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Improvement of the performance of microwave X band absorbers based on pure and doped Ba-hexaferrite

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ABSTRACT

Optimum Fe/Ba mole ratio is determined in doped Ba-hexaferrite prepared via mechanical activation. X-ray diffractometer (XRD), scanning electron microscope (SEM), vibrating sample magnetometer (VSM) and vector network analyzer are used to analyze phases, structures, electromagnetic and microwave absorption properties. The mole ratio of Fe/Ba = 10 is detected to be optimum for doping and synthesizing the Ba-hexaferrite. In order to achieve high absorption in X band the ions of Zr^{4+}–Sn^{4+}–Ti^{4+}–M^{2+} ($M = Mg^{2+}$, Zn^{2+}, Cu^{2+}, Co^{2+}) are used as dopants. The results indicate the formation of single phase Ba-hexaferrite in either pure or doped compounds without any non-magnetic intermediate phases and with spherical and hexagonal morphologies respectively for the pure and doped ferrite. It is found out that $BaCo_2Zr(SnTi)_{0.5}Fe_8O_{19}$ compound has the maximum saturation magnetization (49.80 emu/g). Also the composite of $BaCo_2Zr(SnTi)_{0.5}Fe_8O_{19}$ 50 wt% in epoxy resin exhibits a minimum reflection loss of −29 dB at 12.2 GHz with 2.6 GHz bandwidth.

1. Introduction

In order to minimize electromagnetic wave interferences and their reflections, using advanced types of microwave absorbents is essential [1–3]. Among materials with microwave absorption capacity, ferrites with hexagonal and spinel structures are promising candidates. High electric conductivity of spinel ferrites results in reduction of their magnetic permeability at high frequencies because of eddy current loss. In contrast, hexagonal ferrites have larger intrinsic magnetic fields [4–7]. Hexaferrites such hexagonal barium ferrite (Ba-hexaferrite), exhibit unique properties such as high saturation magnetization, high uniaxial magnetic anisotropy, high Curie temperature and good chemical stability, that present them as useful materials for various applications in radar absorbing materials (RAMs), recording media, microwave devices, electrical machines, sensors and permanent magnets [8–11]. There are several methods to synthesize Ba-hexaferrite submicron particles, including chemical methods like sol–gel [12], hydrothermal [13], co-precipitation [14], microemulsion [15], sonochemical [16], and solid state methods like mechanical alloying or activation [17]. Synthesis method plays an important role in structural and magnetic properties of the material. Among the synthesis methods, mechanical activation is a simple and a low cost method in which synthesized powders exhibit unique properties because of the large specific area of grain boundaries and high volume fraction of atoms located at these boundaries [18–22]. One of the main drawbacks in the synthesis of Ba-hexaferrites is the simultaneous formation of non-magnetic intermediate phases like hematite (Fe_2O_3) and barium monoferrite ($BaFe_2O_4$). Therefore, high milling times (> 40 h) [23–25], high sintering temperatures (> 1373 K) [25–27] and high milling speeds (> 400 rpm) [28,29] are employed to solve this problem. One of the main parameters in the synthesis of barium hexaferrite is the Fe/Ba mole ratio. Taking the proper ratio, results in induction of lattice imperfections in the structure and increasing the kinetics of diffusion, and moreover reducing the heat treating temperature. Also, by using the optimum ratio it is possible to eliminate non-magnetic phases. The high anisotropy field (1.36 MA/m) and resonance frequency (47.6 GHz) of this compound makes it unusable as radar absorbent at the X-band [1,30–33]. It has been reported that by substitution of Fe^{3+} with Sn, Ti and Zr cations, the saturation magnetization increases and the coercive force decreases leading to enhancement of microwave absorption [34–41]. However, most of the studies on the microwave absorption of doped barium hexaferrite have been investigated on the K_u, K and K_a bands and there are few ones on the X-band [42–51]. Accordingly, the optimization of Fe/Ba mole ratio and dopant contents are highly effective strategies to tailor the magnetic and the microwave absorption properties of the final

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Table 1
Conditions used for the synthesis of pure and doped barium hexaferrite.

Sample	Ball to powder ratio (BPR)	Milling time (h)	Rotating speed (rpm)	Sintering temperature T (°C)	Sintering time t (h)
Pure Ba-hexaferrite	20:1	10	300	900	2
Doped Ba-hexaferrite	30:1	20	300	1000	5

material. In this work, the optimization of the mole ratio of Fe/Ba for the synthesis of pure phase barium hexaferrite by mechanical activation method has been investigated using XRD. Thereafter, the optimized hexaferrite has been substituted with tetravalent cations, namely Ti^{4+}, Sn^{4+} and Zr^{4+} (used as constant dopants) and four divalent cations of Zn^{2+}, Cu^{2+}, Co^{2+} and Mg^{2+} in various contents in order to achieve the highest X band microwave absorption.

