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Characterization of Cobalt Ferrite Nano-particles Synthesized by Polymerized Complex Sol-gel Method

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ABSTRACT

In this study, single-phase cobalt ferrite nanoparticles with nearly spherical morphology and an average size of approximately 30-50 nm were synthesized via a straightforward sol-gel method. This method relied on a polycondensation reaction between citric acid, benzoic acid, and ethylene glycol, carried out without adjusting the pH and under strongly acidic conditions, eliminating the need for gel aging. These characteristics render the method flexible and practical for synthesizing various types of nanocomposite materials.
The thermal behavior, phase evolution, and magnetic properties of the products were assessed using DTA, XRD, and VSM techniques. Additionally, SEM and TEM were utilized to examine particle size and morphology. Thermal studies indicated a three-step process: loss of residual water in the gel, de-esterification, and decarboxylation of COOH groups, culminating in the formation of cobalt ferrite.
The impact of calcination temperature on the phases and magnetic properties of the powders was thoroughly analyzed. Heat treatment in air at 750°C for 1 hour successfully produced pure spinel cobalt ferrite nanoparticles. In contrast, annealing at 850°C for the same duration resulted in the highest magnetization among all samples tested. This investigation established a clear relationship between the microstructure of the nanoparticles and their magnetic properties, highlighting the importance of anealing temperature in the magnetic behavior of cobalt ferrite nanoparticles.

Keywords: Cobalt ferrite, Nanopowder, Polymerized complex, Magnetic properties

Introduction

Synthesis and characterization of spinel ferrites have been intensively studied because of fundamental understanding as well as their applicability in a variety of areas such as high-density information storage system [1], ferro fluid technology [2], biological and clinical applications [3] and gas sensors [4].
Among spinel ferrites, cobalt ferrite, $CoFe_2O_4$, has attracted considerable attention due to its large magneto-crystalline anisotropy, high coercivity, moderate saturation magnetization, large magneto-strictive coefficient, chemical stability and mechanical hardness [5]. Several chemical methods like sol-gel [6], micro-emulsion [7], citrate gel [8], co-precipitation [9], auto-combustion synthesis [10] and hydrothermal process [11] have been reported to synthesize cobalt ferrite nano-particles.
Recently, polymerized complex (PC) which is a modified Pechini type [12,13] sol-gel method, in which a solution of ethylene glycol, citric acid and metal ions is polymerized to a polyester network, has been employed to process nano size ceramic powders at a relatively low temperatures [13-15]. In this research, nano particles of cobalt ferrite were prepared by a PC method. The effects of the synthesis parameters on the powder particle characteristics were also investigated crystalline

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Experimental Procedure

Iron citrate, cobalt nitrate, citric acid, ethylene glycol and benzoic acid all of analytical grade were used as starting materials. A mixed solution with $4.61C_6H_8O_7$: $13.84(CH_2OH)_2$: $0.7C_6H_5COOH$: $2FeC_6H_7O_7$: 1Co $(NO_3).6H_2O$: $300H_2O$ molar ratio was used to prepare Co-Fe complex solution.

First, the citric acid was dissolved in water at 60°C followed by addition of iron citrate and cobalt nitrate to the solution under stirring until a bright brownish homogenous solution achieved. Then ethylene glycol and benzoic acid were added to the solution. The pH of solution was 1.4 in this step of the process. Then stirring was being continued, without pH adjusting, at 80°C for 1.5 hour to obtain a dark brownish viscose gel. The obtained gel was dried at 150°C for 24 hours under the aire atmosphere. Annealing process was carried out at 550, 650, 750 and 850°C for 1 hour by a heating rate of 10°C/min.

