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Short communication

Preparation and electromagnetic properties of in-situ Ba_{0.8}Sr_{0.2}TiO₃/YFeO₃ composites

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Abstract

Ba_{0.8}Sr_{0.2}TiO₃/YFeO₃ (BST/YIP) composites with giant dielectric constants and high saturation magnetizations were synthesized via the in-situ growth solid-state method. The BST/YIP composites were sintered at 1350 °C, using Ba_{0.8}Sr_{0.2}TiO₃ and Y₃Fe₅O₁₂ as raw materials. The phase composition and surface morphology of the composites were investigated using XRD and SEM, respectively. The dielectric and magnetic properties of the composites were also studied. The results show that the BST/YIP composites have giant dielectric constants and high saturation magnetization. For the 40% BST/60% YIP composite, the dielectric constant and saturation magnetization are about 100,000 and 6.6, respectively. The giant dielectric constants of the BST/YIP composites are mainly attributed to the Maxwell–Wagner polarization.

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1. Introduction

With the rapid development of scientific technology, mirocomputer, radio communication technology and internet technology, the requirements of miniaturization, integration and intelligentization for the electronic devices become more and more stringent which needs the development of new materials or the enhancement of the properties of the existing materials [1]. As driven by the impetus of smaller and smaller feature sizes of devices in microelectronics, materials with high performance and multifunctional properties are always attracting the attention of researchers [2]. Dielectric materials with large dielectric constants have played a significant role in miniaturizing the size of modern electronic devices [3]. Recently, some common dielectric materials with high dielectric constants, such as lead zirconate titanate (PZT), cubic perovskite-related compound CaCu₃Ti₄O₁₂ [4-6] and BaTiO₃ have been widely studied. However, these single-phase compounds are lack of multifunctional

properties. Meanwhile, the magnetoelectric composite showing both high dielectric and magnetic properties have recently attracted increasing interest because of their significant technological promise in novel multifunctional devices [7,8]. These composite materials have been identified to be an enable solution to fabricate miniaturized filters and antennas, electromagnetic interference devices, and so on [9].

Barium strontium titanate (BST), an ABO₃-type perovskite ferroelectric oxide, which is a solid solution of barium titanate (BaTiO₃) and strontium titanate (SrTiO₃), is a well known dielectric and ferroelectric material [10– 12]. Consequently, using (Ba_{1-x}Sr_x)TiO₃ as a host material can be expected to develop new magnetoelectric composite materials with higher performance. On the other hand, as a typical orthoferrite, YFeO₃ is a G-type antiferromagnet with a high Néel temperature (T_N =645 K) and a high resistivity [13]. YFeO₃ single crystal shows weak magnetic and the magnetic properties are also expected to be effectively modified by doping rare earth element. The significance for applications in magnetic field sensors and optical devices in the fields of ultrafast optical switches, light spot position measurements and magneto-optical

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current sensor has been reported in the previous work [14–16].

In this paper, we synthesized a new kind of magneto-electric $Ba_{0.8}Sr_{0.2}TiO_3/YFeO_3$ (BST/YIP) composites by the in-situ growth solid-state method. And the dielectric and magnetic properties of the composites were investigated.

2. Experimental

The BST/YIP composites were prepared by the in-situ growth solid-state method. Commercial BaCO₃, SrCO₃ and TiO₂ were weighed, mixed thoroughly and calcined at 1150 °C in air for 2 h to prepare Ba_{0.8}Sr_{0.2}TiO₃ powder. Meanwhile, Y₃Fe₅O₁₂ (YIG) was prepared from reagent-grade Fe₂O₃ and Y₂O₃, which were also weighed, mixed thoroughly and calcined at 1250 °C for 6 h. The above calcined BST and YIG powders were ball milled into micrometer-size powders. The purities of the above starting materials are all higher than 99%. According to different mass ratios, BST and YIG powders were mixed first, and then the obtained final powder mixtures were pressed into disks after adding some aqueous 5 wt% PVA solution. Final sintering was carried out at 1350 °C in air for 2 h.

