Synthesis of nano $Co_{1-x}Ni_xFe_2O_4$ by sol-gel method and its properties

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ABSTRACT

Nano ferrites have received considerable attentions due to their various applications such as magnetic or catalyst materials. In this work, nickel-cobalt ferrite ($Co_{1-x}Ni_xFe_2O_4$) was synthesized by sol-gel method using stearic acid. The effects of calcination temperature and nickel/cobalt ratios on the formation of structure were also investigated. XRD results show that all samples which were calcined from 6000C to higher temperature for 1 hour were in single cubic spinel phases. The magnetic properties include saturation magnetization (M_s) and coercivity (H_c) have been also investigated by using vibrating sample magnetometer (VSM). The saturation magnetization and coercivity of $CoFe_2O_4$ calcined at 600^0C for 1hour is 74.4 emu/g and 1519.13 Oe, respectively. The saturation magnetization and coercivety of substituted materials decrease with increasing Ni content.

Keywords: *Nano $Co_{1-x}Ni_xFe_2O_4$, magnetic materials, sol-gel method.*

1. INTRODUCTION

Ferrite compounds are magnetic materials which are used in many technological applications because of their good combination of magnetic and electrical properties. The spinel ferrite (MFe_2O_4, where M are Zn, Mn, Ni, and Co) is a kind of material system for high-frequency passive components because of its high permeability, resistivity and permittivity [1-3]. Among magnetic nano powders, $Co_{1-x}Ni_xFe_2O_4$ nano powders are a class of magnetic material with excellent performance, prominently chemical stability, resistance to oxidation, moderate saturation magnetization, high mechanical strength [4-6], inverse spinel structure, a well-known hard magnetic material [5-8], having large magnetic anisotropy, high coercivity, and high Curie temperature around 793 K. Ferrite compounds was synthesized by using co-precipitation, micro emulsion, thermal decomposition, hydrothermal, sol-gel method [1,5,6,7]. In this study, $Co_{1-x}Ni_xFe_2O_4$ was

prepared by sol-gel method with stearic acid that has not been reported.

On the other hand, combination of soft and hard magnetic (MFe_2O_4, with M are Ni, and Co) properties make them the promising candidate for many different electronic applications such as in the telecommunication field or recording technology and biomedical [4-7]. In this study, Ni-Co ferrite was synthesized by sol-gel method and effect of Ni/Co ratios on the spinel structure, magnetic properties of $Co_{1-x}Ni_xFe_2O_4$ were also investigated.

2. METHODOLOGY

For the preparation of nickel-cobalt ferrites by sol-gel method, an appropriate amount of stearic acid (0.132 moles) was first melted in a beaker at 70°C (melting point of stearic acid). Then 250 ml mixed solutions of $Fe(NO_3)_3.9H_2O$ (0.03 moles), $Co(NO_3)_2.6H_2O$ and $Ni(NO_3)_2.6H_2O$ with certain molar ratios (Fe^{3+}: M^{2+} =2:1, M are Ni, Co by ratio of Co : Ni = 0.1:0:9; 0.3:0.7; 0.5:0.5; 0.7:0.3; and 1.0:0, respectively) were added. The mixture was thoroughly stirred by magnetic stirrer in 5 hours to vaporize completely water and homogenous solutions high viscously were formed. The obtained sol was dried in the drier at 200°C for 5 hours. Then solution was ignited in air at 400ºC (Auto-Ignition Temperature of stearic acid) for 30 mins to ensure stearic acid in mixed fired completely and the obtained powders were calcined at different temperatures ranging 500°C, 600°C, 800°C, and 1000°C for 1 hour.

The crystal structure was characterized by an X-Ray diffractometer (Bruker D8 Advance, Germany) with CuKα radiation (λ=0.15406). The morphology of uncoated $Co_{1-x}Ni_xFe_2O_4$ sample was investigated by Field Emission Scanning Electron Microscopy (FESEM, S4800-Hitachi). The magnetic properties of particles were measured at room temperature using a vibrating sample magnetometer (VSM, EV11-VSM, KLA Tencor - USA) with the maximum applied field of 15 kOe

3. RESULTS AND DISCUSSION

3.1. Effect of calcination temperature on the formation structure

To survey effect of calcination temperature, the obtained powders after igniting in air at 400ºC were calcined at different temperatures ranging 500°C, 600°C, 800°C, and 1000°C for 1 hours. The XRD result in *Figure 1* shows that $Co_{1-x}Ni_xFe_2O_4$ (with x=0.5) single phase was formed since 600°C and there are no impurities in all samples. The crystallite size increases from 25.27 nm (at 500°C) to 45.83 nm (at 1000°C) as in *Table 1*. The increasing of crystallite size also completes agree with intensity and broadening of peak from XRD results in *Figure 1*.

