



## Research articles

## Magnetic properties of Ce doped M-type strontium hexaferrites synthesized by ceramic route



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## ABSTRACT

This paper highlights the experimental study of  $\text{Ce}^{3+}$  ions doped M-type hexaferrite  $\text{Sr}_{1-x}\text{Ce}_x\text{Fe}_{12}\text{O}_{19}$  ( $0.00 \leq x \leq 0.30$ ) prepared by adopting the conventional ceramic process. The M-type hexagonal structure is confirmed in the X-ray diffraction measurements. There were two phases recognized for the magnetic powders with varying Ce content, one in-between  $x = 0.00$  to  $0.20$  as a single magneto plumbite segment and the other at  $x \geq 0.20$  as a second phase or the so called  $\alpha\text{-Fe}_2\text{O}_3$  phase. The micrographs of the sintered magnets depicted hexagonal crystal shapes. For magnetic properties a systematic study was done on the room temperature B–H hysteresis measurements. The estimated remanence value first increases with  $x$  in the range of  $x = 0.00$  to  $0.15$  and then showed a decrease at  $x \geq 0.15$ . Besides, the intrinsic coercivity ( $H_{c1}$ ) and magnetic induction coercivity ( $H_{cb}$ ) show decreasing-increasing character while maximum energy product ( $\text{BH}_{\text{max}}$ ) and ratio  $H_k/H_{c1}$  demonstrate increasing-decreasing character in the Ce doping range  $x$  of  $x \leq 0.15$  and  $x > 0.15$  respectively.

## 1. Introduction

In last decade, strontium and barium hexagonal ferrites persist to attract the attention of researchers and scientists in various fields of science and technology [1–9]. The M-type hexagonal ferrites (MHF) fall in the category of magnetic materials (ceramic) and importantly participated in several hi-tech and manufacturing fields owing to their low cost, excellent chemical strength, great uniaxial magneto crystalline anisotropy and last but not the least simple production routes [10–12]. Besides, MHF have been widely applied in the form of permanent magnets, for instance information storage devices, loudspeakers, home appliances, and motors etc., to name a few [13]. In addition, recently there is a vast interest in MHF, as some of them belong to multiferroic materials even at room temperature and have large magneto-electric influence, which is associated with the presence of spinoidal and spiral magnetic structures [14–16]. In several MHF, where ferroelectric and ferromagnetic segments co-exist, one can control these segments by applying either an external electric field or magnetic field. Nevertheless, now a day's, M-type MHF attracts more attention as a

multiferroic i.e. a materials that demonstrates a significant association between its magnetic and dielectric properties [17–19]. All the above materials used in spintronics, which is novel in the field of microelectronics [20–23]. However, at room temperature the ferroelectric state can be achieved by chemical substitutions or by magnificent local strains. Consequently, in solid solutions the concentration of  $d^5$  cations in iron crystallographic positions decreases diamagnetically substituted and dipole-dipole ordering becomes possible at prominent temperatures. In electric field at 110 kV/m, Al-aggregation  $\text{BaFe}_{12-x}\text{Al}_x\text{O}_{19}$  ( $x \leq 1.2$ ) hexaferrites the electric polarization of  $0.6 \mu\text{C}/\text{cm}^2$  and strong coupling between magnetic and electric subsystems at normal temperature were found [24,25]. Moreover, the diamagnetic replacement can basis a canted magnetic structure, which also prop ups the stabilization of the ferroelectric state [26,27]. In literature one can find MHF synthesized by several techniques, including sol-gel and sol-gel auto combustion [13,14], citrate precursor process [15,16], Pechini process [28], solution combustion route [29], modified chemical coprecipitation method [30] hydrothermal method [2,3,31,32] and the ceramic process [12,33]. Among all above routes the ceramic method is

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frequently used to prepare magnetic oxides which require multiple annealing and sintering steps [34]. It is important to recall that in MHF type structure,  $\text{Fe}^{3+}$  ions utilize five opposing sites, i.e., three spin up sites (2a, 2b and 12k) and two spins down sites ( $4f_1$  and  $4f_2$ ). Here 2a, 12k and  $4f_2$  are octahedral sites,  $4f_1$  is tetrahedral site and 2b is bi-pyramidal site [35]. Furthermore, the magnetic moment as well as magneto-crystalline structure appear from the electronic anisotropy of  $\text{Fe}^{3+}$  ions at the above five opposing sites [36]. In order to fulfill various needs in magnetic storing devices, many cations ( $\text{La}^{3+}$ ,  $\text{Pr}^{3+}$ ,  $\text{Sm}^{3+}$ ,  $\text{Nd}^{3+}$ ,  $\text{Co}^{2+}$ ,  $\text{Ti}^{4+}$ ,  $\text{Mn}^{2+}$  etc.,) and cations combinations ( $\text{La}^{3+}$ - $\text{Co}^{2+}$ ,  $\text{La}^{3+}$ - $\text{Zn}^{2+}$ ,  $\text{La}^{3+}$ - $\text{Cu}^{2+}$ ,  $\text{La}^{3+}$ - $\text{Bi}^{3+}$  etc.,) have been tried. A survey of literature for high density perpendicular recording media suggest that in  $\text{La}^{3+}$  substituted strontium hexaferrite nanoparticles prepared by citrate precursor method show increase in coercivity value by increasing  $\text{La}^{3+}$  composition and has high value at  $x = 0.63$  [37–40]. Recently, structural and magnetic properties of M type Ce substituted barium hexaferrites prepared by Sol-gel auto combination method were studied [13]. Microwave absorption properties of Ce-substituted barium ferrite prepared by the citrate sol-gel method were also investigated recently [15]. A study regarding improvement in the magnetic properties of strontium hexaferrite via Al doping is discussed very recently by Wang et al. [41]. Effect on structural and magnetic properties due to replacement of  $\text{Sr}^{2+}$  ions by rare-earth ions ( $\text{RE}^{3+}$ ) ions (RE: Y, La, Ce, Pr, Nd, Sm, and Gd) have been investigated for the case of Sr ferrites [42]. A systematic study on structural and magnetic properties of M-type strontium hexaferrite particles, prepared by a modified chemical coprecipitation method is done by Zi et al. [30]. There are also reports about the compound  $\text{Sr}_{1-x}\text{La}_x\text{Fe}_{12-x}\text{Cu}_x\text{O}_{19}$  in the doping range of ( $0.0 \leq x \leq 0.35$ ) prepared via the standard ceramic route revealed that the coercivity decrease linearly by increasing  $x$  (i.e.,  $0.0 \leq x \leq 0.35$ ) just like work of Yang et al. [43,44]. The strontium based MHF containing  $\text{La}^{3+}$  and  $\text{Bi}^{3+}$  with the formula  $\text{Sr}_{1-x}\text{La}_x\text{Fe}_{12-x}\text{Bi}_x\text{O}_{19}$  ( $0.0 \leq x \leq 0.5$ ) prepared by sol-gel auto combustion technique have also been studied in recent times [45]. Moreover, study of Co-Zr substituted strontium hexagonal ferrites  $\text{SrCo}_x\text{Zr}_x\text{Fe}_{12-x}\text{O}_{19}$  with ( $0.0 \leq x \leq 1.0$ ) produced by sol-gel process also showed some intriguing properties [46–48]. The presence of magnetically ordered states and ferroelectric properties open new opportunity for wide practical application of the MHF. The key point for such applications is controlling the magnetic properties of such specimens. The analysis of experimental data of multicomponent oxides so as the hexaferrites confirmed that their physical properties are directly depended on concentration of diamagnetic substitute, type of crystal structure, crystalline size [27] and even confirmed the anion stoichiometry [25]. In this paper we expose the study of  $\text{Ce}^{3+}$  ions doped Sr

