Characterisations of $Ce_xY_{3-x}Fe_5O_{12}$ (x = 0, 0.5, 1 and 1.5) thin films prepared using sol-gel method

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$Ce_xY_{3-x}Fe_5O_{12}$ (x = 0, 0.5, 1 and 1.5) thin films were prepared using sol-gel method. The X-ray diffraction patterns of the films show that the as-prepared film is amorphous and different x values need different annealing temperatures in order to crystallise the films. The magnetic measurement using a vibrating sample magnetometer shows that all of the films are hard magnetic materials with low saturation magnetisation values. The particles size measurement using a scanning electron microscope shows that all the particles are less than 150 nm.

I. INTRODUCTION

Cerium substituted yttrium iron garnets ($Ce_xY_{3-x}Fe_5O_{12}$; Ce-YIG) have been extensively studied due to their high potential in magneto-optic and microwave applications [1]. They have been prepared as crystals [2], powders [1,3] and also thin films [4]. Various thin film preparation techniques such as laser ablation process [5] and RF magnetron sputtering [6,7] have been employed to prepare the films. However, these techniques required complicated and expensive equipment such as a high power laser source and a high vacuum system. Recently, a simple and inexpensive sol-gel method has become popular among thin film researchers [8]. The technique that is normally used for bulk ceramic material preparation involves 3 steps, that is preparation of a sol, gelation of the sol and removal of the solvent. To transfer the gel into thin film form, a dip coating or spin coating technique is used. Sol-gel can also be used to prepare thick film in one step as the thickness depends only on the precursors concentrations.

In this paper, we report on the characterisations of some Ce-YIG thin films prepared using sol-gel method. The sol was prepared from inorganic precursors and to transform the gel into thin film form a spin coater was used.

II. EXPERIMENTAL DETAILS

To prepare $Ce_xY_{3-x}Fe_5O_{12}$ (x = 0, 0.5, 1 and 1.5) solutions, weighted amounts of yttrium nitrate pentahydrate ($Y(NO_3)_3.5H_20$) and iron nitrate nanohydrate ($Fe(NO_3).9H_2O$) were first dissolved in 2-methaoxyethanol and refluxed at ~80^{o}C for 5hours. The resulting solution was stirred for 48 hours. Then cerium (III) nitrate hexahydrate ($Ce(NO_3)_3.6H_2O$) was added followed by acetic acid and diethylamine. The solution was again stirred for 24 hours. To transform the solution into thin film form, a few drops of the solution was put on a clean quartz substrate and was spin coated at 3500 rpm for 30 seconds. The film was heated at 150^{o}C to get rid of residual organic compounds. Then annealing process was carried out at 700^{o}C for 2 hours in air.

The films microstructures and magnetic properties were studied using an x-ray diffractometer (XRD) and a vibrating sample magnetometer (VSM), respectively. The films grain sizes were measured using a scanning electron microscopy (SEM).

III. RESULTS AND DISCUSSIONS

The films microstructures were studied using an XRD and the obtained results were compared with the joint committee on powder diffraction standard (JCPDS) cards. Fig. 1 shows the typical XRD spectrum for the as-prepared films. They are amorphous. Annealing the films at 700^{o}C for 2 hours crystallise the x = 0 and 1 films into tetragonal Ce-YIG, but x = 0.5 and 1.5 are still amorphous (Fig. 2). The film x = 1.5 shows some peaks only after further annealing at 800^{o}C for 2 hours (Fig. 3). The film crystallise into mixed structure of orthorhombic and cubic Ce-YIG. These results show that different annealing temperatures are required to crystallise $Ce_xY_{3-x}Fe_5O_{12}$ films with different x values.

