The effect of temperature on synthesis of $MgFe_2O_4$ based on natural iron sand by Co-precipitation method as adsorbent Pb ion

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The effect of temperature on synthesis of $MgFe_2O_4$ based on natural iron sand by Co-precipitation method as adsorbent Pb ion

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Abstract. Magnetic materials of magnesium ferrite ($MgFe_2O_4$) based on natural iron sand has been successfully prepared as an adsorbent of Pb ions. This material was synthesized by co-precipitation method with natural iron and $Mg(CH_3COO)_2.4H_2O$ as precursor powders which were mixed with 2.0 M NH_4OH at a synthesis temperature of 50, 70 and 90 °C. The material characterizations were carried out using X-Ray Diffraction (XRD), Scanning Electron Microscopy (SEM), and Vibrating Sample Magnetometer (VSM). The XRD pattern showed that $MgFe_2O_4$ has single phase and its crystallite size increased and the powder density decreased as the temperature increased. The optimum temperature for the synthesis of $MgFe_2O_4$ was found at 50 °C, when the saturation magnetization and coercivity were at the highest values of 51.73 emu/g and 308 Oe, respectively. The result also showed that $MgFe_2O_4$ as an adsorbent of Pb ions has adsorption capacity of 143.07mg/g and removal efficiency of 97.3%.

1. Introduction

Iron sand is a one of abundant natural mineral in Indonesia. Natural iron sand contains high iron (Fe) content and can be processed into magnetic material to have a high economic value. One of the magnetic materials prepared from natural iron sand is based on spinel ferrite. Spinel ferrite possess two different crystal structures: tetrahedral and octahedral. The chemical formula of the spinel ferrite is M-Fe_2O_4, where *M* is a divalent of metal ion such as Cu, Zn, Ni, Co, Fe, Mn, Mg or their mixtures. Spinel ferrite has widely known for its low coercivity as well as high permeability and resistance; but, coercivity is low when the particle size is in nanometer scale [1].

The magnesium ferrite ($MgFe_2O_4$) is one of spinel ferrites which have high saturation magnetization, Curie temperature and electrical resistivity [1]. It is also known for its unique chemical properties and thermal stability. The synthesis methods for $MgFe_2O_4$ production developed so far included sol-gel [2], hydrothermal [3], polyol, co-precipitation [4], combustion [5], and solvothermal [6]. The $MgFe_2O_4$ was categorized as an *n*-type semiconductor materials so it can be applied as adsorption, sensor, Magnetic Resonance Imaging (MRI) materials [7], magnetic separation, hyperthermia treatment [2], and drugs delivery [8, 9]. In its application as heavy metal ion adsorbents,

optimum conditions will be obtained if the material has a small particle size, large absorbent surface area, high saturation, and low coercivity [10].

In this research, the $MgFe_2O_4$ from natural iron sand was synthesized by coprecipitation method that is advantageous for its relatively simple procedure at low temperatures. The synthesis temperature was varied at 50, 70 and 90 °C. $MgFe_2O_4$ properties were analyzed by X-Ray diffraction (XRD), scanning electron microscope (SEM), and vibrating sample magnetometer (VSM).

2. Experiment method

Iron sand powder from Deli Serdang River of North Sumatra was used as raw material The precursor solution was made from the mixture of iron sand (2.72 g) and magnesium acetate tetrahydrate, $Mg(CH_3COO)_2.4H_2O$ (5.91 g), dissolved in 40 mL HCl solution (5.1 M). The solution was stirred using magnetic stirrer at 500 rpm and then filtered with whatman filter paper (125 µm diameter). Subsequently, the all solution was added dropwise into 99 ml NH_4OH 2 M solution at temperatures of 50, 70, and 90 °C, named as T50, T70, and T90 samples. The solutions were continuously stirred for 120 minutes at 500 rpm to form dark precipitate. The precipitate was washed with distilled water to reach neutral pH and dried in the oven at 100 °C to evaporate the water. The resulted $MgFe_2O_4$ powder was tested using pycnometer, X-Ray diffraction (XRD - Rigaku SmartLab, wavelength $\lambda = 1.5418$ Å), scanning electron microscope (SEM - Hitachi 3500 SU), and vibrating sample magnetometer (VSM - Electromagnetic VSM250) for the analysis of powder density, phase, microstructure, and magnetic properties respectively. The Pb ion adsorption was carried out by mixing 50 mg of $MgFe_2O_4$ powder into a 25 mL Pb ion solution in water. The mixture was then shaken with a shaker mill for 30 minutes. Finally, the ion content of Pb before and after adsorption was tested using atomic absorption spectroscopy (AAS - Shimadzu AA6800).

