Orissa Journal of Physics
ISSN 0974-8202

Vol. 30, No. 1
February 2023
pp. 25-30

Studies of Structural and Dielectric Properties of $(Bi_{0.95}Mg_{0.05})(Fe_{0.95}Zr_{0.05})O_3$ Electroceramics

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Received : 5.12.2022 ; Accepted : 4.01.2023

Abstract. The polycrystalline sample $(Bi_{0.95}Mg_{0.05})(Fe_{0.95}Zr_{0.05})O_3$ was synthesized using solid-state reaction method. The room temperature X-ray diffraction analysis confirms the sample is formed in a single phase rhombohedral structure. The dielectric properties of the sample has been studied varying the frequency from 1 kHz to1000 kHz and temperature from 25^0C to 500^0C which provide some important properties of the prepared sample.

Keywords: multiferroics; solid-state reaction; dielectrics; polarization.

1. Introduction

The multiferroics materials are the materials which have the ferroelectric, ferromagnetic, and also ferroelastic behavior in a single phase simultaneously (1,2). Becaause of huge applications in sensors, transducers, energy harvesting devices, transformers, magnetic memories, and microwave and spintronic devices, more attentions are drawn for synthesis of multifunctional materials. Among the different lead free multiferroics materials, $BiFeO_3$ is one of the most widely used multiferroics material having a perovskite (ABO_3) rhombohedral structure and R3c space group symmetry. $BiFeO_3$ posses higher values of Curie temperature (T_c =850°C) and Neel temperature(T_N= 370 °C) (3). Again it has both magnetic and ferroelectric orders above room temperature. But still many inherent problems are faced in BFO because of high leakage current. In recent past, suitable materials are synthesized to overcome the above mentioned problems of $BiFeO_3$ by doping other elements at the Bi and Fe sites (4-6). Doping of La^{3+}, Eu^{3+}, Gd^{3+}, Sr^{2+}, Ba^{2+} , Ca^{2+} at Bi site of $BiFeO_3$ increases the ferroelectric behavior where as doping of Cr^{3+}, Ni^{3+}, Ti^{4+} , Zr^{4+}, Mn^{4+} at Fe-site improves the magnetic behaviors (7,8). In this work, we have tried to improve dielectric and ferroelectric behaviors of BiFeO3 by synthesizing solid solutions of $(Bi_{0.95}Mg_{0.05})(Fe_{0.95}Zr_{0.05})O_3$. In this communication, dielectric properties of $(Bi_{0.95}Mg_{0.05})(Fe_{0.95}Zr_{0.05})O_3$ electroceramics have been presented briefly.

2. Experimental

The desired sample ($Bi_{0.95}Mg_{0.05})(Fe_{0.95}Zr_{0.05})O_3$ was prepared by mixed oxide method at a temperature of 850°C by taking analytical grade (purity>99.9%) oxides such as Magnesium Oxide (MgO), Bismuth oxide (Bi_2O_3), Iron oxide (Fe_2O_3), and Zirconium oxide (ZrO_2) procured from the M/S Loba Chemicals. Above ingredients weigh in a suitable stoichiometric ratio and were homogeneously mixed and ground for 4 h in air and then using methanol for another 4 h. Then the prepared sample was heated at various temperatures and finally heated at 850°C for 5 hours by cylindrical alumina crucible (M/S ANTS ceramics).

$$Bi_2O_3 + MgO + ZrO_2 + Fe_2O_3 \text{ (heat treatment at 800°C)}$$

$$= (Bi_{0.95}Mg_{0.05})(Fe_{0.95}Zr_{0.05})O_3$$

The fine powder of calcinated sample was prepared and then homogeneously mixed by using one or two drops of PVA solution to prepare circular pellets with diameter about 1.2 cm and thickness about 0.1cm to 0.2 cm applying a hydraulic pressure of 4×10^6 N/m^2 by using KBR press (M/S Techno search Instruments Co.). The prepared pellets were sintered at 850ºC for a period of 6 hours. The calcined powder of $(Bi_{0.95}Ba_{0.05})(Fe_{0.95}Zr_{0.05})O_3$ was used for the structural analysis to get the crystal data using Diffractrometer (Rigaku, Japan) which uses CuKa radiation (λ = 1.5405A°) within Bragg angles (2θ) ranging from 20 ° to 80°. The sintered pellets were electroded by using a high-quality silver Paint (M/s Alfa Aesar) after polishing the surfaces for the dielectric measurements. The pellet was kept at a temperature of 120 °C for 2 hours before performing the experiments. The phase-sensitive meter (PSM) (M/S N4L model PSM 1735, UK) is used for various electrical measurements. The measurements were recorded within frequency range 1 kHz to1 MHz and temperature range 25°C to 500°C.