The phase composition of the BST/YIP composites was analyzed using an x-ray diffractometer (D/max2000PC, Rigaku, Tokyo, Japan). A scanning electron microscopy (JSM-6460, JEOL JSM-6460, Tokyo, Japan) was employed to observe the microstructure of the BST/YIP composites. The dielectric properties of the composites were measured by an impedance analyzer (E4980A, Agilent, Palo Alto, CA). The magnetic hysteresis loops of the composites were measured by a vibrating sample magnetometer 113 (Lake Shore 7410, Westerville, OH).

3. Results and discussion

The XRD patterns of the composites with different BST concentrations are shown in Fig. 1. It is striking that the parent YIG phase cannot be detected in all composites. The main crystalline phases of all the sintered samples can be indexed to be tetragonal perovskite BST phase and orthorhombic perovskite YIP phase (JCPDS card NO. 39-1489). It is worth noting that there is a small amount of unknown phase in all composites. As expected, with increasing the YIG concentration, the diffraction peaks of YIP phase become strengthened gradually.

Fig. 2 shows the backscattered SEM micrographs of natural surfaces of the BST/YIP composites with different BST concentrations. Homogeneous microstructures with few pores can be observed in all the composites. The coexistence of two major phases, i.e. ball-like BST grains (bright) and long-rod YIP grains (dark), can be also observed, which is well consistent with the above XRD analysis results.

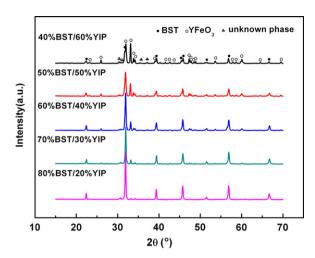


Fig. 1. XRD patterns of the BST/YIP composites with different BST concentrations.

Fig. 3 shows the SEM micrograph and EDS result of the 40% BST/60% YIP composite. It can be found that the white grains are composed of Ba, Ti, Sr, O and small amounts of Fe, C, which confirms that the bright grain is perovskite BST phase. The dark grains are composed of Y, Fe, O and small amounts of Ti. The atom ratio of Y to Fe is 1.09, which means that the dark grain is perovskite YIP phase. It can be deduced that some Ti⁴⁺ cations diffused from BST into YIG and made YIG transform to YIP.

Fig. 4 shows the frequency dependence of the dielectric properties of the BST/YIP composites with different BST concentrations. It can be seen that all the samples show similar dielectric behavior at room temperature. And the value of ε' decreases gradually with increasing frequency. The composites possess giant values of ε' in low frequency range. Giant values of ε' are mainly due to the presence of interfacial polarization (also named as Maxwell-Wagner polarization) at the interfaces of the two constituents of the composites [3]. When further increasing the frequency to a certain value all the sample show a Maxwell-Wagner relaxation, i.e., the ε' displays a step decrease at a certain frequency and at same time the $tan \delta$ shows a relaxation peak. According to the two-layer model of the Maxwell-Wagner interfacial polarization, the space charge polarization arises due to the inhomogeneous dielectric structure of the materials. Obviously the dielectric constant originated from the space charge polarization is proportional to the charge amount at the interfaces of the two phases. The space charge amount can be expressed as the following equation [17]:

$$Q = \frac{V\varepsilon_0(\gamma_1\varepsilon_2 - \gamma_2\varepsilon_1)}{\gamma_1d_2 + \gamma_2d_1}S\tag{1}$$

where Q is the charge amount at the interface of the two phases. V is the applied voltage. S is the area of the interfaces of the two phases. γ_1 , γ_2 , ε_1 , ε_2 , d_1 and d_2 are the conductivities, dielectric constants and thickness of the two