based MHF having following formula  $\text{Sr}_{1-x}\text{Ce}_x\text{O}_6\text{Fe}_2\text{O}_3$  or  $\text{Sr}_{1-x}\text{Ce}_x\text{Fe}_{12}\text{O}_{19}$  (from here onward we will call it SCFO) in the form of powders and magnets, within the doping range  $0.00 \leq x \leq 0.30$  prepared by the conventional solid state ceramic process. The main aim behind this work is to probe composition effects of  $\text{Ce}^{3+}$  ions on the magnetic and microstructure related properties of SCFO magnets.

## 2. Materials and methods

### 2.1. Materials

We highlight that seven samples of MHF doped with  $\text{Ce}^{3+}$  ions having following formula  $\text{Sr}_{1-x}\text{Ce}_x\text{Fe}_{12}\text{O}_{19}$  (SCFO) with  $0.00 \leq x \leq 0.30$ , were prepared in succession by the standard ceramic process by choosing raw materials of AR grades namely  $\text{SrCO}_3$  (99 wt %),  $\text{Ce}_2\text{O}_3$  (99 wt%) and  $\text{Fe}_2\text{O}_3$  (98 wt%).

### 2.2. Methods

In order to achieve required mixtures, a planetary ball mill was used with hardened steel balls containing 10 mm average diameter and having a speed of 310 rpm. Briefly, as a first step, mixed powders prepared by the wet mixing method presintered in air, inside a box furnace at about 1280 °C for 2 h. A vibration mill was used to crush down the particles less than 100  $\mu\text{m}$ , and then sent to a ball mill where they wet milled with appropriate additives (0.5% wt. of  $\text{CaCO}_3$ , 0.3% wt. of  $\text{SiO}_2$ , 0.2% wt. of  $\text{Al}_2\text{O}_3$  and 0.3% wt. of  $\text{H}_3\text{BO}_3$ ) for about 14 h. This final milled product was trampled into disc-shaped pellets having roughly 15 mm thickness and 30 mm diameter under 310 MPa in a pulsed magnetic field aligned parallel to the pressing direction and having value of 900 KA/m. For the second step, the green compacts were sintered again in air at the temperature 1220 °C for 1.5 h inside the box furnace. The structural information regarding phase components of the presintered powder samples were carried out by an analytical diffractometer (X'Pert Pro) in continuous mode using  $\text{Cu K}\alpha$  radiation having  $\lambda = 1.5406 \text{ \AA}$ . In order to investigate morphologies, a field emission scanning electron microscopy FE-SEM (HITACHI S-4800) was used. Magnetic properties were carried out at room temperature by using a permanent magnetic measuring system (NIM-2000HF, made by the National Institute of Metrology of China).