3. Results and Discussions

3.1. *Structural Study*

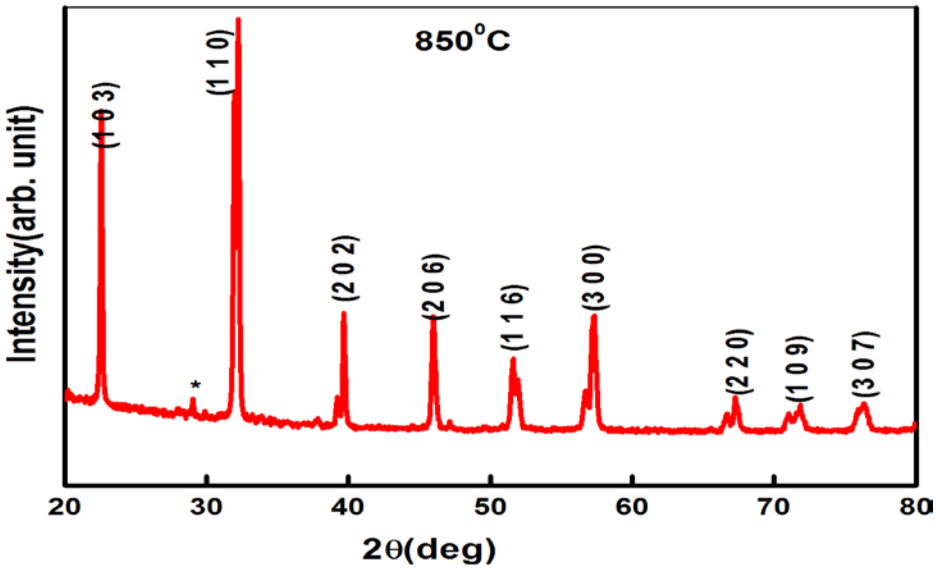

Fig.1: XRD pattern

The X-Ray Diffraction pattern of the prepared polycrystalline sample ($Bi_{0.95}$ $Mg_{0.05}$) ($Fe_{0.95}Zr_{0.05})O_3$ at room temperature for 2θ ranging from 20^o to 80^o is depicted in Fig1. The prominent diffraction peaks are indexed by using POWD software. The basic XRD data consists of all the prominent peaks and confirms that the sample is formed in a single-phase with a prominent impurity peak at $2\theta \approx 29.18^0$(marked as *). Using estimated lattice parameters a=5.5806A^o and c= 13.8616A^0 of unit cell of rhombohedral crystal system indexing of all the prominent peaks were done by computer software POWD (9).

3.2 *Dielectric Study*

The anomalies obtained in the dielectric parameters curves confirm the ferroelectric behavior of the sample(10,11). All the information about different polarizations, relaxation process, cause of dielecric loss, etc can be obtained from dielectric study. The dielectric specimen gets polarized by applying an ac field. Because of polarization, the dielectric parameters of the specimen get changed. The relative permittivity (ε_r) of the sample can be measured at given frequency and temperature using the formula:

$$\varepsilon_r = \frac{C_p t}{\varepsilon_0}$$

Where C_p is parallel capacitance which can be obtained experimentally, t is thickness of the pellet used for measurement and ε_0 is permittivity of free space. The loss in the current component takes place due to temporal phase shift in the driving force and the electrical polarization in the sample. This in turn is the cause of tangent loss of the sample. Both relative permittivity and tangent loss of the material change with the frequency of the applied ac electric field and also on the temperature.