The in-plane magnetisations properties were measured using a vibrating sample magnetometer. Fig. 4 shows the typical hysteresis loop for all of the films. Table I summarises the saturation magnetization (Ms) and coercivity (Hc) for the crystalline films. All of the films are hard magnetic materials with very low Ms values compared to the bulk YIG value of 26.94 emu/g. There are two possible reasons for the low Ms values. Firstly, the effect of size of the particles and shape of the materials. Sanchez *et al.* [3] has also reported the reduction of the Ms values for YIG with nanosize particles. Their estimated critical diameter value below which a spherical particle will be single domain is < 200 nm [3,9]. As our measured particles sizes are also < 200 nm (Fig. 5), the low Ms values could be due to the nanosize particles. Similar reduction of the magnetization was also observed in small iron [10],

γ-Fe_2O_3 [11], $BaFe_{12}O_{19}$ [12] and $MnFe_2O_4$ [13] particles. The second possible reason is the effect of substituting Ce in YIG that increase the lattice parameter of the YIG as the substituted ionic radius (1.14 Å) is bigger than the Y^{3+} ionic radius (1.015 Å). This will reduce the magnetic moment per unit mass of YIG. Ibrahim *et al*. [5] reported Ms value of ~19.71 emu/g and 16.16 emu/g for $CeY_2Fe_5O_{12}$ films prepared using pulsed laser ablation deposition in argon and oxygen atmosphere, respectively. This technique has been reported producing thin film with micron size particles [14-16]. Hence, the low Ms values of the present films are believed to be due to the small particles of the films.

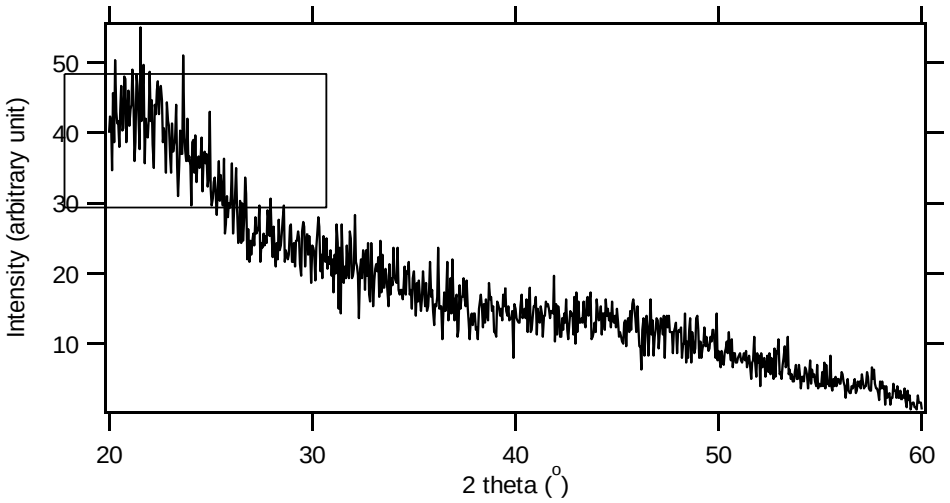

FIG. 1. The typical XRD spectrum for the as-prepared films.

