

Synthesis and Characterization of Manganese Ferrite (MnFe_2O_4) Nanoparticles by Coprecipitation Method at Low Temperatures

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Abstract—Research on the synthesis and characterization of manganese ferrite nanoparticles with the coprecipitation method at low temperature has been carried out. The purpose of this study was to determine the effect of low temperature on the microstructure of MnFe_2O_4 nanoparticles. The synthesis of nanoparticles carried out using the coprecipitation method, characterization using XRD and SEM. XRD measurement data were processed using *Software Origin 9.0*, obtained a crystalline phase diffraction pattern of MnFe_2O_4 and a crystal with a spinel structure. The crystallite size obtained was 5,79 nm at 90 °C, 5,65 nm at 120 °C and 5,47 nm at 150 °C with an average lattice parameter of 9,16 Å. Crystallite size can be controlled using the synthesis method at low temperatures. The surface morphology of the SEM test showed that an average grain size of 5,93 nm was spherical with quite various sizes and clots.

Keywords—manganese ferrite, coprecipitation method, XRD, SEM

I. INTRODUCTION

The nanoparticles have particle sizes in the nanometer range, with an estimate of fewer than 100 nanometers. Particle size and microstructure can be determined by analyzing nanoparticles [1]. Ferrite is a type of magnet that is made from an oxide-based material. Manganese ferrite is corrosion-resistant, chemically stable, and can be made into both hard and soft magnets [2]. Mn, Fe, Zn, Ba, Ca, Co, Ni, and Sr are the elements used as ferrite's base material [3]. Soft ferrite (high permeability and specific resistance, low coercivity), hard ferrite (high coercivity and remanence powers, hexagonal crystal structure), and garnet-structured ferrite (typically based on temperature) are the three types of ferrite [4]. Ferrite is commonly used in software such as microwave units, computer memory chips, and data storage media for drugs [5].

Manganese ferrite nanoparticles (MnFe_2O_4) are soft ferrites with higher magnetic permeability and lower resistivity than CoFe_2O_4 and NiFe_2O_4 . The synthesis of MnFe_2O_4 by coprecipitation method and magnetism characterization at low temperature (< 300 °C) and room temperature (25 °C) have

been successfully carried out to obtain the smallest grain size at room temperature, but a hematite impurity phase (Fe_2O_3) still exists [6]. Moment of magnetic attraction with a magnetic spin of 5 μB . Manganese ferrite (MnFe_2O_4) has a higher magnetic susceptibility than other ferrites such as Fe_3O_4 , CoFe_2O_4 , and NiFe_2O_4 . The milling technique was used to study the effect of La concentration on the structure and characteristics of manganese ferrite, and the wave absorption was found to be 96,50 % [7].

The results of research on manganese ferrite using different methods revealed a difference in grain size ranging from the smallest (4 nm) at 320 °C to the largest (154,1 nm) at 420 °C [6]. However, from all research studies on MnFe_2O_4 carried out at high temperature and room temperature, in this study the characterization of nanoparticles using low temperature was to determine the effect of low temperature variations on the microstructure produced in the synthesis process carried out by the coprecipitation method. Based on previous studies, synthesis and characterization of manganese ferrite (MnFe_2O_4) nanoparticles will be carried out using the coprecipitation method at low temperatures.

II. METHODS

A. Method of Coprecipitation

The nanoparticles were produced using the coprecipitation process, which involved dissolving MnCl_2 and 2FeCl_3 with a composition of 3,1 grams of Mn and 16,2 grams of Fe in 25 ml of distilled water until homogeneous. The mixture was then stirred until homogeneous when adding 3,37 ml HCl (70 %). After that, mix 8 grams of NaOH in 25 ml of distilled water until homogeneous, then drop the homogeneous mixture into the mixture of NaOH and distilled water, stirring for 1 hour at 600 rpm with a magnetic stirrer to prevent sedimentation. The sediment is washed three times to remove as much salt as possible. After washing, the precipitate will be put in a furnace for 4 hours at 90 °C, 120 °C, and 150 °C. The following is the equation for nanoparticle reaction [6]:

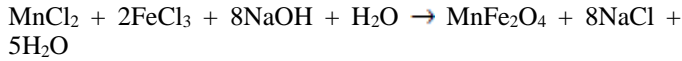


TABLE I. THE VARIANCE OF FURNACE TEMPERATURES WAS USED AS A SYNTHESIS PARAMETER FOR MnFe_2O_4 NANOPARTICLES

No	Mass MnCl_2 (g)	Mass 2FeCl_3 (g)	Furnace Temperature °C
1	3,1	16,2	90 °C
2	3,1	16,2	120 °C
3	3,1	16,2	150 °C

B. X-Ray Diffraction

XRD (X-Ray Diffraction) microstructure characterization of MnFe_2O_4 nanoparticles, including crystal structure, lattice parameters, and grain size [8]. The Scherer equation can be used to determine the grain size of nanoparticles [9].

$$t = \frac{k\lambda}{B \cos\theta} \tag{1}$$

Where t is the grain size, k is the Scherer constant (0,89), λ is the wavelength, B is the width of the half crest, and θ is the angle [10].

C. Scanning Electron Microscopy (SEM)

SEM was used to analyze and quantify the nanoparticles surface morphology. SEM is used to determine particle size and examine the surface morphology of nanoparticles [11]. The operating theory of a scanning electron microscope (SEM) allows it to fire high-energy electron beams into the surface of materials. The reflection of the beam from the surface of the surfaces causes electron reflections. The scattered electrons will reveal details about the sample's state, such as the nanoparticles' surface shape.

III. RESULTS AND DISCUSSION

A. Diffraction Pattern

The furnace result of MnFe_2O_4 nanoparticles with temperature variations is shown below.

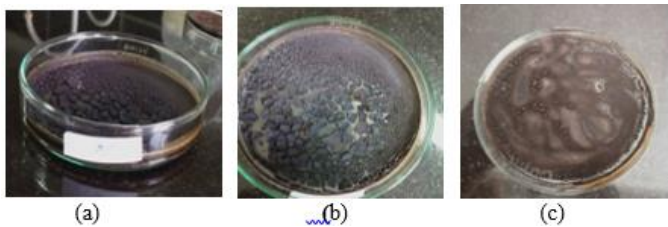


Fig. 1. Sample varie of temperature (a) 90° C (b) 120° C (c) 150° C.

The perfect characterization was obtained after a four-hour furnace operation. The furnace results were crushed with a spatula before being analyzed with XRD.

