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# Electrical properties of $x\mathrm{NiFe_2O_4} + (1-x)\mathrm{Ba_{0.7}Sr_{0.3}TiO_3}$ composites

D.R. Patil, S.S. Chougule, S.A. Lokare, B.K. Chougule *

*Composite Materials Laboratory, Department of Physics, Shivaji University, Kolhapur 416004, India*

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**Abstract**

The standard double sintering ceramic method was used to prepare the magnetoelectric (ME) composites with ferrite and ferroelectric phases. The crystalline phases of the sintered samples were studied by the X-ray diffraction technique. The dielectric parameter such as dielectric constant was determined as a function of frequency (in the range 100 Hz–1 MHz) and temperature (room temperature to 650 °C) at different test frequencies. The DC resistivity as a function of temperature and AC conductivity as a function of frequency were studied for different compositions. Effect of resistivity on ME voltage coefficient is explained.


*Keywords:* XRD; DC resistivity; Dielectric constant; AC conductivity; ME effect

## 1. Introduction

Much interest has been recently created in magnetoelectric materials that generate electrical response (ME output) by application of external magnetic field (called $\mathrm{ME_H}$ effect) or magnetization by external electric field (called $\mathrm{ME_E}$ effect) [1]. In 1894, Curie pointed out the possibility of the polarization of the asymmetric molecular body under an external magnetic field. Subsequently in 1957, Landau and Lifshitz argued that the ME effect could principally exist in some magnetically ordered crystals [3]. The ME effect was observed for the first time in antiferromagnetic $\mathrm{Cr_2O_3}$ in 1961 and later some single phase crystal families were discovered to have ME effect. However, these single phase materials show so weak a ME effect that they have not yet found any technological applications. Alternatively, piezoelectric/ferrite (e.g. $\mathrm{BaTiO_3/CoFe_2O_4}$) composite ceramics have been found to exhibit a larger ME effect than that single phase materials which results from the coupling of magnetic–mechanical–electrical interactions [2].

The ME effect occurs as a product property between piezoelectric and piezomagnetic phases and is not observed in the individual component phases [4]. In order to obtain good ME response first, no chemical reaction should occur between piezoelectric and piezomagnetic materials during the sintering process. Second, the resistivity of the magnetostrictive phase should be as high as possible to prevent the leakage of charges through low resistance path. Third, mechanical defects such as pores at the interface between the two phases should not exist in the composites for good mechanical coupling [5]. Important applications of these materials include magnetic electric sensors in radioelectronics, optoelectronics, microwave electronics and transducers in instrumentation [1].

Literature survey indicates that not much work on the physical properties of magnetoelectric composites has been done. The work available in literature is confined to the measurement of ME effect only. As the ME coefficient of composites largely depends on equilibrium between these two phases, it is necessary to study in detail the electrical behaviour and dielectric properties of these composites. The present paper reports studies on electrical properties such as DC resistivity, AC conductivity and dielectric constant of the present composites in order to understand the conduction mechanism and dielectric behaviour. The resistivity plays a vital role in getting good ME output as mentioned above.

Nickel ferrite is a well-known spinel magnetic material suited for high frequency applications in the telecommunication field due to its high resistivity and low eddy current losses. In addition, $\mathrm{Ba_{0.7}Sr_{0.3}TiO_3}$ (BST) has large figure of merit and superior dielectric and piezoelectric properties.

* Corresponding author. Tel.: +91 231 2609227; fax: +91 231 2691533.
*E-mail addresses:* bkchougule@yahoo.com, sucomposites@yahoo.co.in (B.K. Chougule).

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## 2. Experimental

### 2.1. Preparation

The composites containing two individual phases viz. ferrite ($NiFe_2O_4$) and ferroelectric ($Ba_{0.7}Sr_{0.3}TiO_3$) were prepared by standard double sintering ceramic method. The ferrite phase was prepared through solid-state reaction between AR grade NiO and $Fe_2O_3$ in appropriate molar proportions as starting materials. These basic oxides were mixed and ground in agate mortar for couple of hours and presintered at 900 °C for 9 h. The ferroelectric phase was prepared following the same route by mixing AR grade $BaCO_3$, $SrCO_3$ and $TiO_2$ in their appropriate molar proportions and presintering them at 1000 °C for 10 h. The ME composites were prepared by mixing 15, 30 and 45 mol% of $NiFe_2O_4$ phase with 85, 70 and 55 mol% of $Ba_{0.7}Sr_{0.3}TiO_3$ phase, respectively. The composite mixtures were presintered at 1100 °C for 11 h. The pellets having 3–4 mm thickness and 10 mm diameter were prepared using the hydraulic press. The pellets were sintered finally at 1200 °C for 12 h to yield the final product.

