



## Research articles

# Magneto-optical properties of $\text{BaCr}_y\text{Fe}_{12-y}\text{O}_{19}$ ( $0.0 \leq y \leq 1.0$ ) hexaferrites



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## ABSTRACT

In this study, nanocrystalline  $\text{BaCr}_y\text{Fe}_{12-y}\text{O}_{19}$  ( $0.0 \leq y \leq 1.0$ ) hexaferrite powders were prepared by sol-gel auto combustion method and the effect of  $\text{Cr}^{3+}$  ion substitution on morphology, structure, optic and magnetic properties of Barium hexaferrite were investigated. X-ray powder diffraction (XRD) analyses confirmed the purity of all samples. The XRD data shows that the average crystallite size lies between 60.95 nm and 50.10 nm and same was confirmed by Transmission electron microscopy. Transmission electron and scanning electron microscopy analyses presented the hexagonal morphology of all products. The characteristic hysteresis ( $\sigma$ - $H$ ) curves proved the ferromagnetic feature of as grown nanoparticle samples. Specific saturation magnetization ( $\sigma_s$ ) drops from 46.59 to 34.89 emu/g with increasing Cr content while the coercive field values lie between 770 and 1652 Oe. The large magnitude of the magnetocrystalline (intrinsic) anisotropy field, ( $H_a$ ) between 11.0 and 12.6 kOe proves that all products are magnetically hard. The energy band gap values decrease from 2.0 eV to 1.84 eV with increasing Cr content. From  $^{57}\text{Fe}$  Mössbauer spectroscopy, the variation in line width, isomer shift, quadrupole splitting and hyperfine magnetic field values were determined and discussed.

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## 1. Introduction

The technology is one of the defining elements in the world history and it has the potential to reform the human civilization [1]. Electromagnetic devices are perceived as the next important thing to date have been used in aerospace, medical, power generations and there are reaping benefits from the unique services it provides [2–5]. However, with fast technological developments and large use of electronic devices such as electromagnetic (EM) waves in wireless communication at higher frequencies, microwave filter, telecommunication etc. are creating serious problems with, related to EMI (electromagnetic interference) which have been attracting more attention over civilian and military circumferences [6]. These extensive growth devices are focused to the EM interference and which can be induced by output electric and magnetic field.

Therefore, scientists and researchers are focusing more on microwave absorbing materials [5,7–9].

According to Snoek's limit, the conventional spinel ferrites cannot be considered as an ideal candidate for the microwave absorber enhancement as they do not function well in the higher frequency (GHz) range due to the lower complex permeability ( $\mu_r$ ) [10]. Nevertheless, M-type hexaferrites materials possess those desired properties such as higher saturation magnetization, excellent thermal and chemical stability, adjustable anisotropy and corrosion resistance and greater coercivity, which makes use in the development of magnetic-optic and microwave absorber materials with a GHz frequency range [5,7,8]. M-type hexaferrites with general formula  $\text{MFe}_{12}\text{O}_{19}$  (M = Sr or Ba) are typically obtained by single or multiple cation substitutions for  $\text{Fe}^{3+}$  and or  $\text{M}^{2+}$  ions and produced through different synthesizing methods such as sol-gel auto-combustion, co-precipitated, ceramic, solid state, microwave assisted, and hydrothermal methods [8–12]. M-type hexaferrites are considered widely as important materials for recording media, permanent mag-