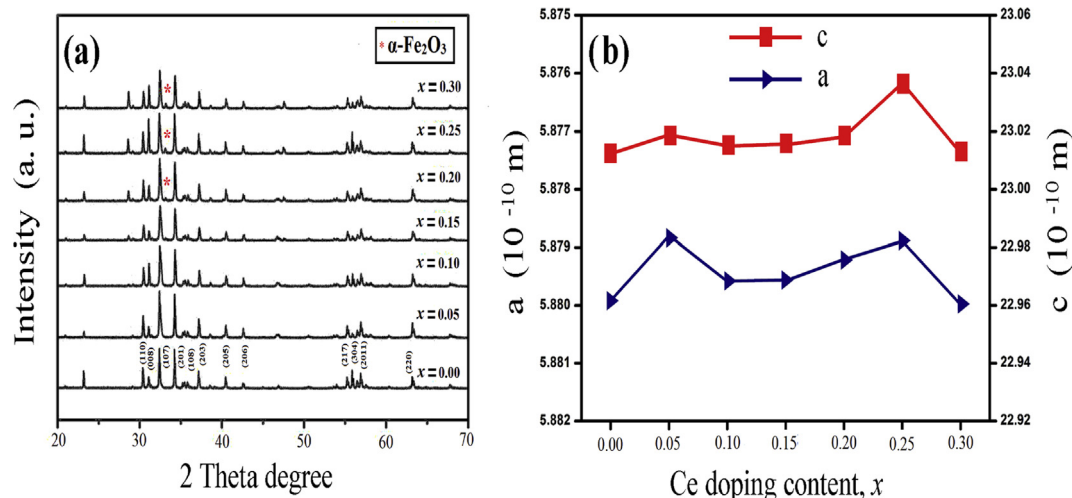
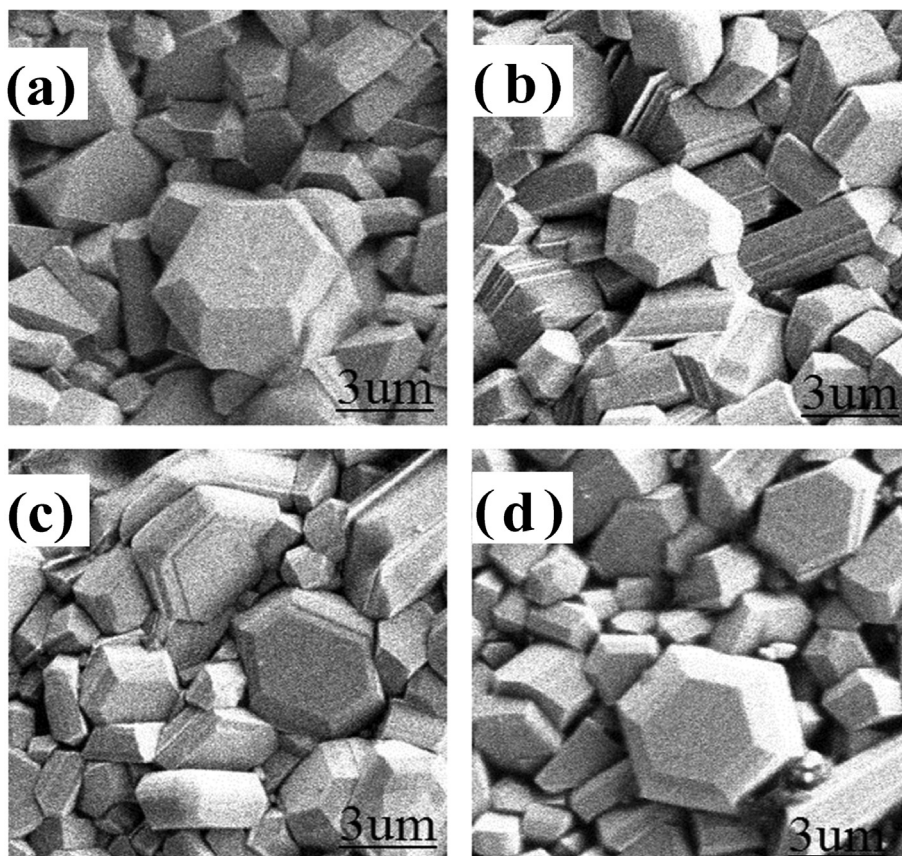


Fig. 1. (a) XRD pattern for the MHF powders of SCFO ( $0.00 \leq x \leq 0.30$ ), (b) Plot of lattice parameters  $a$  and  $c$  versus  $x$  of SCFO ( $0.00 \leq x \leq 0.30$ ).



**Fig. 2.** FE-SEM images of the SCFO ( $0.00 \leq x \leq 0.30$ ) magnets sintered at  $1220^\circ\text{C}$  for 1.5 h (in air) with Ce composition ( $x$ ) of: (a)  $x = 0.00$ , (b)  $x = 0.05$ , (c)  $x = 0.10$ , and (d)  $x = 0.15$ .

### 3. Results and discussion

#### 3.1. Phase identification analysis

The structural analysis of MHF powders of SCFO ( $0.00 \leq x \leq 0.30$ ) acquired by X-ray diffraction (XRD) measurements is depicted in Fig. 1. The diffraction peaks are indexed by reason of hexagonal magneto plumbite crystal anisotropy having space group  $P6_3/mmc$  (JCPDS card no.33-1340).

It is evident from Fig. 1(a) that all precursors have almost same XRD pattern that point toward a single magneto plumbite structure and no secondary phase's sign is seen with the increase of Ce doping content  $x$  from 0.00 to 0.15 due to entering various cations in the crystal structure of hexaferrites. This suggests that  $\text{Ce}^{3+}$  ions have gone into the M-type lattice. With  $x \geq 0.20$ , the XRD patterns show a desired phase of the strontium MHF and also an undesired phase or roughly speaking as a second hematite phase in the form of  $\alpha\text{-Fe}_2\text{O}_3$  with space group (R-3c). The occurrence of this sort of phase at  $x \geq 0.20$ , suggests that not all  $\text{Ce}^{3+}$  ions have entered in the MHF lattice of SCFO. In addition, it is evident that the peak intensities of XRD patterns changes by increasing  $x$ . The variation could be thought as due to occupying of the crystallographic sites in the crystal lattice. The strongest peak corresponds to (1 0 7), for the SCFO magnetic powders with  $x$  range of 0.00–0.30. In Fig. 1(b) we present results for the variant of the lattice constants  $a$  and  $c$  with different Ce doping content ( $x$ ) for the powders of SCFO ( $0.00 \leq x \leq 0.30$ ). These values of lattice constants  $a$  and  $c$  as a function of  $x$  are estimated from the values of the interplanar distances  $d_{hkl}$  given by the following formula,

$$d_{hkl} = \left( \frac{4}{3} \cdot \frac{h^2 + hk + k^2}{a^2} + \frac{l^2}{c^2} \right)^{-\frac{1}{2}} \quad (1)$$

where  $h$ ,  $k$  and  $l$  are the Miller indices and  $d_{hkl}$  is the inter-planer spacing.

