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Structural, magnetic and microwave absorption characteristics of $BaCo_xMn_xTi_{2x}Fe_{12-4x}O_{19}$

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ABSTRACT

The effect of $Mn^{+2}Co^{+2}Ti^{+4}$ substitution on microwave absorption has been studied for $BaCo_xMn_xTi_{2x}Fe_{12-4x}O_{19}$ ferrite–acrylic resin composites, where x varies from 0.3 to 0.5 in steps of 0.1, in frequency range from 12 to 20 GHz. X-ray diffraction (XRD), scanning electron microscope (SEM), vibrating sample magnetometer, and vector network analyzer were used to analyze the structures, electromagnetic and microwave absorption properties. The results showed that, the magnetoplumbite structures for all samples have been formed. Based on microwave measurement on reflectivity, $BaCo_xMn_xTi_{2x}Fe_{12-4x}O_{19}$ may be a good candidate for electromagnetic compatibility and other practical applications at high frequency.

1. Introduction

The increase in electromagnetic pollution due to the rapid development of gigahertz (GHz) electronic systems and telecommunications has resulted in a growing and intense interest in electromagnetic-absorber technology. Electromagnetic interference (EMI) can cause severe interruption of electronically controlled systems. It can cause device malfunctions, generate false images, increase clutter on radar and reduce performance because of system-to-system coupling. These are some of the reasons why the use of self generated electromagnetic radiation apparatuses, which include cellular telephones, wireless computer and pagers, are strictly prohibited in certain areas, for example, in hospitals, banks, petrol stations and inside airplanes. To overcome the problems created by EMI, electromagnetic wave absorbers with the capability of absorbing unwanted electromagnetic signals are used, and research on their electromagnetic and absorption properties is still being carried out [1,2]. Recent developments in microwave absorber technology have been resulted in materials with high wave absorption coefficient, good physical performance and lower production cost [3,4]. There are a variety of absorber materials that can be used to suppress EMI depending on whether they are suitable for low and high frequency application [5–9]. As far as thickness and working frequency bandwidth are concerned, magnetic composites have obvious advantages. The magnetic fillers often used in such composites are ferrite materials, such as spinel ferrites and hexaferrites [10,11]. Hexaferrites with planar magnetic anisotropy are greatly used as electromagnetic wave absorbers in GHz range. Barium ferrite powders are ideal fillers for the development of electromagnetic attenuation materials at microwave, due to their low cost, low density, high stability, large electrical resistively, and high microwave magnetic loss [12–15]. Many works have been reported on barium ferrites for use as electromagnetic materials [16–20].

In our previous paper [17–19] the microwave attenuation properties have been studied different doped ferrites. Here we will report on the relationship between magnetic properties and microstructure for $BaCo_xMn_xTi_{2x}Fe_{12-4x}O_{19}$. The magnetic properties and microwave absorbing characteristics were investigated. The reason for choosing those substitution compounds is their different static magnetic properties (especially, the coercivity and saturation magnetization) at the critical substitution ratio for in-plane anisotropy. The predicated reflection loss demonstrates that $BaCo_xMn_xTi_{2x}Fe_{12-4x}O_{19}$ may be a good candidate for wave absorbing materials with low reflectivity at microwave frequency.

2. Experimental

2.1. The preparation of ferrite powders

For selection of the composition and stiochiometry of barium ferrite, the M-type barium ferrites with different composition were carried out with a conventional powder fabrication. The samples were synthesized from stoichiometric mixtures of Fe_2O_3, TiO_2, Co_3O_4, $BaCO_3$ and $MnCO_3$. Mixtures were crushed for 2 h and sintered in air at 1160 °C for 8 h. The heating rate of samples was from the room temperature to 600 °C with 6 °C min^{-1} and they were kept at 3 °C min^{-1} to the final sintering

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temperature. Finally the sintered ferrites were crushed again for 5 h to obtained fine powders with the particle size between 1 and 4 μm.