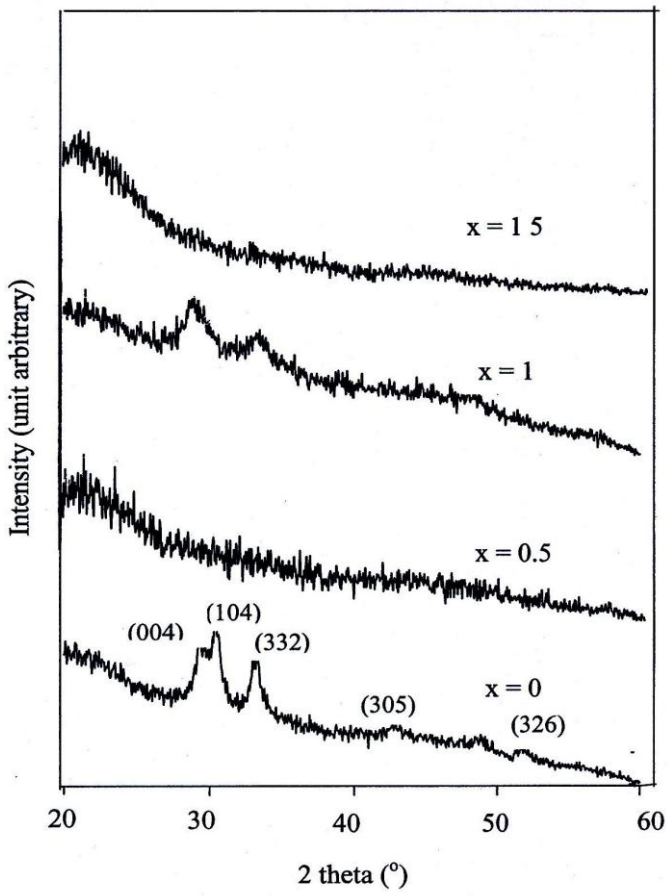

FIG. 2. The XRD spectra for all films annealed at 700°C for 2 hours.

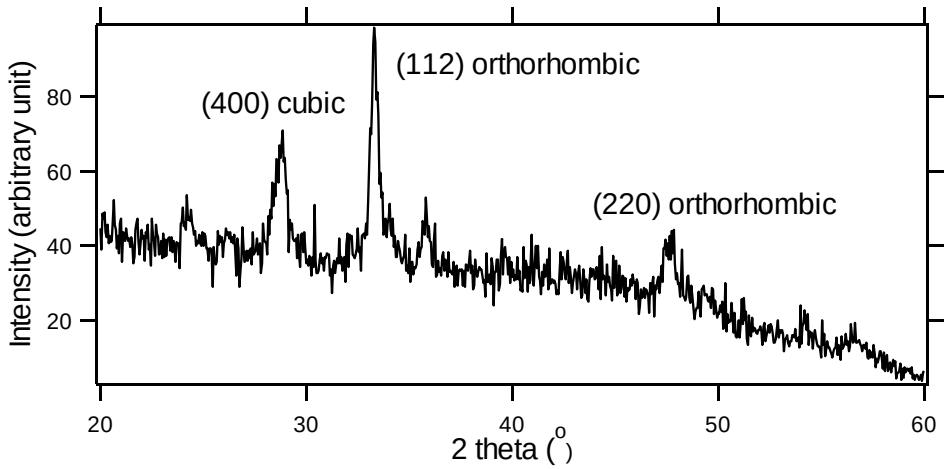

FIG. 3. The XRD spectrum for the film x = 1.5 annealed at 800°C for 2 hours.

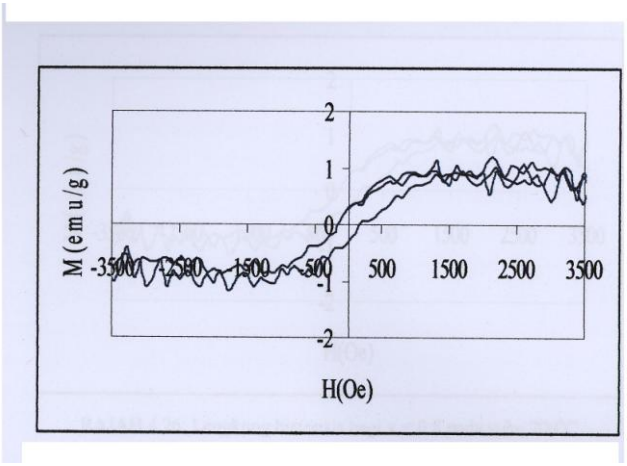

FIG. 4. The typical in-plane hysteresis loop for all of the crystalline films.

