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Multiferroic (NiZn) Fe_2O_4–$BaTiO_3$ composites prepared from nanopowders by auto-combustion method

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Abstract

Nickel zinc ferrite (NZF) and barium titanate (BT) were prepared by auto-combustion synthesis as an effective, simple and rapid method. Multiferroic composites with the general formula $yNi_{1-x}Zn_xFe_2O_4-(1-y)BT$ ($x=0.3$, 0.5, 0.7, $y=0.5$) were prepared by mixing NZF and BT powders in a liquid medium in the ball mill. The FEG micrographs indicated the primary particle size less than 100 nm for both, barium titanate and nickel zinc ferrite phases. X-ray analysis and Raman spectroscopy indicated the formation of well crystallized structure of NZF and BT phase in the composite powders and ceramics, with a small contribution of the secondary phase. The homogenous phase distribution in obtained composites was also confirmed. Impedance spectroscopy measurements were carried out in order to investigate the electrical resistivity of materials, showing that grain boundaries have greater impact on the total resistivity than grains. Saturation magnetization and remnant magnetization continuously decrease with barium titanate phase increase.

Keywords: A. Powders: chemical preparation; B. Composites; C. Impedance; D. $BaTiO_3$ and titanates; D. Ferrites

1. Introduction

Miniaturization of the solid-state electronics is achieved by downscaling and multifunctionality. Ferroics and multiferroics are among the most attractive multifunctional materials [1]. These nanostructured materials stimulated a sharply increasing interest to their significant technological promise in novel devices due to fact that the combination of dissimilar materials in ferroic-based oxide nanocomposites resulted in totally novel functionality.

The term multiferroic (MF) was first used by Schmid in 1994. His definition referred to multiferroics as a single phase materials which simultaneously possess two or more primary ferroic (ferroelectric, ferromagnetic and ferroelastic) properties. Today the term multiferroic has been expanded to include materials which exhibit any type of long range magnetic ordering, spontaneous electric polarization, and/or ferroelasticity. Working under this expanded definition the history of magnetoelectric multiferroics can be traced back to the 1960s [1–3]. In the most general sense the field of multiferroics was born from studies of magnetoelectric systems [4–6]. After an initial burst of interest, research remained static until early 2000. In 2003 the discovery of large ferroelectric polarization in epitaxially grown thin films of $BiFeO_3$ [7] and the discovery of strong magnetic and electric coupling in orthorhombic $TbMnO_3$ and $TbMn_2O_5$ have stimulated activity in the field of multiferroics. Besides scientific interest in their physical properties, multiferroics are interesting due to their potential applications as transducers, actuators, switches, magnetic field

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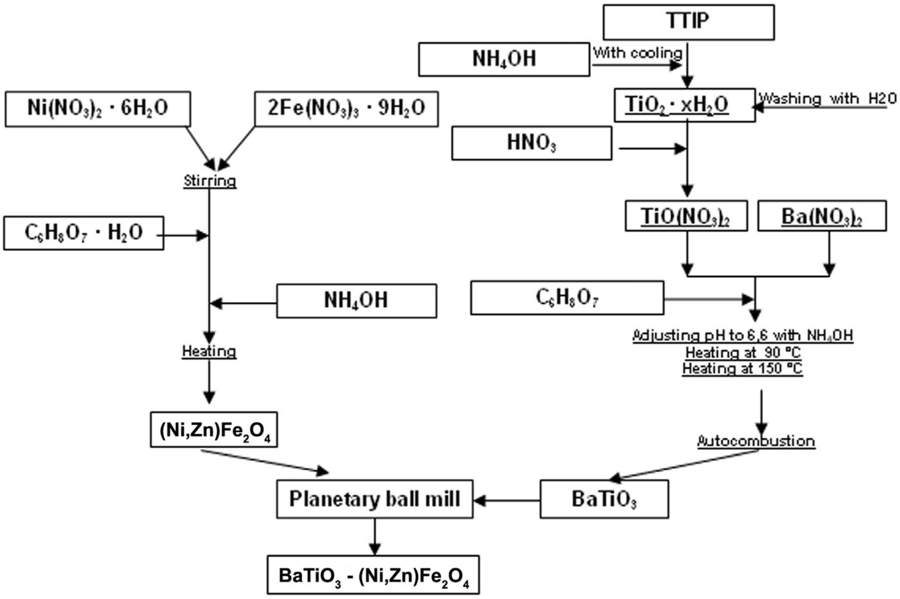

Fig. 1. Scheme of NZF, BT and composites preparation.