As already mentioned, the obtained values for the respective lattice constants  $a$  and  $c$  are plotted in Fig. 1(b), show variation with the increase of Ce content ( $x$ ). Fig. 1(b) shows that the values of lattice parameters  $c$  and  $a$  increase with increasing  $\text{Ce}^{3+}$  aggregation  $x$  for the powders of SCFO ( $0.00 \leq x \leq 0.30$ ). The increase in lattice parameters  $c$  and  $a$  is ascending from the point that the average ionic radii ( $1.26 \text{ \AA}$ ) of  $\text{Sr}^{2+}$  and ( $1.143 \text{ \AA}$ ) of  $\text{Ce}^{3+}$  are higher as equated to the ionic radius ( $0.78 \text{ \AA}$ ) of  $\text{Fe}^{3+}$ . The variation of  $c/a$  (lattice parameter ratios) as a meaning of  $\text{Ce}^{3+}$  aggregation  $x$  for the MHF is confirmed in Fig. 3. It is obvious from Table 1 and Fig. 3 that ratios of  $c/a$  are slightly increasing with the increasing of ( $x$ ) aggregation of  $\text{Ce}^{3+}$  from 0.00 to 0.30. The  $c/a$  is exploited to confirm the structure type i.e for MHF structure can be assumed if the ratio is lower than 3.98. The  $c/a$  ratios are within the range of 3.910–3.916, specifying the MHF structure. It is obvious from Table 1 that with the variation of  $\text{Ce}^{3+}$ ,  $x$  aggregation the unit cell volume ( $V_{\text{cell}}$ ) increases from  $x$  aggregation 0.00–0.30 and  $686.026(\text{A}^0)^3$  to  $692.096(\text{A}^0)^3$ . The increase of  $V_{\text{cell}}$  with increasing  $\text{Ce}^{3+}$   $x$ -aggregation may be ascribed to the lattice parameters ( $c$  and  $a$ ) as shown in Table 1 and Fig. 3. Furthermore, the change in the lattice constant  $c$  is bigger in comparison with lattice constant  $a$ . It is quite evident that both parameters show more or less a similar trend with increasing  $x$  i.e., an increase-decrease region followed by a constant type region and then again an increase-decrease region. It is well established now that the lattice constants should increase if one substitutes higher ionic radii elements [40]. The refined values of lattice parameters of crystal structure with space group  $P6_3/mmc$  are listed in Table 1. The indexing and refinement of lattice parameters are confirmed by using software Checkcell (upgraded version of Celref) by using the least square method. The calculated crystallographic

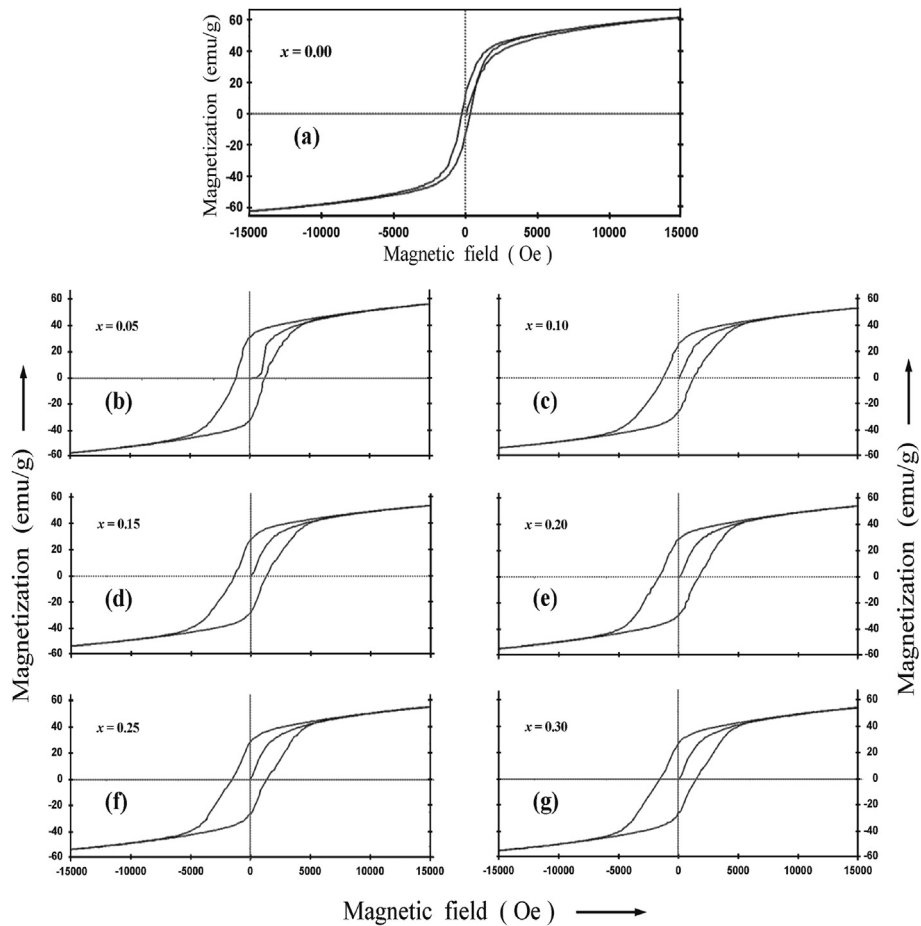


Fig. 3. Magnetization versus applied magnetic field of SCFO hexaferrite; (a)–(f) with different Ce doping content values ( $0.00 \leq x \leq 0.30$ ).

Table 1

Parameters from the structural refinement of  $Sr_{1-x}Ce_xFe_{12}O_{19}$  ( $x = 0.00, 0.05, 0.10, 0.15, 0.20, 0.25, 0.30$ ).

Lattice Parameters	$x = 0.00$	$x = 0.05$	$x = 0.10$	$x = 0.15$	$x = 0.15$	$x = 0.25$	$x = 0.30$
$a = b$	5.8780	5.8804	5.8850	5.8879	5.8897	5.8901	5.8909
$c$ ( $\text{Å}$ )	22.9879	23.0071	23.0136	23.0372	23.0536	23.0680	23.0696
$c/a$	3.910	3.912	3.910	3.912	3.914	3.916	3.916
$\alpha = \beta$ ( $^\circ$ )	90	90	90	90	90	90	90
$\gamma$ ( $^\circ$ )	120	120	120	120	120	120	120
Volume ( $\text{Å}^3$ )	686.026	687.845	688.962	689.478	690.471	691.718	692.096

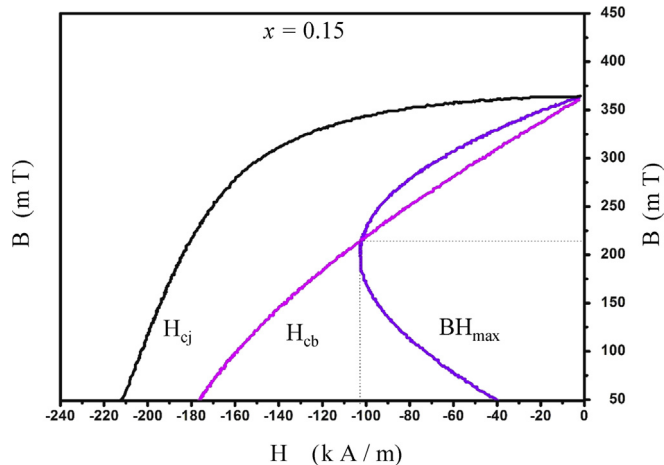


Fig. 4. The usual Demagnetizing curves of MHF magnets SCFO with Ce content  $x$ , at  $x = 0.15$ .

parameters show an increase and variations in lattice parameters with doping of  $Ce^{3+}$  ions which leads to the change in volume of doped sample.