The phase identification of the powder samples were performed by X-ray diffraction (XRD) ,Bruker D8 Instrument, using Cu Ka radiation (λ= 1.5405 A °). Each diffraction pattern was recorded at a step of 0.01° and 1 s per step. All measurements were made at room temperature. The particle morphology was examined by Field Emission Scanning Electron Microscope (FESEM), Model LEO 1530VP and transmission electron microscopy (TEM), JEOL Model 2010, Super twin TEM, with point-to-point resolution of 0.23 nm, line resolution of 0.14nm. The thermal decomposition behavior of the gel was examined by differential thermal analysis (DTA) in air with the heating rate of 10˚C/min on the SDT Q 600 Thermal analysis instrument. Magnetic properties were measured by a vibration sample magnetometer (VSM), Meghnatis Daghigh Co., I.R. IRAN, with a maximum applied field of 10 kOe at room temperatur.

Results And Discussion

Fig. 1 illustrates the DTA curve of the dried gel prepared under the given experimental conditions of the PC method. It could be seen that there is one endothermic and two exothermic distinct peaks which appear about 110, 320 and 510°C respectively. Endothermic reaction related to dehydration of sample and exothermic reaction around 320°C due to de-esterification and de-carboxilation of COOH groups was occurred. exothermic reaction around 510°C due to the formation of cobalt ferrite

Phase evolution was monitored with increasing annaeal temperature of the precursor obtained by the PC method for 1 hour in air at various temperatures and then, phase identification was carried out by XRD.

Fig. 2 shows the XRD patterns of obtained samples. It is seen that the samples annealed at 550°C and 650°C show γ-Fe2O3 (maghemite) traces. By increasing the temperature to 750 and 850°C, cobalt ferrite single phase was achieved with sharp characteristic peak (Fig. 2c and d) related to high degree crystalline and the increase in crystallite size [17].

Regarding to above results, fundamentally this process consists of following stages of chemistry reactions: (1) complexation between acid citric and metal ions in water without aging and pH adjusting. Experiments revealed that using iron citrate in place of iron nitrate [16] was accelerated the complexation. (2) Polymerization, where formed polyester network when treated with ethylene glycol. Benzoic acid acted as a secondary coordinate agent and accelerated the polymerization and gel formation processes rather than other reports [13, 17]. (3) The decomposition of the organic network to obtain powder precursor after heating at moderate temperature. Both SEM and TEM images of obtained powder at 850°C / 1 houre have been shown in Fig. 3 and 4, respectively. Derived powder showed a uniform particle size distribution with spherical shape, a fine powder size in nano-scale range between 30-50 nm, and homogenous microstructure.Magnetic properties of the samples annealed at different temperature were evaluated at room temperature using VSM analysis. The magnetization curves are shown in Fig. 5 and the extracted coercivity (Hc), saturation magnetization (Ms) and remanence magnetization (Mr) values are summarized in Table 1. It demonstrated exist of a relationship between magnetic properties of cobalt ferrite nano-particles and annealing temperature. The coercivity amount of magnetic nano-particles is related to the microstructure, phase composition, particle size, residual strain and many other complex factors [18]. The coercivity of sample annealed at 550°C was small as 567.2 Oe. However, the highest coersivity of 1353.7 Oe was obtained in the sample annealed at 650°C and then drastically was decreased to 587.6 Oe at 750°C. The drastic drop of Hc for the sample annealed at 750°C might originate from the transition of magnetic single domain to magnetic multi-domain within a particle [18]. Decrement of coercivity for the sample annealed at 850°C continued to 577.5 Oe. The Ms increased with annealing temperature with a notice increment occurring 550 and 650°C from 36.9 to 56.2 emu/g. similar trends also was found in Mr from 7 to 33.8. The maximum Ms (79.4 emu/g) and Mr (43.2 emu/g) were achieved in powder annealed at 850°C. Maximum magnetization of resulted powder is favorably closed to cobalt ferrite bulk (80emu/g). Improvement of Ms and Mr was highly depending on growth of cobalt ferrite nano-particles, phase evolution and high degree crystallinity by increasing the annealed temperature.