### 2.2. Characterization

The samples were characterized by using X-ray diffractometer (Philips Model PW 1710) using Cu Kα radiation (λ = 1.5418 Å). Silver paste was applied on the surfaces of pellets for good ohmic contact. The DC resistivity was measured in the temperature range 300–800 K by two-probe method.

The dielectric constant was studied as a function of frequency (100 Hz–1 MHz) and as a function of temperature at test frequencies using LCR meter bridge (Model HP 4284 A). The AC conductivity of the samples was determined from the dielectric parameters using the relation

$$\sigma_{AC} = \omega \acute{\varepsilon} \varepsilon_0 \tan\delta \quad (1)$$

where $\varepsilon_0$ is the permittivity of the free space, $\omega$ the angular frequency and $\tan\delta$ is the loss tangent.

## 3. Results and discussion

### 3.1. XRD analysis

The XRD patterns for the composites with $x = 0.15$, 0.30 and 0.45 are shown in Fig. 1. The peaks in the XRD patterns were identified to be characteristics of both $Ba_{0.7}Sr_{0.3}TiO_3$ and $NiFe_2O_4$ phases. The $NiFe_2O_4$ ferrite phase exhibits a cubic spinel structure with lattice parameter $a = 8.33$ Å. The ferroelectric phase $Ba_{0.7}Sr_{0.3}TiO_3$ shows a tetragonal perovskite structure with lattice parameters $a = 3.98$ Å and $c = 3.99$ Å. The lattice parameters match fairly well with the lattice parameters of the components when present as single phase materials. A comparison between the XRD patterns of composites revels that the intensity of ferrite peaks increases with increasing percentage of ferrite in the composites. The number of peaks of ferrite phase also increase with increasing ferrite content. The lattice parameters for all the compositions are given in Table 1.

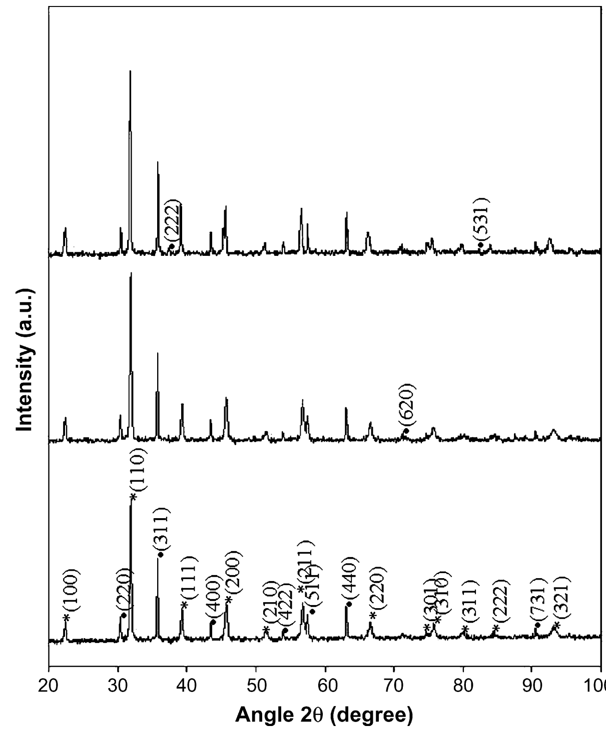


Fig. 1. XRD patterns of $xNiFe_2O_4 + (1 - x)Ba_{0.7}Sr_{0.3}TiO_3$ composites.

### 3.2. Electrical resistivity

Electrical resistivity is an important property as far as the ME composites are concerned. The study of electrical resistivity produces valuable information regarding the behaviour of free and localized electric charge carriers in the samples. The variation of DC resistivity with temperature is shown in Fig. 2. There are two regions of conductivity. The presence of first region observed at

Table 1
Data on electric and dielectric properties of $xNiFe_2O_4 + (1 - x)Ba_{0.7}Sr_{0.3}TiO_3$ ME composites

| Composition $x$ | Lattice parameters | | | $\rho_{RT}$ ($\times 10^9$ Ω cm) | Dielectric constant at $T_c$ | $T_c$ (°C) | $(dE/dH)_H$ (μV/cm Oe) |
|---|---|---|---|---|---|---|---|
| | Ferrite phase | Ferroelectric phase | $c/a$ | | | | |
| 0 | – | $a = 3.97$<br>$c = 3.97$ | 1.00 | 4.4 | – | – | – |
| 0.15 | $a = 8.33$ | $a = 3.96$<br>$c = 3.98$ | 1.00 | 1 | 7358 | 605 | 380 |
| 0.30 | $a = 8.32$ | $a = 3.96$<br>$c = 3.98$ | 1.00 | 0.78 | 4870 | 595 | 343 |
| 0.45 | $a = 8.33$ | $a = 3.96$<br>$c = 3.97$ | 1.00 | 0.56 | 3796 | 590 | 293 |
| 1 | $a = 8.33$ | – | – | 0.03 | – | – | – |