We describe the SEM micrographs of the SCFO powders in Fig. 2((a)–(d)), in order to give an evident visualization about the homogeneous distribution and hexagonal plate like shape of the particles. In addition, it is observed that the Ce composition ( $x$ ) do not affect the average particle size and precursor morphology of the sintered magnets.

### 3.2. Magnetic properties

The magnetic properties of MHF in the form of hysteresis loops for SCFO magnetic powders with different Ce doping content,  $x$  ( $0.00 \leq x \leq 0.30$ ) are shown in Figs. 3 and 4 respectively. The demagnetizing curves (Fig. 3) of sintered magnets were recorded at room temperature for all the precursors by resorting to a permanent magnetic measuring system.

In Fig. 4 below, we describe a quick view of the typical

**Table 2**

Summary of  $B_r$ ,  $H_{cb}$ ,  $H_{cj}$  and  $(BH)_{max}$  values taken from the data of MHF magnets SCFO with different Ce contents,  $x$  ( $0.00 \leq x \leq 0.30$ ).

Ce content, $x$	$B_r$ (mT)	$H_{cb}$ (KA/m)	$H_{cj}$ (ka/m)	$(BH)_{max}$ (kJ/m <sup>3</sup> )
0.00	246	161.3	1710	5.37
0.05	267	150.9	272	12.2
0.10	309	85.9	1131	15.9
0.15	324	61.4	1206	18.0
0.20	226	67.4	1349	5.10
0.25	224	74.3	1574	3.82
0.30	150	170.6	1580	2.85

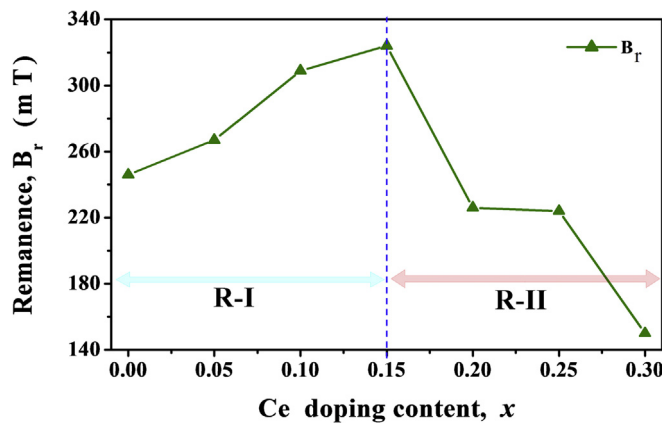


Fig. 5. The remanence ( $B_r$ ) versus Ce doping content ( $x$ ) plotted for the SCFO hexaferrites.

demagnetizing curves of SCFO magnets with constant value of Ce content  $x = 0.15$ .

Furthermore, the parameters determined from the obtained hysteresis loops for instance, the remanence ( $B_r$ ), intrinsic coercivity ( $H_{cj}$ ), magnetic induction coercivity ( $H_{cb}$ ) and maximum energy product ( $(BH)_{max}$ ) respectively are presented in Table 2.

In Fig. 5, we present data of SCFO hexaferrites in the form of remanence ( $B_r$ ) with varying Ce doping content ( $x$ ) in the range of ( $0.00 \leq x \leq 0.30$ ). The  $B_r$  show fairly significant deviation such that initially it increases in the region marked as R-I at  $x = 0.00$  (with  $B_r$  value  $\sim 246$  mT) to  $x = 0.15$  ( $B_r$  value  $\sim 324$  mT). After that it showed a decrease when  $x \geq 0.15$  (with  $B_r$  value  $\sim 324$  mT to 150 mT) marked as region R-II. In our view, the increasing trend in  $B_r$  value in the region R-I ( $0.0 \leq x \leq 0.15$ ) is ascribed to occupancy of different interstitial sites by the  $Fe^{3+}$  ions (as discussed in sec. 1) together with collinear

arrangement of its magnetic moments created by super exchange mechanism. Additionally,  $Ce^{3+}$  ions tend to reside on those sites that contain downward electrons spins in the low doping range, therefore the  $Fe^{3+}$  ions have to forcing upward spins and accordingly increasing the remanence. Alternatively, in  $Sr_{1-x}Ce_xFe_{12}O_{19}$  ( $0.00 \leq x \leq 0.30$ ), the replacement of trivalent of  $Ce^{3+}$  for divalent  $Sr^{2+}$  ions a small quantity of  $Fe^{3+}$  ions will change into  $Fe^{2+}$  ions at octahedral 2a sites to balance the electric charge. This decline the ferromagnetic  $Fe^{3+}-O^{2-}-Fe^{3+}$  super exchange interaction due to valence change [46,47]. Consequently, strong local spin-canting exist the overall magnetic structure remains ferrimagnetic up to  $x \geq 0.20$ . The results were gradual decrease in  $T_C$  by increase in  $Ce^{3+}$  substitution leads to the formation of a spin-canting structure [40,48]. Now let us shed some light on the shrinking of remanence ( $B_r$ ) in the region R-II with  $x > 0.15$ . The increase in doping content of  $Ce^{3+}$  ions together with regards to its non-magnetic character will tend to reduce the already present super exchange effect and hence the  $B_r$  value. Furthermore, in above Ce substitution range R-II (i.e.,  $x > 0.15$ ) the  $\alpha-Fe_2O_3$  phase (as a second phase) will start to show up growing character in its composition with respect to increment in  $x$  and consequently become a cause of further decreasing the  $B_r$  value. Fig. 6(a) describes the dual variation of the magnetic induction coercivity ( $H_{cb}$ ) together with the intrinsic coercivity ( $H_{cj}$ ) as a function of Ce doping content ( $x$ ).

