DIELECTRIC AND ELECTRIC PROPERTIES OF NOVEL CORE-SHELL NANOComposite: SrFe$_{12}$O$_{19}$ – BNT-BT

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Abstract

The dielectric and electric properties of a core-shell SrFe$_{12}$O$_{19}$ – BNT-BT nanocomposite were explored in this study. The desired composition and the existence of the magnetoplumbite SrFe$_{12}$O$_{19}$ and perovskite BNT-BT structures were verified by X-ray diffraction. The dielectric constant values approached the case of BNT-BT due to the small amount of hexaferrite content. The electric properties were also derived.

Key words: multiferroics, dielectrics, core-shell

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1. INTRODUCTION

Presently there are strong ongoing efforts in the research community to remove lead from the industry due to toxicity, which has drastic effects on health (e.g. of industrial workers) and environment [1]. In this respect, promising lead-free composites for dielectric devices are represented by lead-free hexaferrites and perovskites [2–4]. The dielectric performances are considerably influenced by substitutions, sample geometry, preparation method and further processing such as sintering [5]. Sol-gel synthesis is an effective, versatile, and low-cost synthesis technique to prepare nanoparticles with a large number of functionalities [6]. It was proven that the core-shell geometry (through a seed mediated process) is the best approach towards dielectric properties relative to regular nanocomposites [7].

In this study, a novel core-shell composite of strontium hexaferrite and (BiNa)$_{0.5}$TiO$_3$ – BaTiO$_3$, SrFe$_{12}$O$_{19}$ – BNT-BT, was investigated from the point of view of structural, dielectric and electric properties. To the best of our knowledge, this is the first report on the dielectric effect in these particular composites in the form of core-shell.

2. MATERIALS AND METHODS
The core-shell SrFe\textsubscript{12}O\textsubscript{19} (SFO) – Bi\textsubscript{0.5}Na\textsubscript{0.5}TiO\textsubscript{3}-0.08BaTiO\textsubscript{3} (BNT-BT) composite was prepared by a sol gel method based on seed mediated growth. Ba(OAc)\textsubscript{2}.H\textsubscript{2}O and glacial CH\textsubscript{3}COOH were mixed and dissolved, after which Ti(O\textsubscript{i}Pr\textsubscript{4}) was added and slowly heated to give a gel. The pre-formed magnetic core (SFO, corresponding to 2 mol. % with respect to BNT-BT) was added to the gel of BNT-BT precursors. The mixture was then sonicated for 1h and kept at 80°C for another hour, after which a thermal program was started: heating in a Nabertherm furnace with 5°/min room temperature to 700°C, kept for 30 min. at 700°C and then cooled natural to room temperature. The core-shell SFO-BNT-BT was confirmed to conform to the said formula by powder XRD performed after cooling. The powder was then pressed in a 1 mm thick, 10 mm diameter pellet and then sintered at 1100°C for 15 min., followed by natural cooling to room temperature.

The X-ray diffraction (XRD) measurements were performed by Bruker D8 Advance X-Ray Diffractometer. The MAUD program was used for the Rietveld refinement [8]. Dielectric measurements of the sintered ceramic sample were performed using an AGILENT 4294 A Precision Impedance Analyzer (Agilent Technologies, Santa Clara, CA). The electric properties were investigated by a Premier II ferrotester (Radiant Technologies) using a triangular voltage signal. Typical hysteresis and generalized PUND (positive-up-negative-down) characterizations were performed.

3. RESULTS AND DISCUSSIONS

Figure 1 shows the diffraction patterns of the sample A and the results derived from Rietveld refinement are listed in table 1. The hexagonal magnetoplumbite SrFe\textsubscript{12}O\textsubscript{19} phase (space group P6\textsubscript{3}mcm) and the perovskite BNT-BT phase (space group P4bm) were evidenced in the XRD patterns. The desired ratio of the components are roughly obtained. The crystallite size of the SFO phase is 50% higher relative to the BNT-BT phase.
Figure 1. XRD pattern of the composite sample A.

Table 1 Molar compositions, crystallographic data and structure Rietveld refinement parameters.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Contrib. (% mol.)</th>
<th>rSFO (Å)</th>
<th>rBNT-BT (Å)</th>
<th>abSFO (Å)</th>
<th>abBNT-BT (Å)</th>
<th>dsSFO (nm)</th>
<th>dsBNT-BT (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>4</td>
<td>96</td>
<td>5.8822</td>
<td>23.0625</td>
<td>5.5166</td>
<td>3.9176</td>
<td>104.8</td>
</tr>
</tbody>
</table>

Figure 2. Dielectric constant and loss (tan(δ)) of sample A.
The dielectric properties of the composite sample A were studied by measuring the capacitance and dielectric loss, at different frequencies between 1 kHz and 1 MHz. They are represented in figure 2. The dielectric constant values, roughly of the order of $10^3$ at low frequencies, are similar to BNT-BT ceramics [9], due to the small amount of the SrFe$_{12}$O$_{19}$ contribution. The dielectric loss values are relatively low, suitable for dielectric devices.

The electric hysteresis of sample A are shown in figure 3. The parameters of the hysteresis are as follows: maximum polarization of 6 μC/cm$^2$ (continuous line), real remanent polarization of 0.6 μC/cm$^2$ (small dash line, obtained by PUND method) and coercive field of 1500 V/mm. Dynamic dielectric effects are evidenced.

![Electric hysteresis of sample A](image)

Figure 3. Electric hysteresis of sample A.

**4. CONCLUSIONS**

Core-shell SrFe$_{12}$O$_{19}$ – BNT-BT$_{0.08}$ nanocomposite was prepared by sol gel and investigated by X-ray diffraction, Impedance analyzer, Ferrotester and lock-in amplifier. The diffraction is confirming the involved phases. The dielectric constant
values at low frequencies attained about 950, and relatively low dielectric loss. The electric behavior was described.

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