TABLE I. Summary of the coercivity (Hc) and saturation magnetisation (Ms) of all crystalline films.

x	Coercivity, Hc (±1 Oe)	Saturation magnetisation, Ms (±0.01 emu/g)
0	200	1.16
1.0	194	0.16
1.5	192	0.05

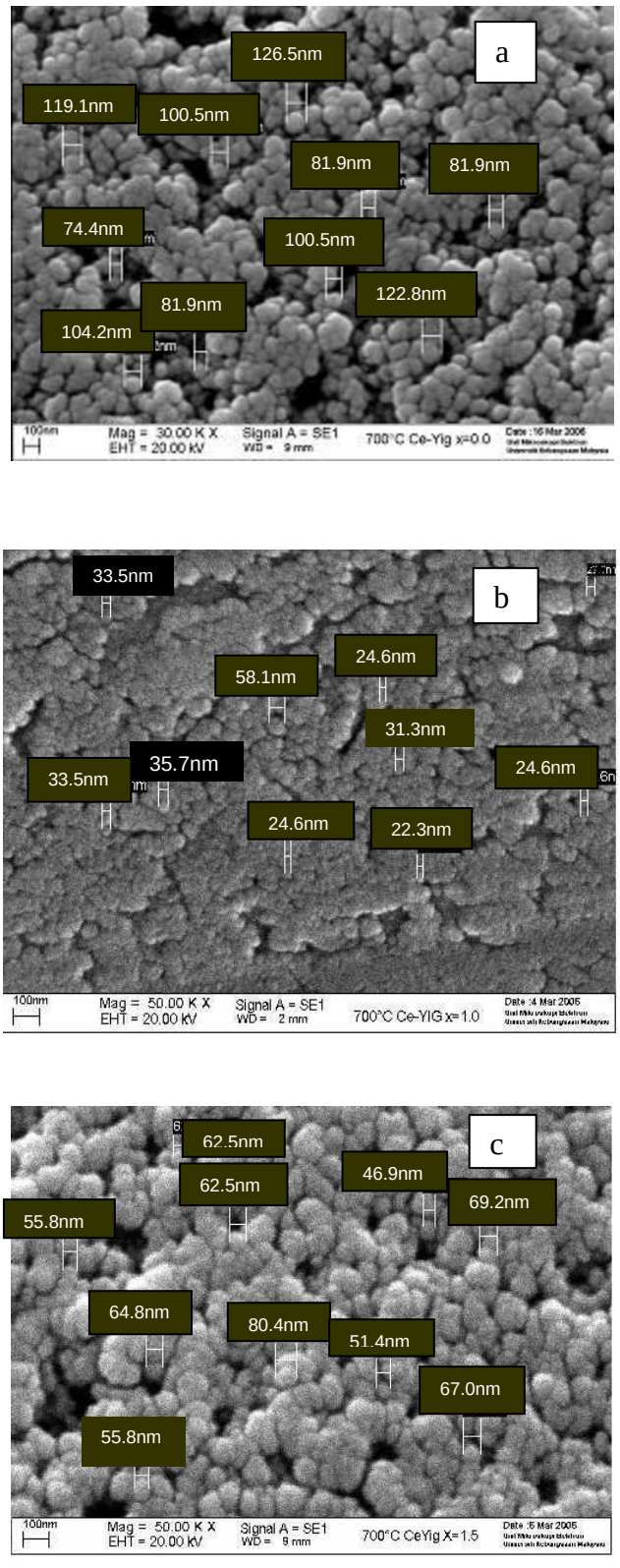

FIG. 5. SEM micrographs of the films (a) x = 0 (b) x = 1 and (c) x = 1.5.

IV. CONCLUSIONS

$Ce_xY_{3-x}Fe_5O_{12}$ (x = 0, 0.5, 1 and 1.5) thin films have been prepared using a sol-gel method. X-ray diffraction characterisation shows that the as-prepared films are amorphous. Different compositions need different annealing temperatures to crystallise them. The crystalline films are hard magnetic materials with very low saturation magnetizations. The low saturation magnetisations are due the nanometer particles of the films.

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