First of all, clearly, observed that both  $H_{cb}$  and  $H_{cj}$  of the SCFO hexaferrites show a rapid decrease with increase in  $x$  values represented by R-I and R-II respectively. The  $H_{cb}$  decreases at  $x = 0.00$  (having  $H_{cb}$  value  $\sim 161.3$  KA/m) to  $x = 0.15$  (with  $H_{cb}$  value  $\sim 61.4$  KA/m). Then it showed an increase from  $x = 0.15$  onwards ( $H_{cb}$  value  $\sim 61.4$  KA/m) to  $x = 0.30$  (having  $H_{cb}$  value  $\sim 170.6$  KA/m). Similarly to above, the  $H_{cj}$  decreases at  $x = 0.00$  ( $H_{cj}$  value  $\sim 1710$  KA/m) to  $x = 0.05$  (having  $H_{cj}$  value  $\sim 272$  KA/m). After that it increases and showed almost a linear increasing character from  $x = 0.10$  ( $H_{cj}$  value  $\sim 1131$  KA/m) to  $x = 0.30$  (having  $H_{cj}$  value  $\sim 1580$  KA/m). In literature [49] one can find a theoretical formula relating inversely the coercivity ( $H_{cj}$ ) with the saturation magnetization ( $M_s$ ) given as under,

$$H_c = a \left( \frac{2K}{M_s} \right) \quad (1)$$

where  $a$ ,  $M_s$  and  $K$  are called, the orientation factor, saturation magnetization and magneto crystalline anisotropy constant respectively. The decrease of  $H_{cb}$  ( $x < 0.15$ ) and  $H_{cj}$  ( $x < 0.05$ ) may be seen from Eq. (1) and from Fig. 6(b) where  $B_r$  and  $H_{cj}$  are plotted together as a function of  $x$ . Besides, the increase of both  $H_{cb}$  and  $H_{cj}$  in the R-II having doping range  $x \geq 0.15$  might be ascribed to the slight decrease seen in the  $B_r$  value in the vicinity of above mentioned range (see Fig. 6(b)). Lastly, there may be an influence of magneto crystalline anisotropy in

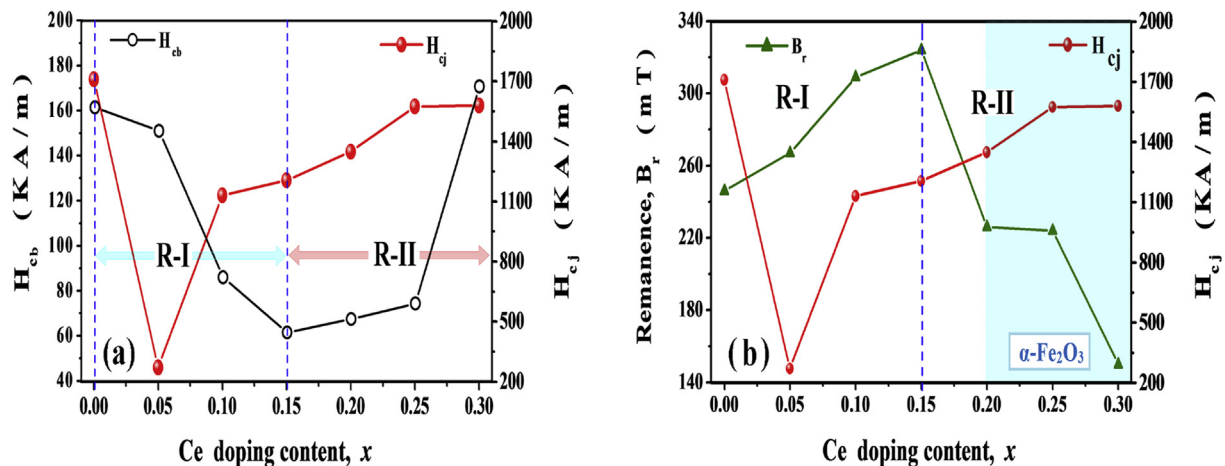


Fig. 6. The plot of  $H_{cb}$  (left vertical axis) and  $H_{cj}$  (right vertical axis) versus Ce doping content  $x$ , for the SCFO hexaferrites. R-I and R-II are regions of investigation.

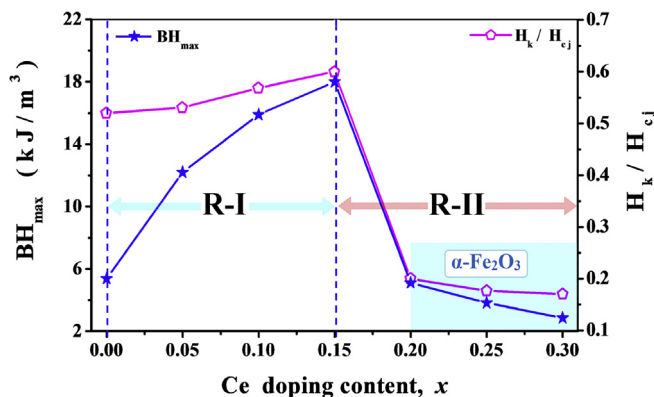


Fig. 7.  $BH_{\max}$  and  $H_k/H_{c_j}$  versus Ce doping content  $x$ , plotted on a dual vertical axis for the SCFO hexaferrites. R-I and R-II are two regions under investigation.

increasing the above parameters ( $H_{cb}$  and  $H_{c_j}$ ).

