Journal of Magnetism and Magnetic Materials 324 (2012) 944–948

Contents lists available at SciVerse ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Study of magnetic properties and magnetoelectric effect in $(x)Ni_{0.5}Zn_{0.5}Fe_2O_4+(1-x)$PZT composites

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ARTICLE INFO

Article history:
Received 16 March 2011
Received in revised form
5 August 2011
Available online 5 October 2011

Keywords:
Magnetoelctric effect
Composites
Single domain
Resistivity

ABSTRACT

Magnetoelectric (ME) composites consisting of ferrite phase (x) $Ni_{0.5}Zn_{0.5}Fe_2O_4$+ferroelectric phase $(1-x)$Pb $Zr_{0.8}Ti_{0.2}O_3$ (Lead Zirconate Titanate—PZT) in which x (mol%) varies between 0 and 1 $(0.0 \le x \le 1.0)$ was synthesized by double sintering ceramic method. The presence of constituent phases of ferrite, ferroelectric and their composites was confirmed by X-ray diffraction studies. The hysteresis measurement was used to study magnetic properties such as saturation magnetization (M_S) and magnetic moment (μ_B). The existence of single domain (SD) particle in the ferrite phase and mixed (SD+MD) particle in the composites was studied from AC susceptibility measurements. ME voltage coefficient for each mol% of ferrite phase was measured as a function of applied DC magnetic field and at the same time influence of magnetic field on ME response and resistivity of composites was studied. The maximum ME voltage coefficient of 0.84 mV/cm Oe was observed for 15% of ferrite phase and 85% of ferroelectric phase in the composites.

1. Introduction

Composite materials are gaining increasing importance due to their fundamental and technological applications in electronic industry. Composite materials consisting of ferroelectric and ferromagnetic materials are expected to exhibit new properties such as magnetoelectric, magnetooptic and other new coupling properties. From the effect of coupling between magnetization and electric polarization writing of a data with an electric field and reading it with a magnetic field, and vice versa, was made possible [1–5]. Certainly this property offers additional advantage in designing the electronic devices [6].

The magnetoelectric materials are a kind of interesting new materials used for sensors, processors, actuators, memory system, etc. [7]. Initially the ME effect was observed in a single crystal (Cr_2O_3) where ME effect was due to the local interaction between the ordered magnetic and ferroelectric sub-lattices [8]. Later on a variety of single phase ME materials were discovered for the last four decades for the study of ME effect [9]. However, most of the single phase materials have weak ME voltage coefficient and involve large processing technique; in addition they are expensive and suffer from degradation under cyclic conditions and are not useful in device applications [10]. Composite materials are the ultimate solutions to these limitations. Composite is a combination of at least two chemically distinct materials with an interface separating the components that find equilibrium between ferroelectric and magnetic structures preserving both properties close to the room temperature. The main advantages of ME composites are that they are cheaper and easier to fabricate and one can control the molar ratio of phases, grain size of each phase and densification [11]. Following the concept of product (combined) property, Suchetelene [1] suggested that the composites of piezomagnetic and magnetostrictive phases must be electromagnetically connected through stress medium. In this the ME effect was due to the combined deformation of the defused structure of the ferroelectric and ferrite components. Primary deformation of the magnetostrictive phase causes polarization of the ferroelectric particles of composites and electric polarization of ferroelectric materials causes change in magnetization of the ferrite phase due to mechanical coupling in ferrite and ferroelectric phases. Various research groups focused on the synthesis methods to synthesize bulk composites and the layer of heterostructure [5]. Among these methods, double sintering solid state reaction method was widely used and the physical property of resulting materials was studied extensively. In addition, in this method one can control the molar ratio of phases and grain size of each phase with sintering temperature [11,12].

In this article we discussed the magnetic properties and magnetoelectric effect of the composites consisting of Ni–Zn ferrite with PZT ferroelectric phases at room temperature. The soft ferrite ($NiFe_2O_4$) is a promising material as it has low anisotropy and high initial permeability. At the same time the larger magnitude of magnetic moment may be due to the ion

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doi:10.1016/j.jmmm.2011.09.016

rearrangement and ultimately favors better ME effect. ME voltage coefficient of the composite can be measured by putting additional amount of zinc oxide as one of the divalent components in nickel ferrite, which is one of the conditions to increase the efficiency of ME conversion factors. In addition, zirconium-doped lead titanate has the advantage of superior coupling factor with good piezoelectric properties [13]; therefore measurements of magnetoelctric effect and magnetic properties of this system were expected to be higher at room temperature

2. Experimental

2.1. Preparation of ME composites

ME composite $[(x)\ Ni_{0.5}Zn_{0.5}Fe_2O_4+(1-x)PbZr_{0.8}Ti_{0.2}O_3]$ was prepared using standard double sintering solid state reaction method. The $Ni_{0.5}Zn_{0.5}Fe_2O_4$ ferrites were prepared by maintaining the stoichiometry of high purity AR grade NiO, ZnO and Fe_2O_3 raw powders and then milled together for 2–3 h and presintered at 800 °C for 8 h in air. The synthesis of $PbZr_{0.8}Ti_{0.2}O_3$ ferroelectrics was made using AR grade PbO, TiO_2 and ZrO_2 raw powders in molar proportion and then milled for 2–3 h and presintered at 900 °C for 10 h in air. ME composites was prepared by mixing 15, 30 and 45 mol% of ferrite phase with 85, 70 and 55 mol% of the ferroelectric phase and then presintered at 1100 °C for 10 h. Afterwards these compositions were mixed with 2% of polyvinyl alcohol as a binder and then pressed into pellets of 15 mm diameter and 2–3 mm thickness using a hydraulic machine. The samples in the forms of pellets were finally sintered at 1150 °C for 12 h in a programmable furnace at the heating rate of 100 °C/h and slowly cooled to room temperature at the same rate.