For the composition of $BaCo_xMn_xTi_{2x}Fe_{12-4x}O_{19}$ ($x = 0.3$, 0.4 and 0.5), the Fe^{+3} was partially replaced by Mn^{+2}, Co^{+2} and Ti^{+4}. Three samples of hexagonal ferrite powder namely, ferrite "A" of composition [$BaCo_{0.3}Mn_{0.3}Ti_{0.6}Fe_{10.8}O_{19}$], ferrite "B" of composition [$BaCo_{0.4}Mn_{0.4}Ti_{0.8}Fe_{10.4}O_{19}$] and ferrite "C" of composition [$BaCo_{0.5}Mn_{0.5}Ti_{1.0}Fe_{10}O_{19}$] were synthesized. Typical platelets have diameter to thickness ratios (D/t) ranging from 3 to 10. The composite specimens were prepared by mixing doped barium ferrites and acrylic resin powder with concentration of 70:30 by weight. Mixture of ferrite powders with acrylic resin were plasticized and fired at 220 °C and 5.5 Mpa. The pressed composites were in the form of cylindrical with the thickness of 2 mm and the diameter of 40 mm.

2.2. Measurement of properties

The identification of the crystalline phase was carried out on a X-ray powder diffractometer operating at 40 kV and using Cu Kα radiation. Scanning electron microscopy (SEM) examinations were performed using a PHILIPS XL 400. Specimens were coated by a thin gold layer by sputtering technique for SEM observation. Vibrating sample magnetometer was used to determine the hysteresis loops of ferrite samples at room temperature. Variation of the reflection loss in (dB) versus frequency in the range of 12–20 GHz has been investigated.

3. Results and discussion

3.1. Microstructure characteristics

The XRD patterns for the calcined powders of $BaCo_xMn_xTi_{2x}Fe_{12-4x}O_{19}$ are shown in Fig. 1. It is observed that the samples consist of the pure barium ferrite phase. The peaks for the doped barium ferrite appear at the same position as for the undoped ferrite, but with different intensities. In the doped ferrite cases, the dopants of Mn^{+2}, Co^{+2} and Ti^{+4} seem to be rearranged in the hexagonal structure to fulfill the formation of single hexagonal phase. It is generally recognized that the vacancy sites of partial deprivation of Ba^{+2}, Fe^{+3} and O^{-2} can be filled by these dopant ions.

Fig. 2 shows the microstructures of eroded surface of prepared ferrites. It is found that doping with small amount of Mn–Co–Ti does not significantly affect grain size and morphology. The grains are typical platelet morphology with grain size of about 3–6 μm. Some intergranular pores are present in all samples. Apparently, the Mn–Co–Ti doping does not contribute to the microstructure change. This indicates that the grain size is not the most likely cause of the enhanced coercivity observed in the doped samples.

3.2. Magnetic properties

The hysteresis loops of the as-synthesized barium ferrites are shown in Fig. 3. In general, substitutions lead to a decrease of H_c through the reduction of the magnetocrystalline anisotropy of the barium ferrite. It is observed that the undoped sample possess the largest coercive force (H_c), the largest hysteresis loop area and the highest B_r than those of other samples, while the former two factors may lead the larger hysteresis loss. The H_c of pure barium ferrite is very high (about 258.7 kA m^{-1}), which is due to strong uniaxial anisotropy along the *c*-axis of *M*-hexaferrite. On the other hand, Mn, Co and Ti substitution led to a rapid decrease of H_c from 258.7 kA m^{-1} ($x = 0$) to 16.3 kA m^{-1} ($x = 0.3$). The moderately low H_c indicates that the domain wall motion is the dominant magnetization mechanism and the sample is a soft magnetic material.