The plot of maximum energy product ( $BH_{\max}$ ) versus different Ce compositions  $x$  is shown in Fig. 7. First consider the left side axis of  $BH_{\max}$  value, that show an increasing trend in the region R-I with  $0.00 \leq x \leq 0.15$ , while there is a decrease in its value in the region R-II ( $x \geq 0.15$ ). Looking at Figs. 5 and 6 and also from the information given in Table 1, it is clear that the  $BH_{\max}$  has shown a decreasing trend in R-II region at  $x = 0.15$  (with  $BH_{\max} \sim 18.0 \text{ kJ/m}^3$ ) to  $x = 0.30$  (having  $BH_{\max} \sim 2.85 \text{ kJ/m}^3$ ). This can be attributed to the variations of the remanence ( $B_r$ ) and intrinsic coercivity ( $H_{c_j}$ ) as depicted in Fig. 6. It is well accepted that the  $BH_{\max}$  is predictable by the product of  $B_r$  and  $H_{c_j}$ , and is judged as maximum area (in the second quadrant) of the hysteresis loop. Therefore, above discussion confirmed that, the  $BH_{\max}$  has shown a decrease with respect to increase in Ce doping  $x$  for the case of SCFO. Now let us turn to the right hand side axis of above Fig. 7, where the  $H_k/H_{c_j}$  ratio of the SCFO ( $0.00 \leq x \leq 0.30$ ) magnets as a function of  $x$  is presented. Here  $H_k$  and  $H_{c_j}$  are the knee field and coercivity respectively. Actually,  $H_k/H_{c_j}$  is considered as a vital ratio in regard to magnetic properties of the permanent magnets. It can, by instinct, index the rectangularity of the demagnetizing curves as well as validate the stability of the magnets under dynamic working environment [50]. As seen from Fig. 7, in region R-I, the  $H_k/H_{c_j}$  ratio has shown an increase similar to  $BH_{\max}$  with increasing Ce content and in the region R-II it has shown a decrease in its value. The observed character of  $H_k/H_{c_j}$  ratio is accordance with the results revealed in Figs. 3 and 4.

#### 4. Conclusion

The M-type hexaferrites (MHF) doped with  $\text{Ce}^{3+}$  ions having the general formula  $\text{Sr}_{1-x}\text{Ce}_x\text{Fe}_{12}\text{O}_{19}$  (SCFO) with  $0.00 \leq x \leq 0.30$ , were successfully prepared in succession by the standard ceramic process. XRD data confirmed the phase compositions of the powder samples by highlighting two phases, first as a single magneto plumbite phase with increasing  $x$  upto  $x \leq 0.20$ , and second as  $\alpha\text{-Fe}_2\text{O}_3$  when  $x \geq 0.20$ . The microstructures also confirmed that the grain particles were of hexagonal platelet like shape. For investigation of magnetic properties, a permanent magnetic measuring machine was used to record the measurements at room temperature. The estimated remanence value ( $B_r$ ), showed an increase with  $x$  upto  $x = 0.15$  and then a decrease when  $x \geq 0.15$ . The other parameters like intrinsic coercivity ( $H_{c_j}$ ) and magnetic induction coercivity ( $H_{cb}$ ) showed a decreasing-increasing trend in the above regions of Ce doping. However, the maximum energy product ( $BH_{\max}$ ) and ratio  $H_k/H_{c_j}$  have displayed increasing-decreasing character in the regions of  $x \leq 0.15$  and  $x > 0.15$  respectively with respect to increasing Ce doping content. Finally, the MHF with Ce doping (like SCFO) with  $x \leq 0.15$  having high  $B_r$  and low  $H_c$  values, are of great interest for the future magnetic applications.