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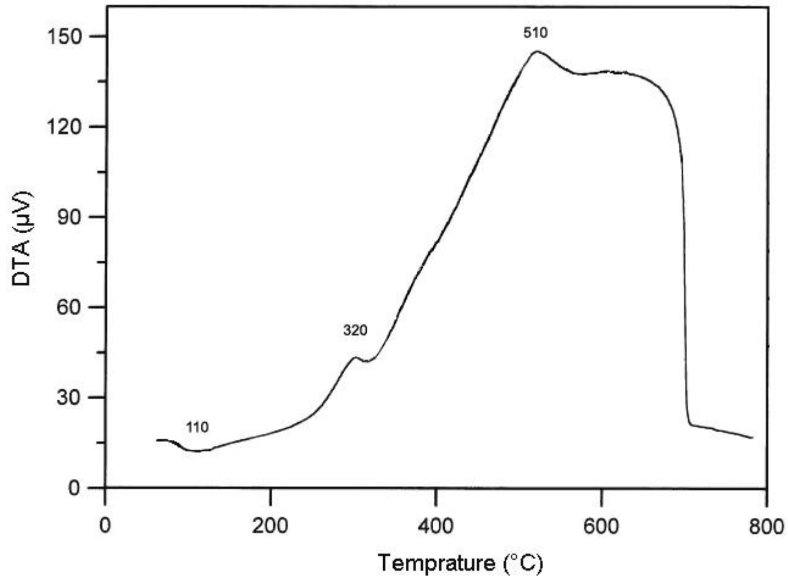

Fig.1. DTA curve of the dried gel prepared under the given experimental conditions.

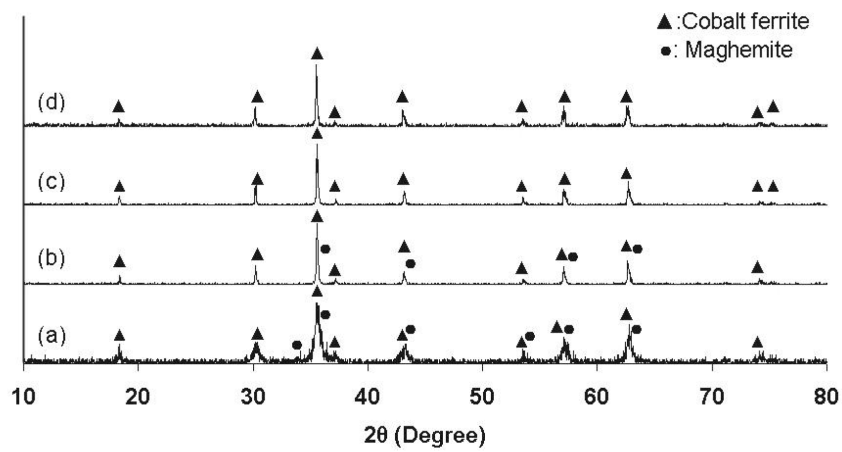

Fig. 2. X-ray diffraction patterns of samples annealed at different temperatures.

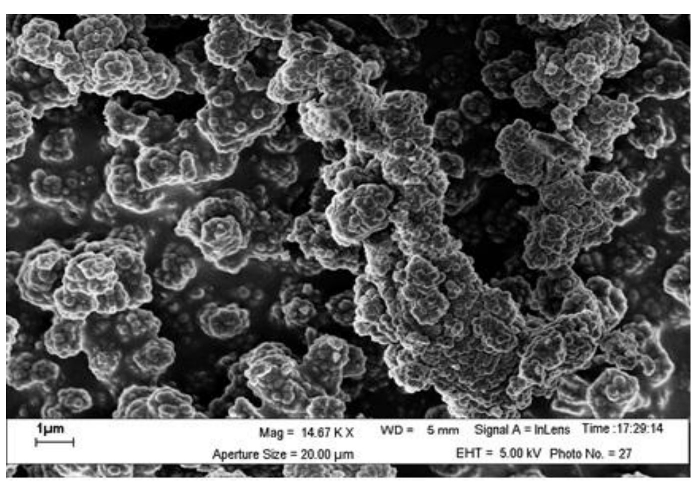

Fig.3. SEM image of precursor annealed powder at 850°C.