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nets, signal processing devices, telecommunication, super capacitor, microwave filters, magneto-optic media, and microwave devices etc. [9–16]. Since the beginning of 1950s,  $\text{BaFe}_{12}\text{O}_{19}$  hexaferrite has been a well-known microwave absorbing and magnetic material and researchers are developing the magnetic, microwave and dielectric properties of this hexaferrite using d-block elements such as Co, Ti, Ni, Cu, Mn, Ce, Al and Ti [15–18]. For example, Vadivelan et al. used copper to be substituted instead of  $\text{Fe}^{3+}$  ions of barium hexaferrite [19], Ti was substituted with  $\text{Fe}^{3+}$  ions of the single crystal  $\text{BaFe}_{12}\text{O}_{19}$  in a study conducted by Vinnik and co-workers [20], and a similar study by Ghzael et al. reported that Bi was substituted with  $\text{Fe}^{3+}$  ions of barium hexaferrite [18]. It was observed that the dopant elements and synthesis methods have a vital role in boosting magnetic, electric and microwave absorption properties of M-type hexaferrites. To further develop the magnetic properties of  $\text{BaFe}_{12}\text{O}_{19}$ , in this study Cr ions were doped with Ba ions in  $\text{BaFe}_{12}\text{O}_{19}$  hexaferrites. These novel magnetic materials,  $\text{BaCr}_y\text{Fe}_{12-y}\text{O}_{19}$  ( $0.0 \leq y \leq 1.0$ ) nanocrystalline hexaferrite powders, were synthesized by sol-gel auto-combustion method and their structures, magnetic properties were studied in detail.

## 2. Synthesis

### 2.1. Materials and instruments

Barium chloride  $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$  (99.9%), iron nitrate  $\text{Fe}(\text{NO}_3)_3 (\geq 96\%)$ , chromium (III) chloride hexahydrate  $\text{CrCl}_3 \cdot 6\text{H}_2\text{O} (\geq 98\%)$ , citric acid ( $\text{C}_6\text{H}_8\text{O}_7$ ) and ammonia solution ( $\text{NH}_3$ ) (25%) were purchased from Sigma-Aldrich. XRD measurements were conducted using Rigaku Benchtop Miniflex X-ray diffraction (XRD) with  $\text{Cu K}\alpha$  ( $20^\circ$ – $70^\circ$ ). Morphological analyses were done using HR-SEM, JEOL JSM- 6490 with EDX. Spectral analyses of samples were done via Bruker ATR-FT-IR spectrophotometer ( $400$ – $4000 \text{ cm}^{-1}$ ). Thermo Scientific, Evolution 300 PC model spectrophotometer (Praying Mantis diffuse reflectance accessory) was used for percent diffuse reflectance (DR%) measurements. The room temperature magnetization analyses were done via vibrating sample magnetometer (VSM) (10 kOe).

### 2.2. Method of preparation

Chemical sol-gel combustion route was employed for the synthesis of  $\text{BaCr}_y\text{Fe}_{12-y}\text{O}_{19}$  ( $0.0 \leq y \leq 1.0$ ) nanocrystalline hexaferrite powders. The specific amounts of  $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$ ,  $\text{Fe}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$  and  $\text{CrCl}_3$  were dissolved in deionized  $\text{H}_2\text{O}$ . The molar ratio of Fe: Ba was maintained at 10, while that of nitrates to citrate was 1:1. The pH value of the solution was fixed to 7.5 via  $\text{NH}_3$  solution (25 wt%). The solution was heated at  $85^\circ\text{C}$  to drive off the ammonia resulting in a colloidal sol which dried and transformed to a gel. The temperature was then adjusted to  $300^\circ\text{C}$ , at that point self-propagating decomposition takes place, a light-weight mass swelled up and ignited in a violent exothermic reaction that propagates through the entire sample in about 20 s, as the citric acid polymerized and evolved  $\text{CO}_2$ , while the cations were completely converted  $\alpha\text{-Fe}_2\text{O}_3$  and  $\text{BaCO}_3$  and  $\text{CrCO}_3$ . The driving force of this thrilling exothermic redox reaction was the combustion of  $\text{NH}_4\text{NO}_3$