Cho and Kim have also reported similar results for Co–Ru substituted barium ferrite [20]. However, only linear decrease in coercivity was found in Co–Ti, Co–Sn and Co–Ir substituted barium ferrite [21–23]. It is observed that coercivity is dependent upon the grain size. Based on SEM images of our doped samples described

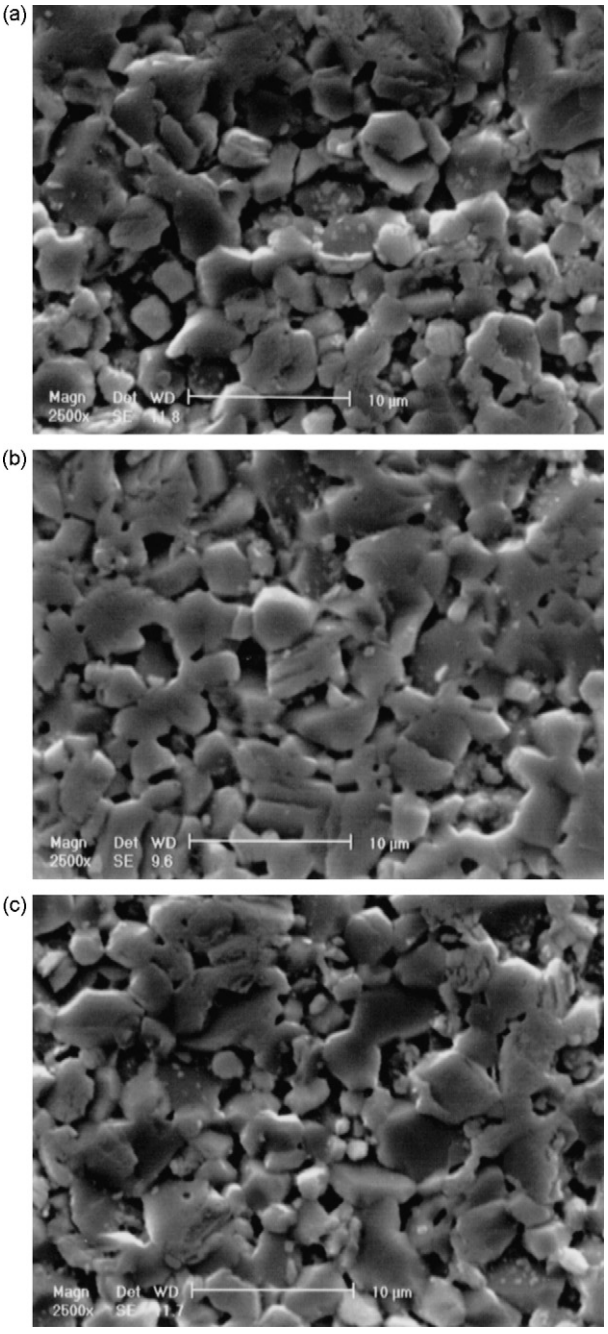

Fig. 2. SEM photographs of eroded surface of (a) ferrite "A", (b) ferrite "B" and (c) ferrite "C".

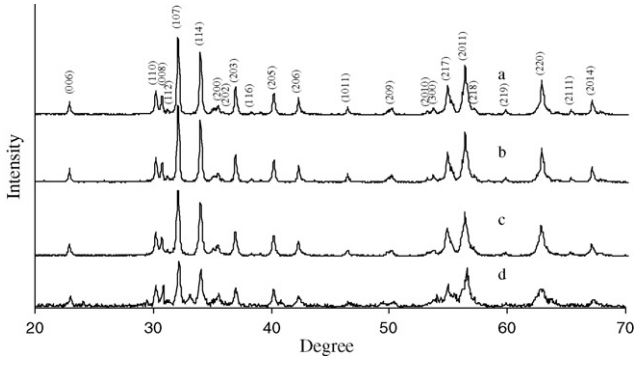

Fig. 1. XRD pattern of calcined powders at 1160 °C: (a) undoped ferrite, (b) ferrite "A", (c) ferrite "B" and (d) ferrite "C".