3.2.1. Dependence of dielectric parameters on frequency

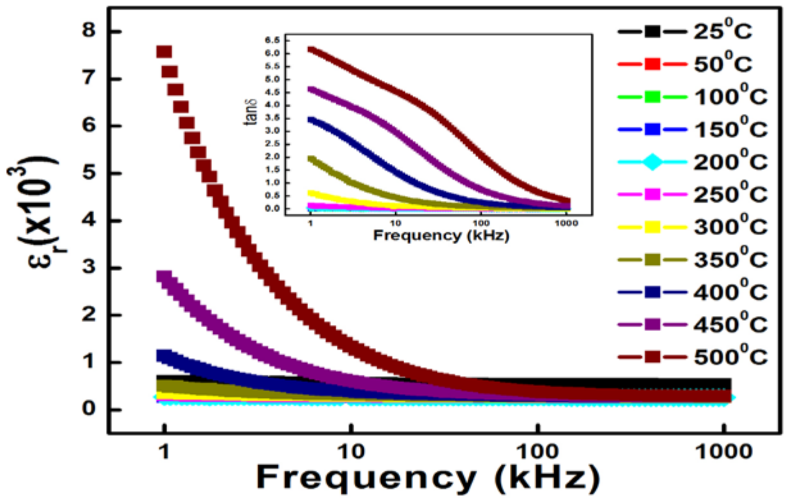

Fig.2: Frequency dependence of dielectric parameters

The variation of the dielectric parameters (relative permittivity (ε_r) and tangent loss (tan δ)) of the sample $(Bi_{0.95} Mg_{0.05}) (Fe_{0.95}Zr_{0.05})O_3$, with frequency (1kHz to1000kHz) at constant temperatures (25^0C to 500^0C) is shown in Fig.2.The graphs confirm that the relative permitivity at lower frequency range has higher values and then starts decreasing with increase in frequency, hence dielectric dispersion takes place at lower frequencies. The decrease in permittivity (ε_r) as well as tangent loss (tan δ) with increase in frequency proves the general characteristics of dielectrics and ferroelectric behavior of the synthesized sample. Again different polarization processes such as dipolar polarization, atomic polarization, ionic polarization, space charge polarization and electronic polarization are the cause of such variation in dielectric parameters. The small displacement produced because of these polarizations is the cause of the net polarization of the material (12). Such variations of dielectric parameters may be explained by a two-layer model given by Maxwell and Wagner (13).

3.2.2. Dependence of dielectric parameters on temperature

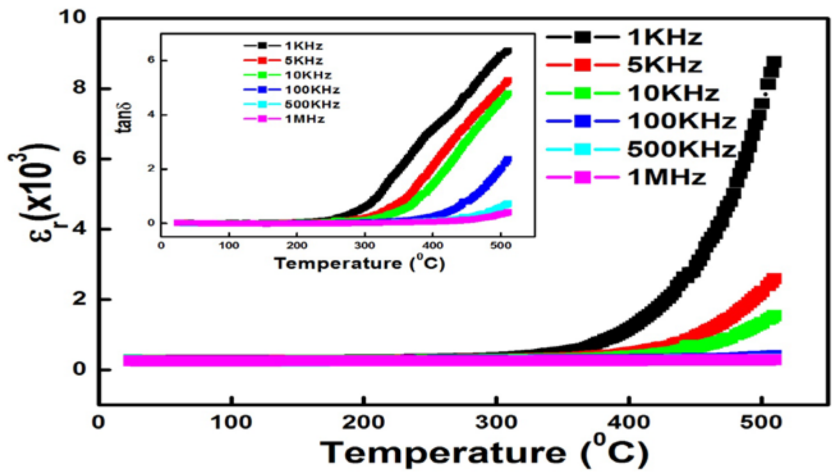

Fig.3: Temperature dependence of dielectric parameters

The temperature dependence of dielectric parameters of $(Bi_{0.95}\ Mg_{0.05})(Fe_{0.95}Zr_{0.05})O_3$ electroceramics, are shown in fig 3. As observed for a particular frequency, relative permitivity (ε_r) increases with increase in temperature and attains a maximum value. The temperature for which permittivity becomes maximum is known as Curie temperature (T_c) (14). Again it is also observed that the maximum value of dielectric constant decreases with rise in frequencies. Similar trends are observed dependence of tangent loss on temperature is taken into consideration. The increase in tanδ is because of defects such as thermally induced motion of charge carriers, oxygen vacancies and presence of the impurity phase of the synthesized sample.

4. Conclusions

The electro-ceramics $(Bi_{0.95}\ Mg_{0.05})\ (Fe_{0.95}Zr_{0.05})O_3$ was prepared by the solid state reaction route. The XRD spectral analysis confirms the sample is formed in a rhombohedral crystal system. The study of variations of relative permittivity (ε_r) and tangent loss (tan δ) with frequency and temperature confirms the dielectric properties of the synthesized material. Because of higher dielectric constant and lower tangent loss, this material is very useful for various multifunctional electronic devices.

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