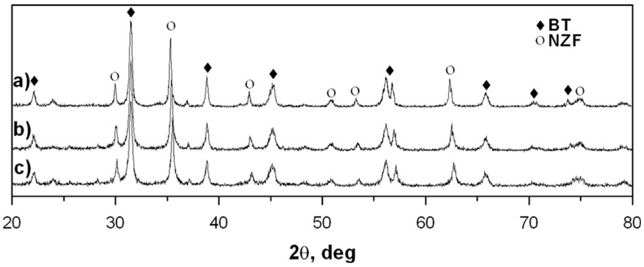

Fig. 2. The X-ray diffraction patterns of (a) NZF(30–70) – BT, (b) NZF(50–50) – BT and (c) NZF(70–30)–BT powders.

sensors, new types of electronic memory devices, capacitive/inductive passive filters for telecommunications, etc. [8,9]. A large number of publications have been dedicated to multiferroics, dealing with theoretical, experimental, and application aspects [2,10]. In spite of hundreds of publications focused to single or composite multiferroic materials in the last years, they remain highly controversial concerning their preparation methods, phase stability, intrinsic polarization and switching, ferroelectric, ferromagnetic and magnetoelectric properties, etc. [10].

Multiferroic properties can appear in a large variety of materials [11]. The ferroelectric–ferromagnetic composites, as two-phase multiferroic materials, are desired not only for the fundamental research of magneto-electric effect, but also for the potential applications in many electronic devices [12]. The most widely studied systems correspond to Co or Ni ferrites, with PZT, PNT, BT or BST [13]. Among them, the Ni–Zn ferrites/$BaTiO_3$ systems need to be further investigated because of high electrical resistivity, chemical stability and excellent electromagnetic properties of the Ni–Zn ferrites, and high permittivity, low dielectric loss and high tunability of $BaTiO_3$ [4–6]. Those composites have attracted considerable attention as a new class of nanoferrites, expanding their use in other areas, such as drug delivery, heterogeneous catalysis, levitated railway system, magnetic-refrigeration, microwave devices, antennas, etc. [14].

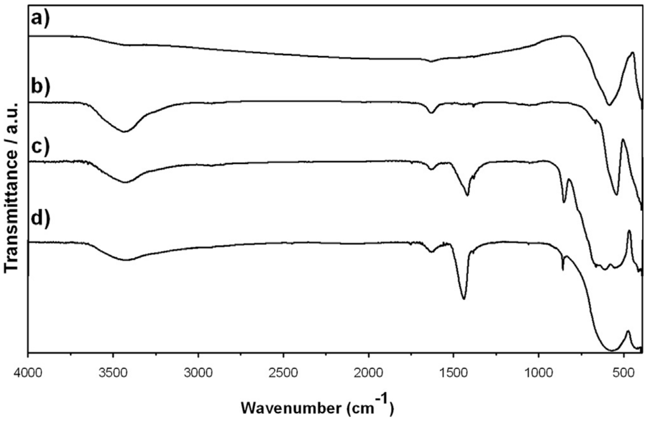

Fig. 3. FT-IR spectra of (a) NF, (b) ZF, (c) BT and (d) NZF(70–30) – BT powders.

To obtain the multiferroics, several routes for conventional material fabrication are being applied. Popular techniques within the multiferroic community are: solid state synthesis, hydrothermal synthesis, sol–gel processing, vacuum based deposition or other wet chemical synthesis methods. However, some types of multiferroics require specific processing conditions within more appropriate techniques. Consequently, multiferroic composites request methods for the synthesis both components: ferroelectric and ferromagnetic.

Ferrites crystallize in three crystal type: spinel, garnet type and magnetoplumbite type [15,16]. Meanwhile, the main attention is stressed to spinel type of ferrites that can be synthesized by a sol–gel method, conventional solid state reaction, mechanical attrition, hydrothermal synthesis, self-propagating combustion method, thermolysis, wet chemical co-precipitation technique, self-propagating, microemulsion,