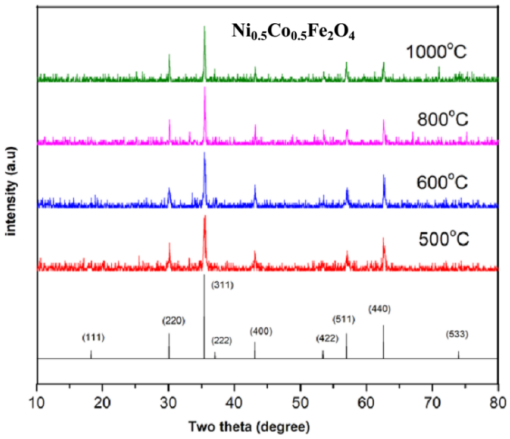

Figure 1. XRD patterns of $Co_{1-x}Ni_xFe_2O_4$ (with x =0.5) calcined at 500°C, 600°C, 800°C and 1000°C for 1 hour

Table 1. The crystallite size of $Co_{1-x}Ni_xFe_2O_4$ (with x = 0.5)

Temperature (°C)	Crystallite size (nm)
500	25.27
600	36.00
800	42.58
1000	45.83

The crystallite size of $Co_{1-x}Ni_xFe_2O_4$ (with x = 0.5) calcined at 1000°C for 1 hour is greater than the results in Gharagozlou's report [9] (34.3 nm) and Rao 's report [10] (7.5 nm).

3.2. Effect of Ni/Co ratios on the material's structure.

To investigate the effect of the ratio of Ni/Co, $Co_{1-x}Ni_xFe_2O_4$ powders were synthesized at 600°C for 1 hour calcination with different values of x: 0; 0.3; 0.5; 0.7; 0.9, respectively.

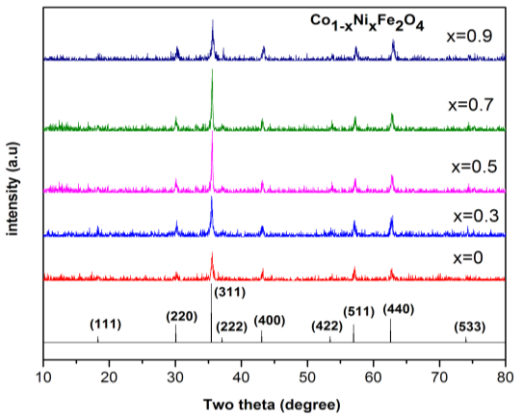

Figure 2. XRD patterns of $Co_{1-x}Ni_xFe_2O_4$ which were calcined at 600°C for 1 hour.

The XRD patterns are shown in *Figure 2*. There is no impurity detected in all samples indicating that single-phase cubic structure of $Co_{1-x}Ni_xFe_2O_4$ was successfully synthesized by sol-gel method with stearic acid. The diffraction lines corresponding to a cubic, spinel-type and crystalline phase indicate the formation of series of solid solutions between $CoFe_2O_4$ and $NiFe_2O_4$ [7].

On the other hand, the SEM results of $Co_{1-x}Ni_xFe_2O_4$ powders (x = 0, 0.5, 0.9) which are shown in *Figure 3* indicate that the particles are agglomerated sphere. The grains are uniform size and in nano regime and no significant change with ratio of Ni/Co. From the SEM micrographs of the powders, the particle sizes are average in the range of 30- 70 nm.

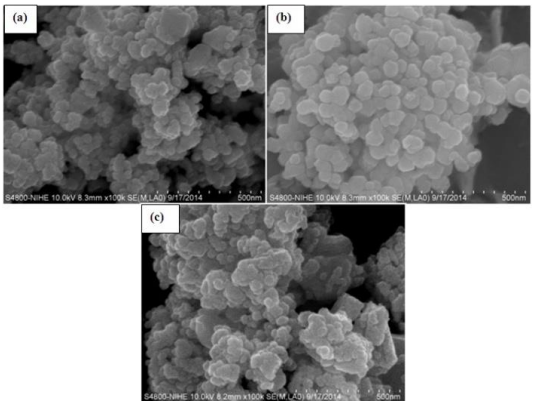

Figure 3. The SEM images of the $Co_{1-x}Ni_xFe_2O_4$ powder; (a) x=0; (b) x=0.5; (c) x=0.9

3.3. Magnetic properties of Co1-xNixFe2O4.

Table 2. The saturation magnetization (M_s), coercivity (H_c), remanence magnetization (M_r)

x	M_s (emu/g)	H_c (Oe)	M_r (emu/g)	M_r/M_s
0	74.4	1519.1	30.7	0.41
0.3	52.5	948.0	19.9	0.38
0.5	52.4	858.6	21.9	0.42
0.7	41.9	504.4	15.6	0.37
0.9	42.8	64.7	10.1	0.24

The magnetic properties of $Co_{1-x}Ni_xFe_2O_4$ at room temperature were also investigated and were indicated in *Table 2* and *Figure 4*.