2. Experimental

The raw materials that are used in this study consist of $BaCO_3$, Fe_2O_3, ZrO_2, SnO_2, TiO_2, MgO, ZnO, CuO and CoO. Pure and doped barium hexaferrite with the $BaFe_{12}O_{19}$ and $BaM_{2x}Zr_x(SnTi)_{x/2}Fe_{12-4x}O_{19}$ (M=Mg, Zn, Cu, Co) formula respectively are prepared using mechanical activation method under the conditions listed in Table 1. In this work, the planetary ball mill (Fritsch Pulverisette 5) is used, it contains two stainless steel cups and 11 steel balls with an average diameter of 16 mm in each cup. In all steps of the milling process, 1 wt% of ethanol was used as a process control agent (PCA). In order to remove Fe/Cr contamination after the milling process, the mixed powder was stirred in 30 mL solution

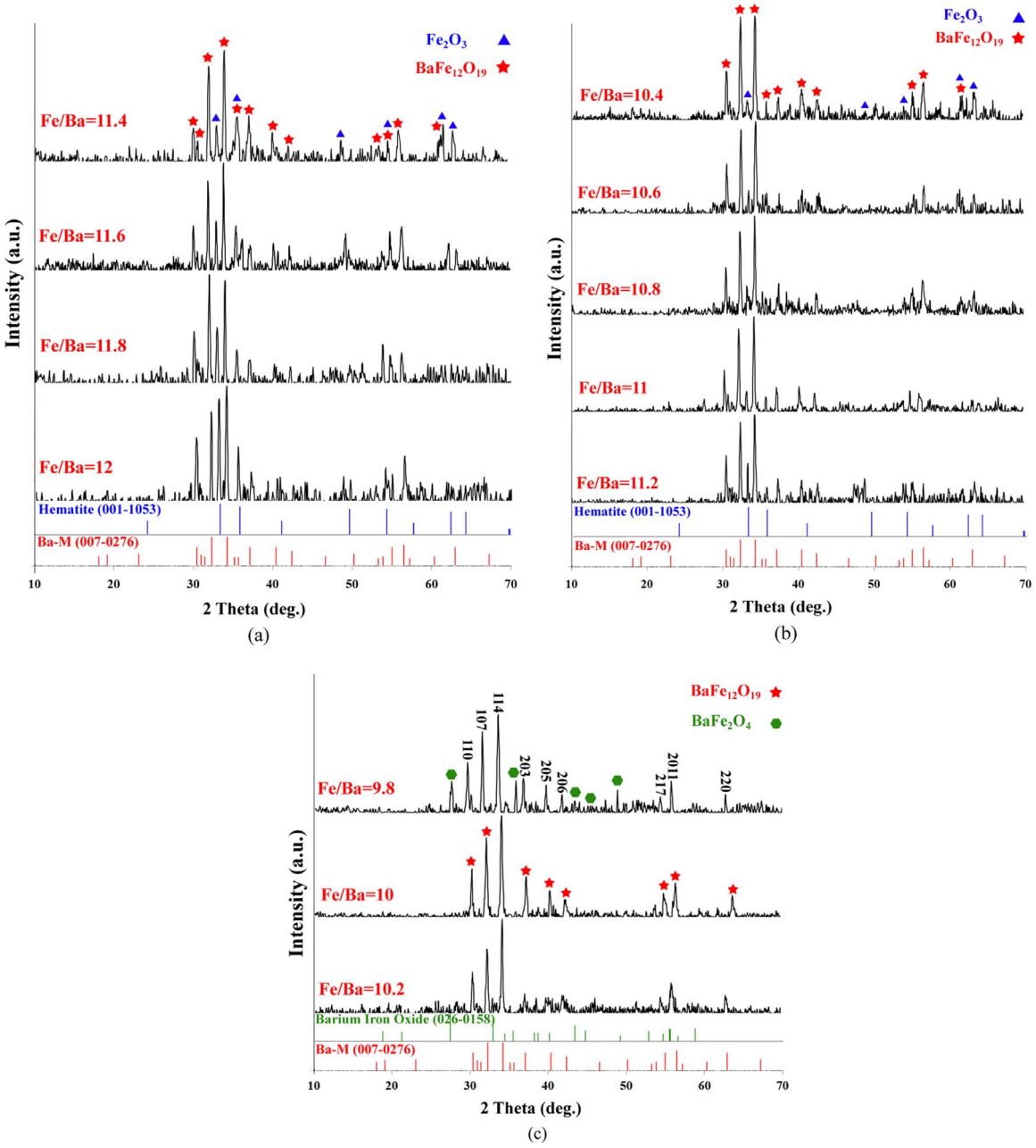

Fig. 1. Diffraction patterns of synthesized samples with the mole ratio of Fe/Ba in the range of, a) 12–11.4, b) 11.2–10.4 and c) 10.2–9.8.