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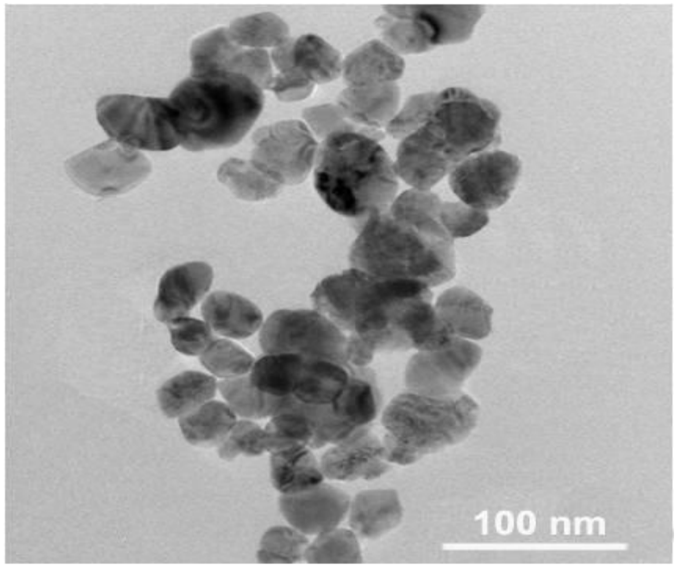

Fig.4. TEM image of sample annealed at 850°C.

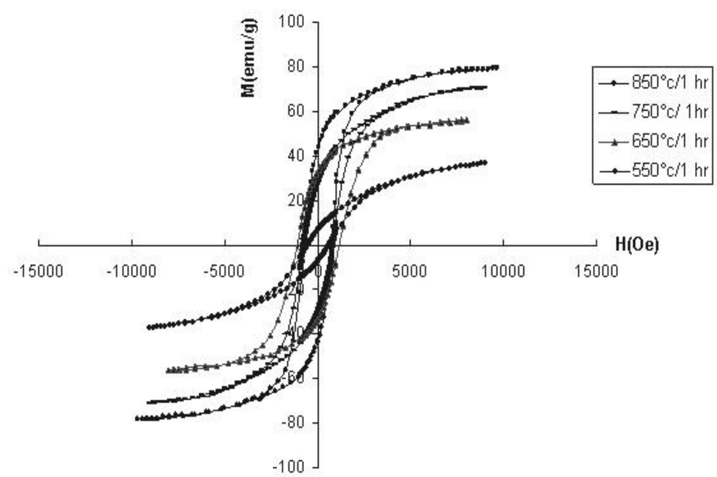

Fig.5. Magnetization curves of samples annealed at different temperatures

Table 1. Magnetic properties of samples annealed at different temperatures

Anealing temperature (°C)	**M_s (emu/g)**	**M_r (emu/g)**	**H_c (Oe)**
550	36.9	7	567.2
650	56.2	33.8	1353.7
750	70.7	29.5	587.6
850	79.4	43.2	577.5

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Conclusion

Cobalt ferrite nano-particles were successfully synthesized by a facile sol-gel route based on polyesterification reaction between citric acid, benzoic acid and ethylene glycol under strong acidic condition of achieved complex solution, without pH adjusting of solution and aging of gel. A pure spinel cobalt ferrite was formed after heat treatment at 750° C for 1 h. It is concluded that there is a relation between magnetic properties and annealing temperature. By increasing the temperature, saturation magnetization and remannence magnetization were achieved 79.4 and 43.2 emu/g at 850°C, respectively that saturation magnetization is favorably closed to cobalt ferrite bulk (80emu/g). The highest coersivity of 1353.7 Oe was obtained in the sample annealed at 650°C.

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