3. Results and discussion

Figure 1 shows diffraction pattern of the $MgFe_2O_4$ samples synthesized with various temperatures. Based on XRD database (PDF card No. 04-102-1070), all data showed that the samples has a single phase of cubic crystal structure. This observation can be used to calculate the lattice parameter and the crystallite diameter using the following equations:

$$a^2 = \frac{\lambda^2}{4sin^2\theta}(h^2 + k^2 + l^2) \quad (1)$$

$$D = \frac{K\lambda}{Bcos\theta} \quad (2)$$

In which D is crystallite diameter, a is lattice parameter, θ is angle of diffraction, h, k, and l are miller index, K is a Scherrer constant (0.9), λ = wavelength of the x-ray, and B is full width at half maximum (FWHM). The calculated lattice parameter and crystallite size were shown in table 1.

Based on table 1, it can be seen that the calculated lattice parameters for all samples are the same with those for $MgFe_2O_4$ material (8.410 Å). This result strongly confirms that the synthesized powder was $MgFe_2O_4$ with crystalline structure. Furthermore, we can also see from table 1 that the crystallite diameter tends to increase with the increase of synthesis temperature, while the crystal structure and cell volume are not affected [11, 12]. From the calculation results, the smallest crystallite size of 27.87 nm is obtained with synthesis temperature of 50 ^oC (T50). Meanwhile, an increase in crystallite size is directly proportional to the number of pores and inversely to powder density [13].

Obtained hysteresis curve from VSM measurement for all samples are shown in figure 2, and their associated magnetic properties in table 2. Table 2 shows that the coercivity and remanence increases with an increase of synthesis temperatures. The magnetic properties of material depend on the composition, crystallinity, size, and phases [11]. Smaller particle size will tend to be single domain, so that the energy barrier will be smaller. As a result, the magnetic moment will be easily switched by the

influence of the external magnetic field. When magnetic field is eliminated the magnetic moment will be more easily to return to its original position, so that the coercivity value tends to decrease that the optimum synthesis temperature is 50 °C with coercivity value of 382 Oe.

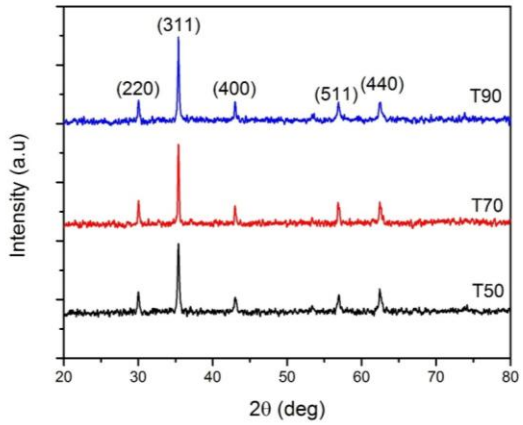


Figure 1. XRD diffraction pattern of $MgFe_2O_4$.

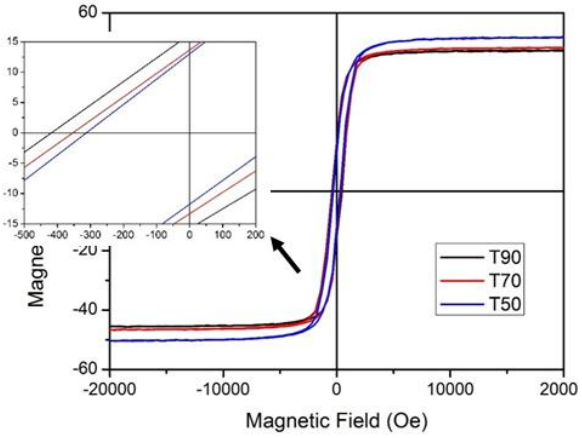


Figure 2. Hysteresis curves of $MgFe_2O_4$.

Table 1. Lattice parameter and crystallite size of $MgFe_2O_4$.

Sample	Powder density (g/cm^3)	Lattice parameter (Å)	Crytallite size (nm)
T50	4.677	8.411	27.87
T70	4.666	8.412	32.22
T90	4.482	8.411	34.30

Table 2. Magnetic properties of $MgFe_2O_4$.

