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The effect of yttrium substitution on the magnetic properties of magnetite nanoparticles

M. Mozaffari *, J. Amighian, R. Tavakoli

Department of Physics, Faculty of Science, University of Isfahan, Hezarjarib, 81746-73441 Isfahan, Iran

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ABSTRACT

Superparamagnetic Y-substituted magnetite ($Y_xFe_{3-x}O_4$,with $x=0.00$, 0.10, 0.15, 0.20 and 0.40) nanoparticles were synthesized via hydrothermal reduction route in the presence of citric acid. The synthesized nanoparticles were characterized by X-ray diffraction (XRD) analysis, Fourier transform infrared (FTIR) spectroscopy, field emission scanning electron microscopy (FESEM), vibrating sample magnetometry (VSM) and gradient field thermomagnetic measurement. The results showed that a minimum amount of citric acid is required to obtain single phase Y-substituted magnetite nanoparticles. Citric acid acts as a modulator and reducing agent in the formation of spinel structure and controls nanoparticle size and crystallinity. Mean crystallite sizes of the single-phase powders were estimated by Williamson–Hall method. Curie temperature measurement of the samples shows that as yttrium content increases, the Curie temperature decreases. Magnetic measurements show that the saturation magnetization of the samples decreases as x increases up to 0.15 and then increases to $x=0.20$ and finally decreases again for $x=0.40$.

1. Introduction

Today a great number of research works have been carried out on ferrites. This is due to their wide applications in many kinds of magnetic devices such as transformers, inductors, magnetic heads, resonance circuits for high frequency devices (ranging from 10^3 to 10^{11} Hz). This is due to the fact that ferrites have high electrical resistivity, low eddy current loss, high initial permeability, moderate saturation magnetization and low hysteresis loss [1].

In the last decades, considerable research works have been devoted to the synthesis and characterization of nanosized materials. In particular iron oxides nanoparticles, such as magnetite (Fe_3O_4) [2], maghemite (γ-Fe_2O_3) [3], and hematite (α-Fe_2O_3) [4] have found a wide-range of applications in different fields [5]. Magnetite (Fe_3O_4) is a common magnetic iron oxide, and it has a cubic inverse spinel structure with oxygen forming an FCC closed packed structure and Fe cations occupy the interstitial tetrahedral (A) and octahedral (B) sites [6].

Magnetite is a ferrimagnet at room temperature and has many applications including ferrofluids [7], ultrahigh density magnetic storage media [8], labeling, detecting and separating in biology [9], catalyzes, as well as contrast agent in magnetic resonance imaging [10–12], and as a mediator in magnetic hyperthermia [13,14].

Various chemical routes have been developed to synthesize nanosized nanoparticles, including mechanochemical processing [15], coprecipitation [16], sol–gel [17], electroxidation [18], hydrothermal [19,20] and hydrothermal reduction [21].

In this work, superparamagnetic Y-substituted magnetite nanoparticles ($Y_xFe_{3-x}O_4$) with different amounts of Y ($x=0.00$, 0.10, 0.15, 0.20 and 0.40) were synthesized by hydrothermal reduction route at 200 °C in the presence of citric acid as a reducing agent and stabilizer, which is an inexpensive and non-toxic reducer. The hydrothermal reduction route is an economic, facile, environmental friendly and efficient method which needs less energy in comparison with other preparation methods [21].

2. Experimental

The raw materials were $Fe(NO_3)_3\cdot 9H_2O$, NaOH and $C_6H_8O_7\cdot H_2O$ all from Merck company and $Y(NO_3)_3\cdot 9H_2O$ from CSTARM Co. all with minimum purities of 99%.

In a typical experiment, 3 mM of $Fe(NO_3)_3\cdot 9H_2O$ were dissolved in 20 ml of deionized double distilled water under continuous stirring. Prior to this step, a stock solution of 3 M was prepared from NaOH and used in all sample preparation processes. NaOH solution was added drop by drop to reach a pH of 9. Vigorous stirring continued for another 15 min to achieve reddish brown slurry. The slurry was then centrifuged and washed with deionized double distilled water to remove excess ions and reach

* Corresponding author. Fax: +98 31 3793 4800.
E-mail addresses: mozafari@sci.ui.ac.ir, mmozafari@hotmail.com (M. Mozaffari).

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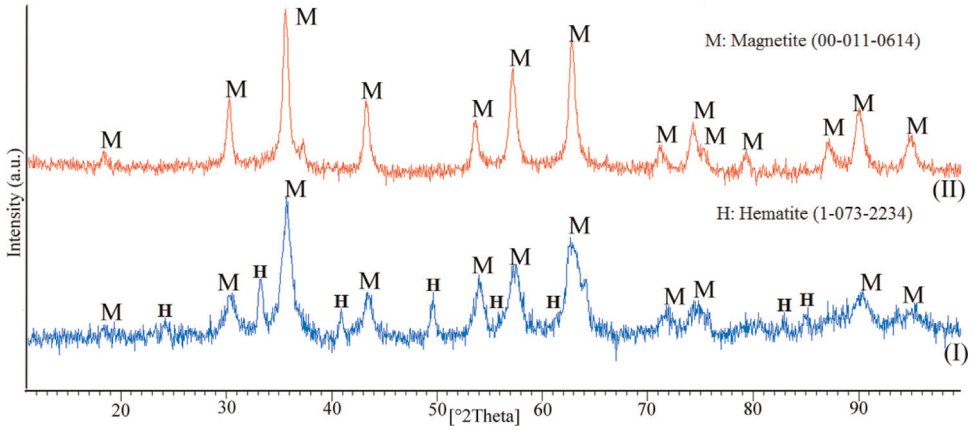

Fig. 1. XRD patterns of the synthesized samples with (I) 1.5 and (II) 2 mM citric acid concentration.