For the series of $Co_{1-x}Ni_xFe_2O_4$ that calcined at 600°C for 1 hour, the saturation magnetization are in range of 42.8 - 74.4 emu/g, higher than those reported by Xiang [11] (29.3 - 56.4 emu/g) and by Tang [12] (42.5 – 67.5 emu/g). The coercivity are in range of 64.7 – 1519.1 Oe, higher than Chen 's study [13] (24.34 – 696.91 Oe). The saturation magnetization value is 74.4 emu/g which is very close to the value of the standard bulk material (80 emu/g for $CoFe_2O_4$ [8]). As the Ni content increases, the coercivity and the saturation magnetization decrease.

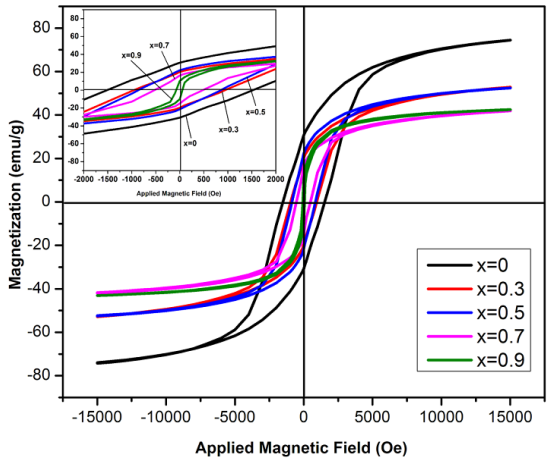

Figure 4. Magnetic hysteresis loop of $Co_{1-x}Ni_xFe_2O_4$ (x= 0, 0.3, 0.5, 0.7, 0.9) at room temperature.

4. CONCLUSIONS

Nickel - Cobalt ferrites $Co_{1-x}Ni_xFe_2O_4$ were synthesized successfully using a sol-gel method. The spinel structure formatted since 600°C in 1h calcination which indicated by XRD patterns. There is no impurity detected in all samples indicating that single-phase cubic structure of $Co_{1-x}Ni_xFe_2O_4$ were successfully formed by this method.

SEM images show the particles $Co_{1-x}Ni_xFe_2O_4$ (x=0.0, 0.5, 0.9) are spherical with somewhat agglomerated. The grains are uniformly sized and in nano regime.

The saturation magnetization and coercivity of $CoFe_2O_4$ are 74.4 emu/g; 1519.13 Oe, respectively. In addition, the saturation magnetization and coercivity decrease with increasing Ni content.

Tổng hợp nano $Co_{1-x}Ni_xFe_2O_4$ bằng phương pháp sol-gel và các tính chất

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TÓM TẮT

Nano ferrite được nhiều quan tâm nghiên cứu do có nhiều ứng dụng như làm vật liệu từ tính và vật liệu xúc tác. Trong bài báo này nickel-cobalt ferrite ($Co_{1-x}Ni_xFe_2O_4$) được tổng hợp bằng phương pháp sol – gel. Sự ảnh hưởng của nhiệt độ nung và tỉ lệ nickel/cobalt đến sự hình thành cấu trúc vật liệu đã được khảo sát. Kết quả phân tích nhiễu xạ tia X (XRD) cho thấy các mẫu được nung ở nhiệt độ từ 600^0C trở lên trong thời gian 1h đều ở dạng đơn pha tinh thể với cấu trúc spinel. Các thuộc tính từ của vật liệu như độ từ hoá bão hoà M_s và lực kháng từ Hc được đo bằng Từ kế mẫu rung (VSM) với kết quả tương ứng của mẫu nung ở 600^oC trong 1h là 74.4emu/g và 1519.13 Oe. Kết quả này giảm khi tăng hàm lượng của nickel.

Từ khóa: *Nano* $Co_{1-x}Ni_xFe_2O_4$*, vật liệu từ tính, phương pháp sol-gel*

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