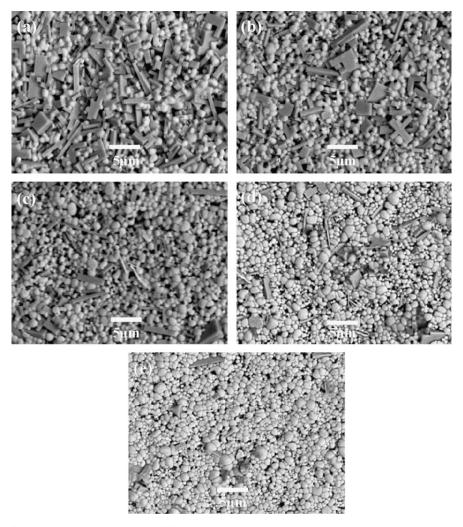


Fig. 2. SEM micrographs of the BST/YIP composites with different BST concentrations: (a) 40% BST/60% YIP, (b) 50% BST/50% YIP, (c) 60% BST/40% YIP, (d) 70% BST/30% YIP and (e) 80% BST/20% YIP.

phases, respectively. For the BST/YIP composites, the giant dielectric constant is influenced by the area of the interfaces of the two phases, which is dependent on the BST concentration and the density of the composite. It is well known that the dielectric constant is also directly dependent on the relative content and density of the material. Consequently, in low frequency range, the value of ε' decreases with increasing BST concentration.

The magnetic hysteresis loops of the BST/YIP composites with different BST concentrations are shown in Fig. 5. It is evident that the saturation magnetization $(M_{\rm S})$ decreases with increasing BST concentration, as expected, because this parameter depends on the total amount of magnetic material. The reduction of these values is mainly due to the dilution of magnetic component. The pure antiferromagnetic YFeO₃ displays a weak ferromagnetic characteristics. Because the alignment of Fe moments in YFeO₃ is not strictly antiparallel but slightly canted, a small net magnetization results give rise to a weak ferromagnetic behavior [18]. The saturation magnetizations of the composites are bigger than that of pure YFeO₃

prepared by the conventional solid-state reaction method [19] probably due to the fact that the doping of ${\rm Ti}^{4+}$ destroys the antiferromagnetic order and the remnant magnetization and the coercive field are 6.6 emu/g and 1.4 kOe, respectively, for the 40% BST/60% YIP composite.

4. Conclusions

The BST/YIP composites with giant dielectric constants and high saturation magnetizations were prepared through the in-situ growth solid-state method. Homogeneous microstructures with ball-like BST grains and long rod YIP grains were observed in the composites. It is the Maxwell–Wagner interfacial polarization that leads to the giant dielectric constant of the composites. The composites also possess higher saturation magnetization than pure YFeO₃ due to the doping of Ti⁴⁺. For the 40% BST/60% YIP composite, the remnant magnetization and the coercive field are 6.6 emu/g and 1.4 kOe, respectively.

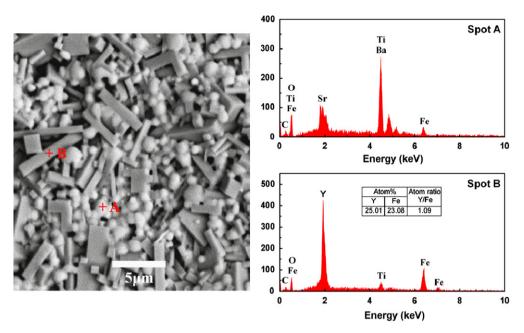


Fig. 3. SEM micrograph and EDS result of the 40% BST/60% YIP composite.

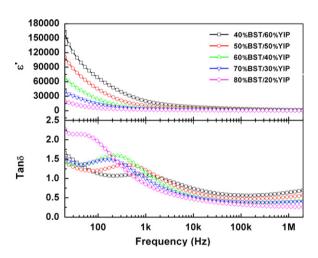


Fig. 4. Frequency dependence of the dielectric properties of the BST/YIP composites with different BST concentrations.

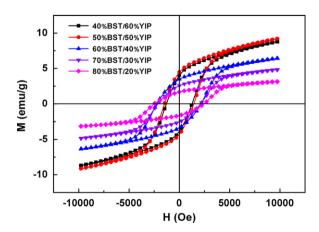


Fig. 5. Magnetic hysteresis loops of the BST/YIP composites with different BST concentrations.

Acknowledgments

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