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Materials Research Bulletin

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Improvement of the physical properties of novel $(1 - y) Co_{0.8}Cu_{0.2}Fe_2O_4 + (y)$ SrTiO₃ nanocomposite

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ARTICLE INFO

Article history: Received 13 June 2012 Received in revised form 29 December 2012 Accepted 7 January 2013 Available online 24 January 2013

Keywords: A. Magnetic materials

B. Chemical synthesis C. X-ray diffraction D. Dielectric properties

ABSTRACT

Magnetoelectric (ME) nanocomposites $(1 - y) \operatorname{Co}_{0.8}\operatorname{Cu}_{0.2}\operatorname{Fe}_2\operatorname{O}_4 + (y) \operatorname{SrTiO}_3 (y = 40, 50, 60, 80 and 100%)$ were prepared by standard ceramic method. Phase formation was checked using X-ray diffraction analysis. Both saturation magnetization (M_s) and Curie temperature (T_c) decrease with increasing SrTiO₃ content. Temperature dependence of the dielectric constant reveals two maxima, one about 550 K corresponds to non-stoichiometry and lattice distortions while the second around 900 K corresponds to the Curie temperature (T_c). The large value of ME output is due to the strain induced by lattice distortion in the ferrite phase by Jahn–Teller ions like Cu. Hence, Jahn–Teller effect in the ferrite leads to polarization in the piezoelectric phase.

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1. Introduction

Simultaneous ferroelectric and ferromagnetic properties can be achieved in multiferroic composites. Such multiferroic composites consist of a ferroelectric and a ferromagnetic phase. The coupling between the two order parameters is through a stress mediation, i.e., a magnetic field induces a distortion of the magnetostrictive phase, which in turn distorts the piezoelectric phase in which an electric field is generated [1]. Accordingly the composite can be considered as a new material with multiferroic properties. The magnetoelectric effect is extrinsic in this case since magnetoelectric effect is not exhibited by any of the constituent phases on their own. Such physical property of the composite is called a "product property" [2], refer to an effect in one of the phases or submaterials which in turn leads to a second effect in the other phase. The advantages of multiferroic composites over single phase multiferroics are (i) the physical properties of the materials can be tailored by the selection of different constituent phases and their volume ratios in order to meet specific applications. (ii) The magnetoelectric coupling effect in multiferroic composites is much higher than single phase ferroelectromagnets. Many researchers have studied Co ferrite and BaTiO₃ [3-5]. No studies are available on Co-Cu

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ferrite and SrTiO₃ composite. SrTiO₃-based compounds have been attracting a considerable interest both from a fundamental point of view and for a wide range of applications, particularly in tunable electronic devices [6]. In the present work, (1 - y) $Co_{0.8}Cu_{0.2}Fe_2O_4 + (y)$ SrTiO₃ (y = 40, 50, 60, 80 and 100%) was selected as a model system. The perovskite oxides are most commonly used in oxygen sensors such as SrTiO₃, the intrinsic ionic point defects in which are strontium and oxygen vacancies [7-9]. Transition metal titanates with a perovskite crystal structure are considered as promising material. The interest in SrTiO₃ is due to (1) the gravimetric capacity matching well with currently available cathode materials, (2) sufficient discharge kinetics for a wide range of applications, (3) good safety characteristics, and (4) a high durability, i.e. relatively negligible irreversible capacity losses are observed, also SrTiO₃ is a semiconductor, the resistance arising from the low electronic conductivity and surface boundary interfaces can have a detrimental effect on performance [10].

2. Experimental procedure

Cobalt Copper ferrite $Co_{0.8}Cu_{0.2}Fe_2O_4$ was prepared using a conventional double sintering ceramic process $Co_{0.8}Cu_{0.2}Fe_2O_4$ using analytical oxides (BDH). CoO, CuO and Fe_2O_3 as starting materials were mixed together in stoichiometric ratios and grinded for 4 h. The mixtures were presintered at 800 °C for 5 h with heating rate of 4 °C/min and cooled to room temperature with

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the same rate as that of heating. After that, the samples were pressed in the form of disc using uniaxial press of 5×10^8 N/m² and finally sintered at 1100 °C for 5 h in air and then slowly cooled to room temperature with rate of 4 °C/min. Similarly SrTiO₃ was prepared starting with SrCO₃ and TiO₂, mixed together in stoichiometric ratios and grinded for 4 h. The mixtures were presintered at 1200 °C for 5 h with heating rate of 4 °C/min and cooled to room temperature with this same rate.

The cobalt copper ferrite (Co_{0.8}Cu_{0.2}Fe₂O₄) powder was good mixed with the ferroelectric phase (SrTiO₃) to prepare the nanocomposites (1 - y) Co_{0.8}Cu_{0.2}Fe₂O₄ + (y) SrTiO₃; SrTiO₃ (y = 40, 50, 60, 80 and 100%) and grinded for 2 h at room temperature. Cold pressing for this composite was carried out using a uniaxial press of value 5×10^8 N/m² into a pellet form. The composite samples were finally sintered at 1300 °C for 3 h. X-ray diffraction was carried out using radiation source Cu K α radiation ($\lambda = 1.5405$ Å). The measurements of magnetic susceptibility χ_{M} as a function of temperature were carried out using Faraday's method at different magnetic field intensities. The accuracy of measuring temperature was ± 1 °C.

The magnetization M (emu/g) was measured at room temperature in a magnetizing field H (Oe) ranging from 0.0 up to 6000 Oe, using a vibrating sample magnetometer (VSM, EG&G model no. 1551, USA). The LCR meter (Fluka model PM6306) was used to measure the ac electrical resistivity and dielectric properties of the investigated samples. The measurements were carried out from room temperature up to 980 K at different frequencies ranging from 100 Hz to 1 MHz.

The polarization P was measured at room temperature as a function of electric field E using a homemade Sawyer-Tower circuit.

The ME outputs were measured using a homemade circuit in which two copper electrodes were brazed to the electrical leads and were kept on either side of the poled sample. The whole sample holder assembly was kept between the pole pieces of a DC electromagnet. All stray pick-ups have been avoided by proper grounding of the experimental set-up. The two end leads from the sample were connected to electrometer Keithly 610 through a shielded cable. The measurement of output voltage is made after stabilization of the magnitude of the magnetic field. The output voltage developed across the sample was measured as a function of increasing DC magnetic field.

3. Results and discussion

Fig. 1 presents the XRD patterns of the composites with different $SrTiO_3$ concentration. The data show two well defined



Fig. 1. XRD patterns of the composites $(1 - y) Co_{0.8}Cu_{0.2}Fe_2O_4 + (y) SrTiO_3$ (y = 40, 50, 60 and 80%).

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