2.2. Characterization

The crystal structure and crystalline phases of the composites as well as their constituent phases were determined by X-ray diffractometer (Philips Model PW-1710) with CuK_α radiation ($\lambda=1.514$ Å) in a wide range of Bragg angle 2θ ($10^\circ \le 2\theta \le 80^\circ$) at a scanning rate of 2°/min. The magnetic hysteresis loop tracer was used to measure the saturation magnetization (M_S) and magnetic moment (μ_B) of the sample and the saturation magnetization (M_S) was estimated by

$$M_S=(1-P)\sigma_s' d_x \tag{1}$$

where P is the porosity and d_x is the X-ray density.

The magnetic moment was estimated by

$$\mu_B=\frac{M\sigma_S'}{5585} \tag{2}$$

where M is the molecular weight and σ_S' is the magnetization per gram mole of the sample. The AC susceptibility measurements of the sample were carried out in the temperature range of 100–800 °C.

The magnetoelectric measurements of the samples were carried out under induced electric polarization (by an applied magnetic field) and induced magnetization (by an applied electric field). Electric and magnetic poling of the composites was made to increase the magnetostriction coefficient of the ferrite phase and the piezoelectric coefficient of the ferroelectric phase. The electric poling was carried out by heating the samples at about 30 °C above the ferroelectric Curie temperature in an external electric field of 2.5 kV/cm. The magnetic poling was carried out by applying DC magnetic field of 6 K Oe at room temperature and in the same setup the static ME voltage coefficient $(dE/dH)_H$ was estimated. The magnetic field applied normal to the flat and polished surfaces of the composite pellets with good electric contacts induces the electric voltage as a function of magnetic field and was measured with high impedance kiethley electrometer (model 2000).

3. Results and discussion

3.1. Phase identification

The X-ray diffraction patterns of ferrite ($Ni_{0.5}Zn_{0.5}Fe_2O_4$) phase (Fig. 1a), ferroelectric (PZT) phase (Fig. 1b) and their composites are presented in Fig. 2. X-ray diffraction patterns of ferrites, ferroelectrics and composites show well defined peaks without intermediate phase formation in the composites. The occurrence of peaks with specific indices indicates spinel and perovskite structure of the samples, which confirms cubic spinel structure in ferrite phase and tetragonal perovskite structure in ferroelectric phase of the composites. However, no structural changes were observed for both the phases in the composites. The lattice parameters for both the phases are given in Table 1. The ferrite phase has a cubic spinel structure with lattice parameter $a=8.342$ Å and the ferroelectric phase has tetragonal perovskite structure with $a=4.128$ Å and $c=4.129$ Å, $c/a=1.0002$. Using the following Scherer's formula average particle size (Table 1) of both the phases was estimated by broadening the XRD peaks of the phases:

$$D_{hkl}=\frac{0.9\lambda}{\beta_{hkl}\cos\theta_{hkl}} \tag{3}$$

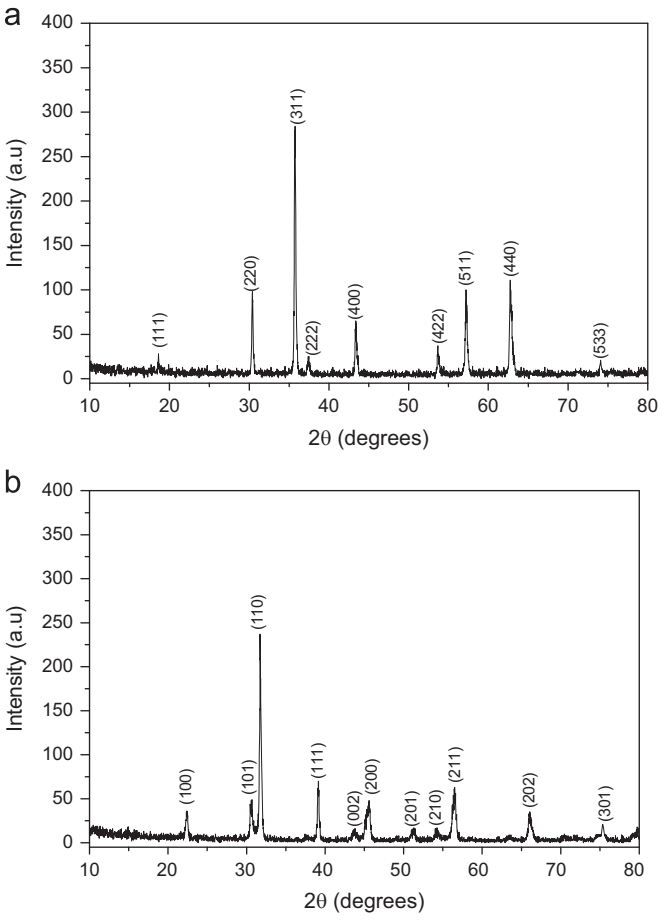

Fig. 1. (a) XRD pattern of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ ferrite phase. (b) XRD pattern of $PbZr_{0.8}Ti_{0.2}O_3$ ferroelectric phase.