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#### References

- [1] J.P. Jakubovics, Magnetism and Magnetic Materials, 1st Ed., The Institute of Metals, London, 1987.
- [2] J.F. Wanga, C.B. Ponton, I.R. Harris, J. Magn. Magn. Mater. 234 (2001) 233–240.
- [3] J.F. Wang, C.B. Ponton, R. Grössinger, I.R. Harris, J. Alloy. Compd. 369 (2004) 170–177.
- [4] M.N. Akhtar, A.B. Sulong, M. Ahmad, M.A. Khan, A. Ali, M.U. Islam, J. Alloy. Compd. 660 (2016) 486–495.
- [5] S.V. Trukhanov, A.V. Trukhanov, V.G. Kostishin, L.V. Panina, I.S. Kazakevich, V.A. Turchenko, V.V. Kochervinskiy, JETP Lett. 103 (2016) 100–105.
- [6] K.M.U. Rehman, X. Liu, Y. Yang, et al., J. Magn. Magn. Mater. 449 (2018) 360–365.
- [7] S.V. Trukhanov, A.V. Trukhanov, et al., J. Magn. Magn. Mater. 426 (2017) 487–496.
- [8] S.V. Trukhanov, A.V. Trukhanov, V.A. Turchenko, V. An, E.L. Trukhanov, D.I. Trukhanova, V.M. Tishkevich, T.I. Ivanov, M. Zubar, V.G. Salem, L.V. Kostishyn, D.A. Panina, S.A. Gudkova, Vinnik, Ceram. Int. 44 (2018) 290–300.
- [9] S.V. Trukhanov, A.V. Trukhanov, V.A. Turchenko, V. An, D.I. Trukhanov, E.L. Tishkevich, T.I. Trukhanova, D.V. Zubar, V.G. Karpinsky, L.V. Kostishyn, D.A. Panina, S.A. Vinnik, E.A. Gudkova, P. Trofimov, A. Thakur, Y. Yang Thakur, J. Magn. Magn. Mater. 457 (2018) 83–96.
- [10] S.-W. Cheong, M. Mostovoy, Nat. Mater 6 (2007) 13.
- [11] M. Gich, I. Fina, A. Morelli, F. Sánchez, M. Alexe, J. Gàzquez, J. Fontcuberta, A. Roig, Adv. Mater. 26 (2014) 4645.
- [12] Y.J. Yang, X.S. Liu, D.L. Jin, Y.Q. Ma, J. Magn. Magn. Mater. 364 (2014) 11–17.
- [13] R.A. Pawar, S.S. Desai, Q.Y. Tamboli, Sagar E. Shirsath, S.M. Patange, J. Magn. Magn. Mater. 378 (2015) 59–63.
- [14] V.P. Singh, G. Kumar, A. Kumar, R.S. Rai, M.A. Valente, K.M. Batoo, R.K. Kotnala, M. Singh, Ceram. Int. 42 (2016) 5011–5017.
- [15] Sun Chang, Sun Kangning, Chui Pengfei, J. Magn. Magn. Mater. 324 (2012) 802–805.
- [16] K. Rana, P. Thakur, A. Thakur, M. Tomar, V. Gupta, J.L. Mattei, P. Queffelec, Ceram. Int. 42 (2016) 8413–8418.
- [17] A.V. Trukhanov, V.O. Turchenko, I.A. Bobrikov, S.V. Trukhanov, I.S. Kazakevich, A.M. Balagurov, J. Magn. Magn. Mater. 393 (2015) 253–259.
- [18] S.V. Trukhanov, A.V. Trukhanov, V.O. Turchenko, V.G. Kostishin, L.V. Panina, I.S. Kazakevich, A.M. Balagurov, J. Magn. Magn. Mater. 417 (2016) 130–136.
- [19] S.V. Trukhanov, A.V. Trukhanov, et al., J. Magn. Magn. Mater. 426 (2017) 554–562.
- [20] K.M.U. Rehman, et al., J. Supercond. Novel Magn. (2017) 1–8, <https://doi.org/10.1007/s10948-017-4272-5>.
- [21] K.M.U. Rehman, et al., Chin. J. Phys. 55 (2017) 1780–1786.
- [22] S.V. Trukhanov, Investigation of stability of ordered manganites, JETP 101 (2005) 513–520.
- [23] S.V. Trukhanov, A.V. Trukhanov, H. Szymczak, C.E. Botez, J. Low Temp. Phys. 149 (2007) 185–199.
- [24] S.V. Trukhanov, A.V. Trukhanov, A.N. Vasil'ev, A. Maignan, H. Szymczak, JETP Lett. 85 (2007) 507–512.
- [25] S.V. Trukhanov, A.V. Trukhanov, A.N. Vasiliev, H. Szymczak, JETP 111 (2010) 209–214.
- [26] S.V. Trukhanov, D.P. Kozlenko, A.V. Trukhanov, J. Magn. Magn. Mater. 320 (2008) e88–e91.
- [27] S.V. Trukhanov, A.V. Trukhanov, S.G. Stepin, H. Szymczak, C.E. Botez, Phys. Sol. State 50 (2008) 886–893.
- [28] S.B. Galvão, A.C. Lima, S.N. deMedeiros, J.M. Soares, C.A. Paskocimas, Mater. Lett. 115 (2014) 38–41.
- [29] V.A. Rane, S.S. Meena, S.P. Gokhale, S.M. Yusuf, G.J. Phatak, S.K. Date, J. Electron. Mater. 42 (2013) 761–768.
- [30] Z.F. Zi, Y.P. Sun, X.B. Zhu, Z.R. Yang, J.M. dai, W.H. Song, J. Magn. Magn. Mater. 320 (2008) 2746–2751.
- [31] T.B. Ghzaiel, W. Dhaoui, A. Pasko, F. Mazaleyrat, J. Alloy. Compd. 671 (2016) 245–253.
- [32] B. Grindl, Z. Beji, G. Viau, A. BenAli, J. Magn. Magn. Mater. 449 (2018) 119–126.
- [33] Y. Yang, F. Wang, X. Liu, J. Shao, S. Feng, D. Huang, M. Li, J. Magn. Magn. Mater. 422 (2017) 209–215.
- [34] J. Li, H.W. Zhang, Q. Li, Y.X. Li, Q.L. Yu, J. Rare Earths 31 (2013) 983–987.
- [35] Y.J. Yang, X.S. Liu, D.L. Jin, Mater. Sci. Eng. B 186 (2014) 106–111.
- [36] J. Park, Y.-K. Kim, S.-G. Kim, S. Kim, L.S.I. Liyanage, J. Lee, W. Lee, G.S. Abo, K.-H. Hur, S.-Y.A., J. Magn. Magn. Mater. 355 (2014) 1–6.
- [37] A. Thakur, R.R. Singh, P.B. Barman, J. Magn. Magn. Mater. 326 (2013) 35–40.
- [38] W.J. Zhang, Y. Bai, X. Han, L. Wang, X.F. Lu, L.J. Qiao, J.L. Cao, D. Guo, J. Alloy.

- Compd. 556 (2013) 20–25.
- [39] K.M.U. Rehman, X. Liu, M. Li, S. Jiang, et al., *J. Magn. Magn. Mater.* 426 (2017) 183–187.
- [40] M. Mozaffari, A. Arab, M.H. Yousefi, J. Amighian, *J. Magn. Magn. Mater.* 322 (2010) 2670–2674.
- [41] H.Z. Wang, Y.N. Hai, B. Yao, Y. Xu, L. Shan, L. Xu, J.L. Tang, Q.H. Wang, *J. Magn. Magn. Mater.* 422 (2017) 204–208.
- [42] B.K. Rai, S.R. Mishra, V.V. Nguyen, J.P. Liu, *J. Alloy. Compd.* 550 (2013) 198–203.
- [43] Long Peng, Lezhong Li, Xiaoxi Zhong, Rui Wang, Tu. Xiaoqiang, *J. Magn. Magn. Mater.* 411 (2016) 62–67.
- [44] Y.J. Yang, X.S. Liu, *Mater. Technol.* 29 (2014) 232–236.
- [45] I.A. Auwal, B. Ünal, H. Güngüneş, S.E. Shirsath, A. Baykal, *Ceram. Int.* 42 (2016) 9100–9115.
- [46] W.M.S. Silva, N.S. Ferreira, J.M. Soares, R.B. daSilva, M.A. Macêdo, *J. Magn. Magn. Mater.* 395 (2015) 263–270.
- [47] S.K. Chawla, S.S. Meena, P. Kaur, P.K. Mudsainiyan, S.M. Yusuf, *J. Magn. Magn. Mater.* 378 (2015) 84–91.
- [48] S.K. Chawla, P.K. Mudsainiyan, S.S. Meena, S.M. Yusuf, *J. Magn. Magn. Mater.* 350 (2014) 23–29.
- [49] C. Kittel, *Introduction to Solid State Physics*, Wiley, New York, 2005.
- [50] Z.M. Wang, *Ferrite Production Technology*, Chongqing University Press, Chongqing, 2013.