

## Dielectric and magnetoelectric properties of (*x*)Ni<sub>0.8</sub>Co<sub>0.1</sub>Cu<sub>0.1</sub>Fe<sub>2</sub>O<sub>4</sub>/(1 – *x*)PbZr<sub>0.8</sub>Ti<sub>0.2</sub>O<sub>3</sub> composites

S.R. Kulkarni, C.M. Kanamadi, B.K. Chougule\*

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

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### Abstract

Ceramic composites of Ni<sub>0.8</sub>Co<sub>0.1</sub>Cu<sub>0.1</sub>Fe<sub>2</sub>O<sub>4</sub> and lead–zirconate–titanate (PZT) were prepared using conventional solid state reaction method. The presence of constituent phases in composites was confirmed by X-ray diffraction (XRD). The variation of dielectric constant with frequency (100 Hz–1 MHz) and temperature has been studied. The variation of loss tangent (tan δ) with temperature (at frequency 1 kHz) has also been studied. The magnetoelectric (ME) output was measured as a function of dc magnetic field. The maximum value of ME output (625 mV/cm) was observed for 25% ferrite + 75% ferroelectric phase. The maximum ME response can be explained in terms of the content of ferrite, permittivity of dielectric material and the intensity of magnetic field. The ME response of these composites was observed to be linear within low dc magnetic field. These composites may form the basis for the development of magnetic sensors and transducers for use in solid state microelectronics and microwave devices.

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### 1. Introduction

The magnetoelectric effect is a product property exhibiting a complex behaviour. The magnetoelectric materials become magnetized when placed in an external electric field and electrically polarized when placed in a magnetic field [1]. Such a property is not shown by their constituent phases [2]. It occurs due

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\* Corresponding author. Tel.: +91 231 2690571x5230; fax: +91 231 2691533.

E-mail address: [bkchougule@yahoo.com](mailto:bkchougule@yahoo.com) (B.K. Chougule).

to interaction between the magnetic and electric dipoles [3,4]. The ME materials are used as magnetic sensors for dc and ac magnetic field measurements, transducers and actuators [5]. The effect can also be used in various applications such as microwave field and current measurement [6,7], integral optics and fiber communication technology [8].

The ME materials can be classified into two groups, single phase and composites. The single phase materials exhibit ME effect due to local interaction between the ordered magnetic and ferroelectric sublattices [1]. The simultaneous coexistence of electric and magnetic dipoles activates the coupling between the spontaneous polarization and spontaneous magnetization. The ME effect in composite materials is observed as a consequence of the concept of product property introduced by Van Suchetelene [2]. A suitable combination of piezoelectric and piezomagnetic phases gives rise to this property. The ME effect obtained in composite ceramics is reported to be larger than that of single phase ME materials [9].

To obtain better ME effect, the magnitude of magnetostriction coefficient of ferrite phase and the magnitude of piezoelectric coefficient of piezoelectric phase must be high. Also the resistivity of both phases should be high in order to avoid the leakage of accumulated charges through the magnetostrictive phase. Ni ferrites are resistive and magnetostrictive under low magnetic field bias [10]. Hence, we have chosen it as a ferrite phase. PZT is one of the ideal piezoelectric material having high piezoelectric constant, high dielectric permittivity and superior coupling factor [11]. The objective of this paper is to investigate the effect of dielectric behaviour on the ME response of these composites.

## 2. Experimental techniques

### 2.1. Preparation of ME composites

The samples were prepared by standard ceramic method which has many advantages over the unidirectional solidification method [12]. The piezomagnetic ferrite phase was prepared by solid state reaction using NiO, CoO, CuO and Fe<sub>2</sub>O<sub>3</sub> in molar proportions as starting materials. Similarly, the ferroelectric phase was prepared by using PbO, ZrO<sub>2</sub> and TiO<sub>2</sub> in molar proportions. The constituent phases were presintered at 900 °C for 10 h separately. After presintering, the constituent phases were ground to fine powder. The composites were prepared with compositions (x)Ni<sub>0.8</sub>Co<sub>0.1</sub>Cu<sub>0.1</sub>Fe<sub>2</sub>O<sub>4</sub>/(1 – x)PbZr<sub>0.8</sub>Ti<sub>0.2</sub>O<sub>3</sub> where x = 0.15, 0.25, 0.35 and 0.45. These composites were again ground for 3 h so as to mix them thoroughly. The powder was then pressed into pellets having diameter of 1.5 cm and thickness 2–3 mm. The pelletized samples were sintered at 1000 °C for 12 h.

### 2.2. Characterization

The samples were characterized by X-ray diffractometer (Philips Model PW 1710) using Cu K $\alpha$  radiation ( $\lambda = 1.5418 \text{ \AA}$ ). The XRD patterns show the presence of constituent phases. The patterns do not indicate any chemical reaction between the components during sintering.

### 2.3. Measurement of dielectric constant and $\tan \delta$

The dielectric measurements were carried out as a function of frequency in the range of 100 Hz–1 MHz at room temperature and temperature at frequencies of 1 kHz, 10 kHz, 100 kHz and 1 MHz

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