

Preface

Today's world demands faster, newer, and different products in the market. In the revolutionary innovations, multifunctional devices have received massive attention. Multiferroics are the special class of multifunctional materials in which two or more ferroic orders, i.e., ferroelectricity, ferromagnetism, ferroelasticity, and ferrotoroidicity coexist at the same temperature. These materials have a wide array of structural arrangements along with specific properties like ferroelectricity, ferromagnetism, charge ordering, superconductivity, and colossal magnetoresistance. Multiferroics have fascinating applications in the field of spintronics and nanoelectronics, such as piezoelectric transducers, magnetic actuators with good mechanical properties, multiple-state memory devices, magnetoelectric sensors, spin valves, spin transistors, *etc.*

Since the discovery of multiferroics, perovskites type bismuth ferrite (BFO) has been one of the most promising candidates. It is lead-free piezoelectric material with a high Curie temperature (T_C) of 1103 K. The bulk BFO has a rhombohedral structure with an $R3c$ space group and possesses a high remnant polarization of 90-100 $\mu\text{C cm}^{-2}$. The super-exchange interactions between two magnetically active Fe^{3+} and non-magnetic O^{2-} ions give rise to G-type antiferromagnetic ordering. Despite the extensive properties of BFO, it is very challenging for various applications due to certain inherent obstacles, such as high dielectric loss, thermal decomposition, and the highly volatile nature of BFO, which results in ionized oxygen vacancies and enhances the leakage current by the formation of different impurity phases ($\text{Bi}_2\text{Fe}_4\text{O}_9$, Bi_2O_3 , *etc.*).

In order to improve the magnetic and ferroelectric properties of BFO, doping on A/B- site in BFO-based systems has been carried out. The investigation comprises of the following stages: (i) the Sm content is fixed at 0.1, *i.e.*, $(\text{Bi}_{0.9}\text{Sm}_{0.1})\text{FeO}_3$ (BSFO) system,

and (ii) the BSFO ceramics are synthesized by the solid-state method. Then the influence of different cationic doping in $(\text{Bi}_{0.9}\text{Sm}_{0.1})\text{FeO}_3$ ceramic on the physical and electromagnetic properties is investigated thoroughly. Generally, the A-site of BFO is preferred to be doped with rare-earth ions, which enhances the magnetoelectric properties. The B-site doping is preferred with transition metals, which improve the leakage current. This work aims to contribute the understanding between the structure and electromagnetic properties of doped $(\text{Bi}_{0.9}\text{Sm}_{0.1})\text{FeO}_3$ ceramic.

The Gd doped $(\text{Bi}_{0.9-x}\text{Gd}_x\text{Sm}_{0.1})\text{FeO}_3$ ($x = 0.0 - 0.1$) ceramics are synthesized successfully using solid-state ceramic route. The increase in grain size up to $x = 0.05$ confirms the solubility limit up to $x = 0.05$. The structural and microstructural studies suggest the transformation of phase from rhombohedral to pseudo-cubic phase for $x \geq 0.075$. Also, the composition with $x = 0.075$, i.e., $\text{Bi}_{0.825}\text{Sm}_{0.1}\text{Gd}_{0.075}\text{FeO}_3$ possesses all the ferroic properties viz., high dielectric permittivity, low tangent loss, and higher magnetization with significantly enhanced as compared to the pure bismuth ferrite and samarium substituted bismuth ferrite. Most importantly, in the temperature range of 80 to 300 °C, the $\text{Bi}_{0.825}\text{Sm}_{0.1}\text{Gd}_{0.075}\text{FeO}_3$ sample follows Arrhenius behavior with significantly enhanced conductivity.

The Mg-doped $(\text{Bi}_{0.9}\text{Sm}_{0.1})(\text{Fe}_{1-x}\text{Mg}_x)\text{O}_3$ ($x = 0.0, 0.025, 0.050, 0.075, \text{ and } 0.1$) perovskite systems are observed in a single phase with $R3c$ symmetry. It enhances magnetic and electrical properties with the least leakage current value is $3.64 \times 10^{-7} \text{ A/cm}^2$ reported for $\text{Bi}_{0.9}\text{Sm}_{0.1}\text{Fe}_{0.95}\text{Mg}_{0.05}\text{O}_3$ ceramic. The grain boundary limited conduction, and ohmic conduction are dominated in almost all samples.

Multiferroic-perovskite based ceramic samples having a general formula of $\{(1-x)(\text{Bi}_{0.9}\text{Sm}_{0.1})\text{FeO}_3 - (x)\text{Ba}(\text{Zr}_{0.15}\text{Ti}_{0.85})\text{O}_3\}$ (BSFO – BZT) ($x = 0.0, 0.1, 0.15, 0.2, 0.25, \text{ \&}$

0.5) are successfully prepared by the conventional solid-state route. The structural analysis confirms that BSFO retains its rhombohedral symmetry ($R3c$ space group) up to the $x = 0.025$; further addition of BZT changes the crystal symmetry to tetragonal with $P4mm$ space group. The SEM micrographs show that the increasing BZT content leads to the reducing grain sizes along with diluted irregular-shaped grains. The sample $x = 0.15$ shows maximum dielectric values among all prepared BSFO-BZT samples. The activation energy values suggest the presence of mixed stoichiometric significance of $ABO_{2.95}$ and ABO_3 below 650 K temperature and $ABO_{2.9}$ afterward. The temperature and frequency-dependent impedance analysis confirm the existence of NTCR (a reverse trend with resistance and temperature) for all samples. The sample $x = 0.20$ shows the least possible leakage current value within the series. The sample $x = 0.15$ satisfies both the parameters; it has the largest dielectric constant and a moderate leakage current density value ($1.2 \times 10^{-9} A/cm^2$), which is lower than pure BSFO.

The outcome of the results may find a new material with high electromagnetic properties and reduced leakage current. The as-prepared material can be a good choice for various applications such as non-volatile ferroelectric random-access memory, magnetic data storage, and other electron device applications.

Keywords: *Bismuth ferrite; Gd-doped; Mg-doped; Anti-ferromagnetism; Leakage current; Electromagnetic properties*