried for 2 h at 200 °C prior to electrical measurements. Electrical measurements of the samples were carried out over a wide frequency range (10^{-1} – 10^7 Hz) at different temperatures (30–500 °C) using a computer-controlled LCR meter/impedance analyzer (Newtons4th Ltd.). Microstructure of the surface was observed using scanning electron microscope. For SEM

analysis the surface of various ceramic powder samples were coated with gold by sputtering method to study the morphology of different crystalline phases. Gold coated ceramic powder samples were also used for energy dispersive X-ray analysis (EDX). EDX spectra were recorded by scanning a representative area of the specimen with electron beam for determination of average composition of the sample.

3. Results and discussion

3.1. XRD and SEM analyses

Fig. 1 shows the XRD patterns of all the sintered BNFO samples. Preliminary examination of the raw X-ray powder diffraction data for all samples suggested that these powders are polycrystalline in nature. The sharp and single reflection peaks were found in the reflection pattern, which are different from those of the ingredient

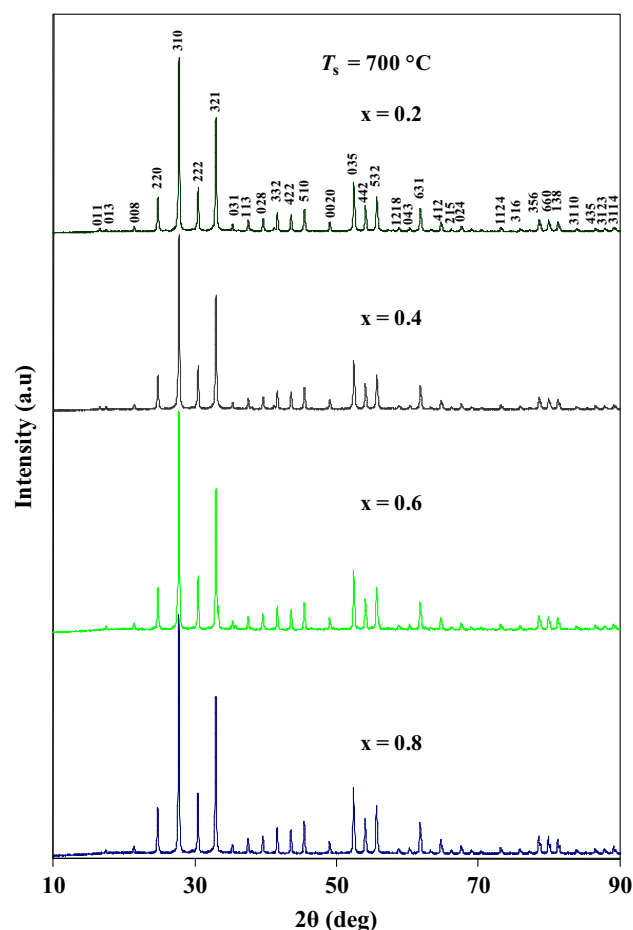


Fig. 1. X-ray diffraction patterns of $(\text{Bi}_2\text{O}_3)(\text{Na}_x\text{Fe}_{1-x}\text{O}_3)$ samples ($T_s = 700$ °C).

Table 1

Real dielectric constant (ϵ'), dielectric loss ($\tan \delta$), dc conductivity (σ_{dc}), ac conductivity (σ_{ac}), dc activation energy (E_{dc}), ac activation energy (E_{ac}) (at high temperature (E_1) and at low temperature (E_2)), peak frequency (f_{M^*}) and complex modulus and impedance relaxation time (τ_{M^*} & τ_{M^*}) of BNFO ceramic.

Sample code	x	ϵ'	$\tan \delta$, (100 °C, 10 Hz)	$\sigma_{dc} \times 10^{-5} (\Omega \text{ m})^{-1}$, (100 °C)	$\sigma_{ac} \times 10^{-6} (\Omega \text{ m})^{-1}$, (100 °C, 10 Hz)	E_{dc} (eV)		E_{ac} (eV)		f_{M^*} (Hz)	$\tau_{M^*} \times 10^{-4}$ (s)	$\tau_{M^*} \times 10^{-4}$ (s)
						E_1	E_2	E_1	E_2			
BNFO1	0.2	477.3	0.43	2.4	3.2	0.62	0.320	0.71	0.44	180	8.83	12.09
BNFO2	0.4	758.3	0.78	3.9	4.1	0.51	0.210	0.64	0.38	179	8.81	8.85
BNFO3	0.6	805.1	0.92	4.2	9.3	0.49	0.165	0.56	0.31	245	6.48	6.50
BNFO4	0.8	1073.8	0.99	4.9	400	0.16	0.123	0.52	0.21	5576	0.28	0.39

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