Sample	Coercivity (Oe)	Saturation (emu/g)	Remanence (emu/g)
T50	308	51.73	13.24
T70	382	48.37	13.79
T90	416	47.41	16.48

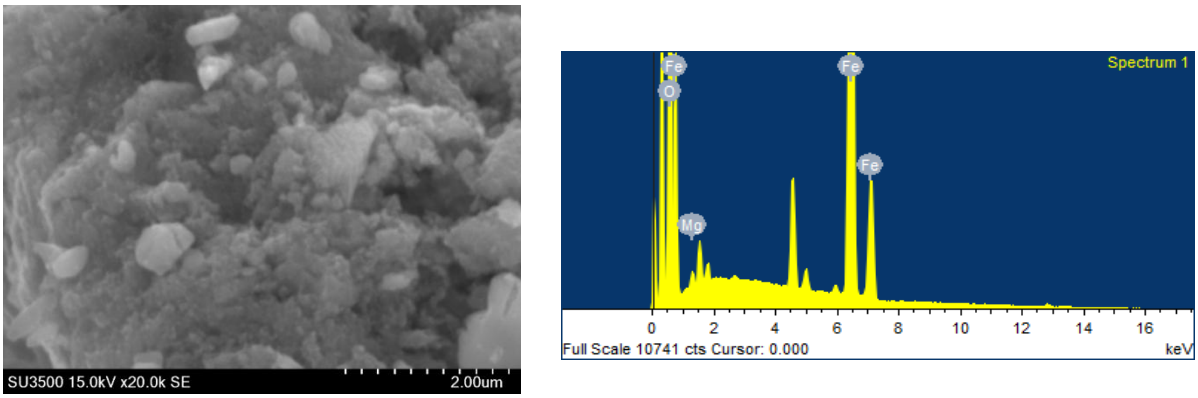
SEM image and its EDX spectra of T50 sample are shown at figures 3a and 3b, respectively. From figure 3a, it can be observed that the particle size is ranged from 0.1 to 0.8 μm and the particle tends to agglomerate. EDX spectra as shown in figure 4b revealed that the T50 sample contains Mg, Fe, and O with the weight percentages of 0.6, 69.9, and 29.5 wt%.

We used obtained $MgFe_2O_4$ powder as Pb ion adsorbent by simply mixing the powder with an artificial waste solution made from Pb salt dissolved in water. The adsorption capacity (q) and removal efficiency (%R) of the powder can be calculated by the following equations:

$$q = \frac{C_o - C_e}{W} V \tag{3}$$

$$\%R = \frac{C_o - C_e}{C_o} 100 \tag{4}$$

where C_o is concentration of Pb ion before treatment, C_e is concentration of Pb ion after treatment, W is particle weight of $MgFe_2O_4$, and V is volume of waste solution. AAS analysis obtained that the adsorption capacity decreases with increasing synthetic temperature. The highest adsorption capacity was obtained for T50 samples at 143.07 mg/g (table 3) due to its small particle size and the largest surface area (table 3). From this result, we can conclude that T50 sample has a superparamagnetic property with high saturation and small coercivity [6, 10].



(a) (b)

Figure 3. (a) SEM image and (b) its correlated EDX spectra of T50.

Table 3. Adsorption properties of $MgFe_2O_4$.

Sample Name	Syntesize temperature (˚C)	Concentration of Pb Ions, (ppm)		Adsorption capacity (mg/g)	Removal efficiency (%)
		Before treatment	After treatment		
T50	50	294	7.86	143.07	97.32
T70	70	294	34.70	129.65	88.20
T90	90	294	156.00	69.00	46 .93

4. Conclusion

Magnesium ferrite ($MgFe_2O_4$) as magnetic material has been successfully fabricated by co-precipitation method. XRD measurement revealed that the resulted $MgFe_2O_4$ powder has a single phase with spinel cubic structure. We also confirmed that the synthesis temperature was found to affect the crystallite diameter of the powder, where the smallest diameter was obtained for the sample synthesized at 50 °C. The magnetic properties of this material such as coercivity and magnetization saturation also confirmed this finding. The application of the powder as Pb ion adsorbent in the artificial waste water showed that the powder has a good adsorption capacity and high removal efficiency.

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