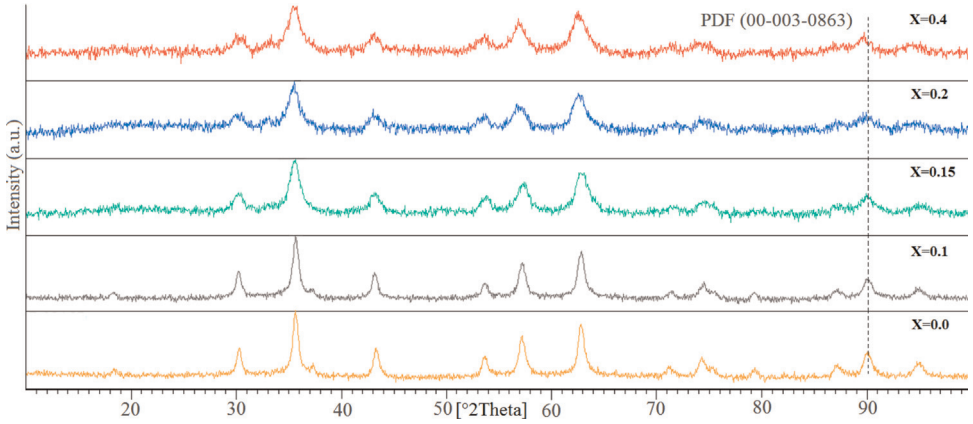

Fig. 2. XRD patterns of $Y_xFe_{3-x}O_4$ nanoparticles (x=0, 0.1, 0.15, 0.2 and 0.4)with 2 mM citric acid. The vertical dashed line at 90° is drawn to guide eye.

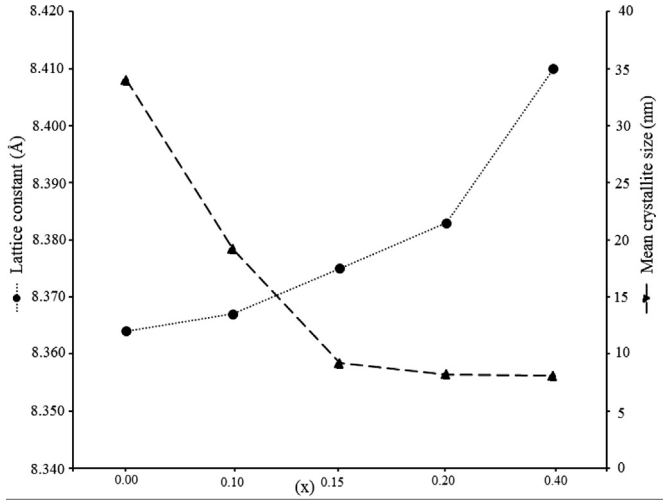

Fig. 3. Variations of lattice constant and crystallite size with respect to x value.

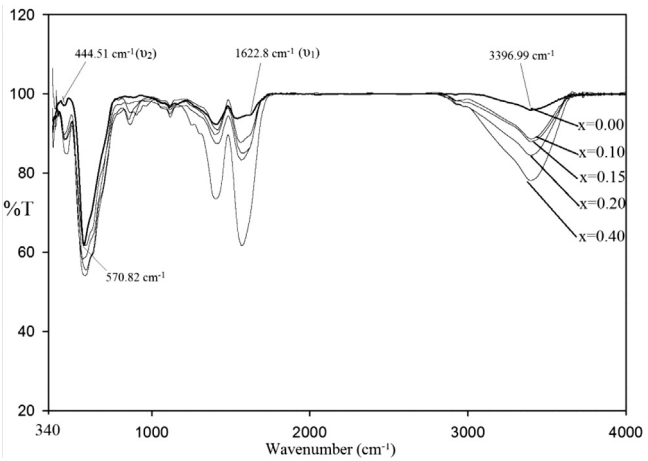

Fig. 4. FT-IR spectra of the Y-substituted magnetite nanoparticles with different x values.

to a pH of 7. To optimize concentration of citric acid, different amounts of the acid (0.5, 1, 1.5 and 2 mM) were added to the iron oxide precursors. After being stirred for another 15 min, the mixtures were transferred into an 80 ml volume Teflon-lined autoclave and were subsequently heated in an oven at 200 °C for 20 h. The autoclave was free cooled after hydrothermal treatment and the precipitate was recovered and washed with distilled water several times and finally rinsed with acetone to eliminate any unwanted impurities. The nanoparticles were dried at room temperature for few days. To synthesize Y-substituted magnetite nanoparticles, appropriate molar ratio of iron nitrate was replaced by yttrium nitrate.

XRD patterns of the nanoparticles were obtained using an X-Ray diffractometer (Philips, X'PERT model), using CuK_α radiation (λ=0.15406 nm). The mean crystallite sizes of the samples were estimated from the widths of the diffraction peaks, using the Williamson–Hall method [15,16]:

$$\beta \cos\theta = \frac{0.9\lambda}{d} + 2\varepsilon \sin\theta$$