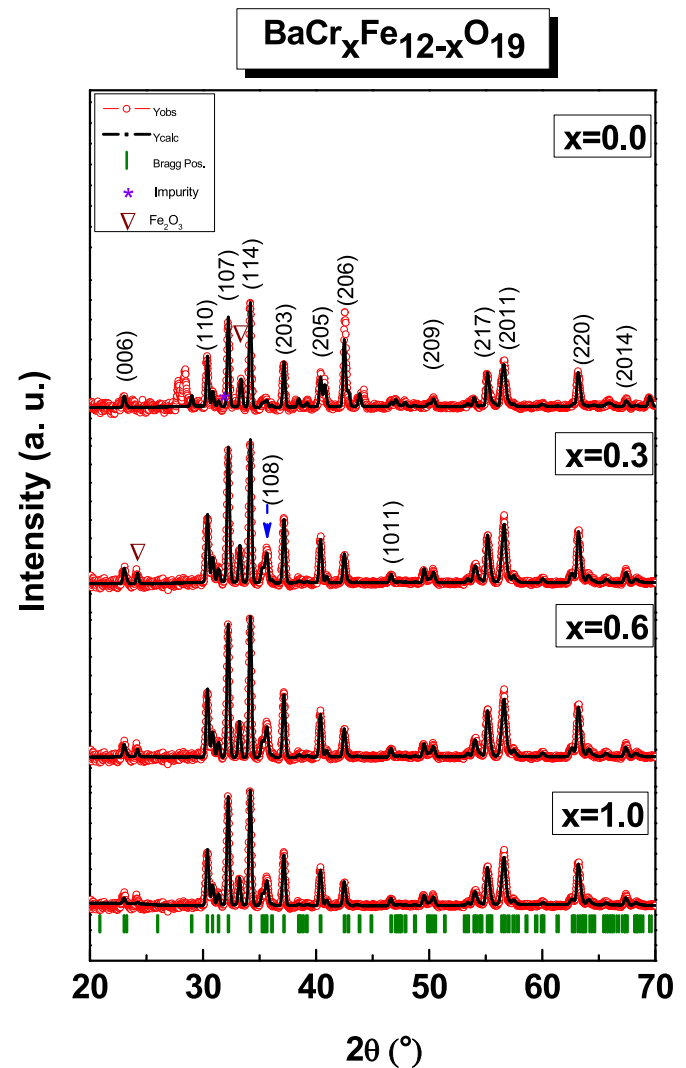


Fig. 1. XRD patterns of  $\text{BaCr}_y\text{Fe}_{12-y}\text{O}_{19}$  ( $0.0 \leq y \leq 1.0$ ) nanocrystalline hexaferrite powders.

formed from neutralization reaction between ammonia and citric acid. Final solid product was calcinated at  $1300^\circ\text{C}$  for 2 h.

## 3. Results and discussion

### 3.1. Structural investigation

The XRD patterns of  $\text{BaCr}_y\text{Fe}_{12-y}\text{O}_{19}$  ( $0.0 \leq y \leq 1.0$ ) nanocrystalline hexaferrite powders are displayed in Fig. 1. The hkl values of the observed peak in Fig. 1 are as follows (1 1 0), (1 0 7), (1 1 4), (1 0 8), (2 0 3), (2 0 5), (2 0 6), (1011), (2 0 9), (2 1 7), (2011), (2 2 0) and (2014) matched well with the barium hexaferrite ICDD Card no: 00-043-0002 confirming M-type barium ferrite structure. Further, trace amounts of hematite ( $\alpha\text{-Fe}_2\text{O}_3$ ) (ICDD PDF#33-0664)

Table 1  
Refined structural parameters for  $\text{BaCr}_y\text{Fe}_{12-y}\text{O}_{19}$  ( $0.0 \leq y \leq 1.0$ ) nanocrystalline hexaferrite powders.

y	a = b(Å)	c (Å)	c/a	V(Å) <sup>3</sup>	d <sub>xrd</sub> ± 0.05 (nm)	χ <sup>2</sup> (chi <sup>2</sup> )	R <sub>Bragg</sub>
0.0	5.884(0)	23.181(2)	3.9397	695.04	61.95	2.86	9.83
0.3	5.884(1)	23.188(6)	3.9409	695.28	55.86	0.85	4.89
0.6	5.884(8)	23.193(8)	3.9413	695.60	52.89	0.83	4.79
1.0	5.884(9)	23.195(0)	3.9414	695.68	50.10	1.20	4.93

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