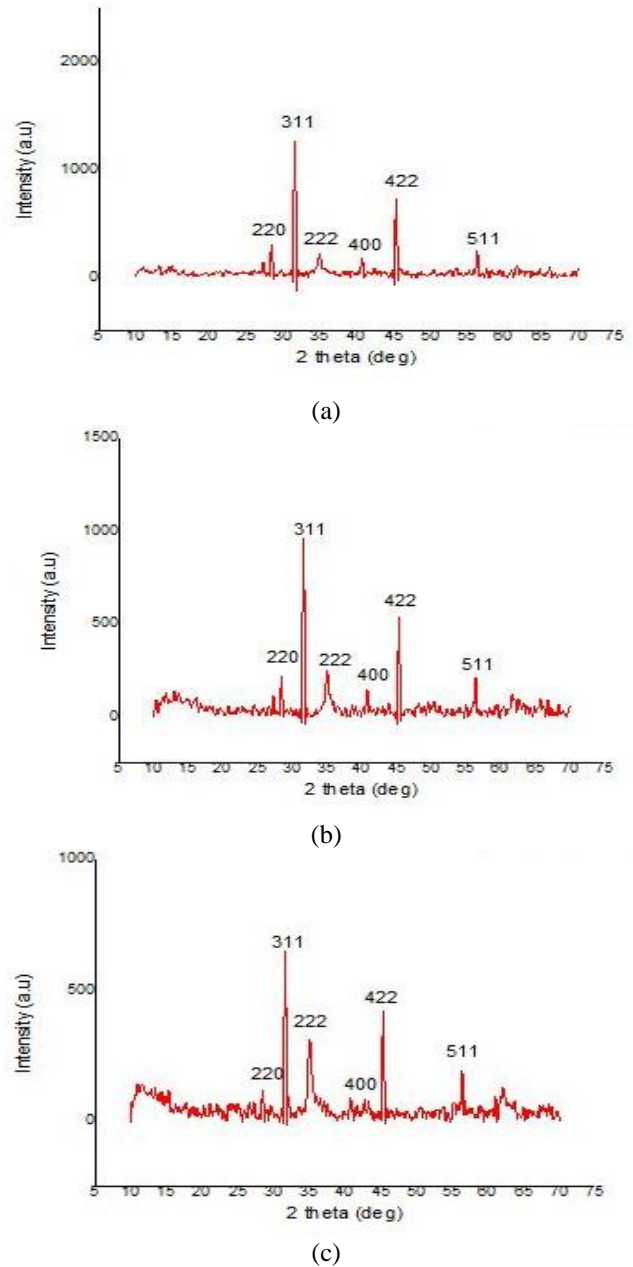


Fig. 2. a) MnFe_2O_4 diffraction pattern at temperature 90 °C, b) MnFe_2O_4 diffraction pattern at temperature 120 °C, c) MnFe_2O_4 diffraction pattern at temperature 150 °C.

The diffraction pattern of MnFe_2O_4 at temperature variations can be seen from Figure 2.a, 2.b, and 2. c above. XRD data was processed using *origin software 9.0*. The crystalline phase of MnFe_2O_4 appears around angle 2θ with the peaks and the hkl plane. The diffraction peaks at angles (2θ) = 28,4°, 31,6°, 35,0°, 40,7°, 45,3°, and 56,4°. The lattice parameters values obtained were 9,02 Å, 9,43 Å, 9,16 Å, 9,04

Å, 9,87 Å, and 8,45 Å. The hkl fields formed include 220, 311, 222, 400, 422, and 511 [12]. The diffraction pattern above shows that the MnFe₂O₄ nanoparticles are crystals in the cubic process with a spinel structure and the Fd3m space group (JCPDS No 75-0034) [13]. Since the coprecipitation approach is used, the sharpness of the diffraction peaks on the XRD test results graph is noticeable. According to the XRD test, there is an impact on shifts in manganese ferrite nanoparticles, as evidenced by clearer and sharper diffraction peaks. The formation of MnFe₂O₄ nanoparticles with strong crystallinity and no impurity phases was demonstrated by XRD findings. The following table 2-4 is a table of XRD data processing at 90 °C, 120 °C, and 150 °C:

TABLE II. XRD DATA PROCESSING AT A TEMPERATURE OF 90 °C

Hkl. Field	Lattice Parameters (Å)	Crystallite Size (nm)	Distance Between Atoms (nm)
220	9,02	6,57	0,31
311	9,43	5,52	0,28
222	9,16	2,7	0,24
400	9,04	5,3	0,22
422	9,87	6,27	0,2
511	8,45	6	0,14

TABLE III. XRD DATA PROCESSING AT A TEMPERATURE OF 120 °C.

Hkl. Field	Lattice Parameters (Å)	Crystallite Size (nm)	Distance Between Atoms (nm)
220	9,02	6,27	0,31
311	9,43	5,52	0,28
222	9,16	2,87	0,24
400	9,04	6	0,22
422	9,87	5,75	0,2
511	8,45	5,3	0,14

TABLE IV. XRD DATA PROCESSING AT A TEMPERATURE 150 °C.

Hkl. Field	Lattice Parameters (Å)	Crystallite Size (nm)	Distance Between Atoms (nm)
220	9,02	6,27	0,31
311	9,43	5,52	0,28
222	9,16	2,5	0,24
400	9,04	5,3	0,22
422	9,87	5,75	0,2
511	8,45	5,11	0,14

At 90 °C, the MnFe₂O₄ sample has the highest degree of crystallinity. Microstructure measurements of MnFe₂O₄ nanoparticles revealed that using low temperatures to produce perfect crystallinity was more effective. Figure 3.a and 3.b show a graph of the relationship between temperature, crystallite size, and lattice parameter:

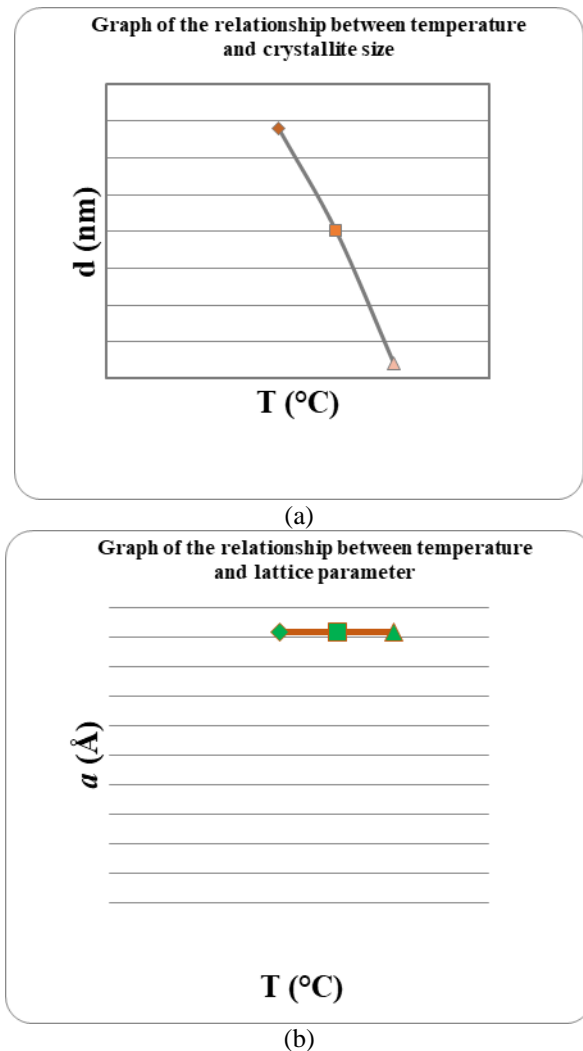


Fig. 3. a) Graph of the relationship between temperature and crystallite size, b) Graph of the relationship between temperature and lattice parameter.

The crystallite size is affected by temperature, as shown in Figure 3.a and 3.b. Due to the effect of the furnace drying temperature, the crystallite size obtained decreases. The smaller the crystallite size, the higher the temperature, and the larger the crystallite size, the lower the temperature. The lattice parameters produced at each low temperature, however, are the same: 9,16 Å. This result shows that temperature does not affect the lattice parameter values. The Scherer equation is used to generate the lattice parameters, which are based on the hkl field specified by the JCPDS data.

B. Nanoparticle Surface Morphology

The resulting particles have a spherical morphology and differ in size, according to the surface morphology obtained from the SEM results at a magnification of 5.00 k. SEM (Scanning Electron Microscopy) images of the surface morphology of MnFe₂O₄ nanoparticles are shown below:

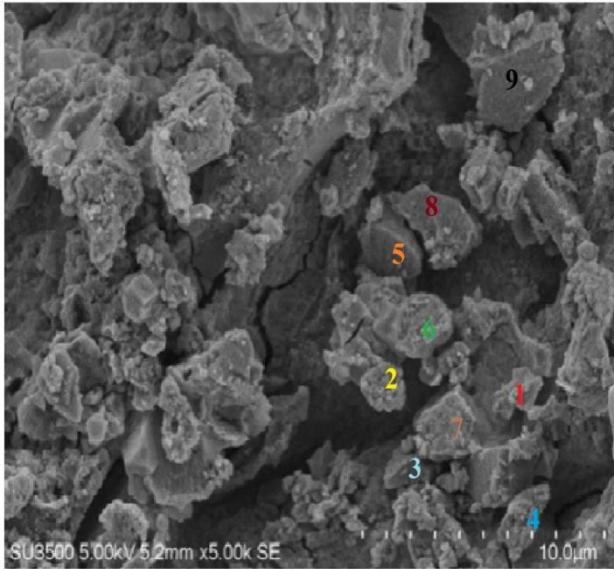


Fig. 4. Morphology of MnFe₂O₄ with magnification of 5.00 k.

TABLE V. THE RESULTS OF TESTING MNFe₂O₄ NANOPARTICLES USING SEM.

No	Grain Size
1	4,33
2	5,15
3	5,34
4	5,56
5	6,07
6	6,27
7	6,43
8	6,46
9	7,78
Average	5,93

Table 4 shows the effects of using *Image J Software* to analyze MnFe₂O₄ nanoparticles; the average grain size is 5,93 nm. Low temperatures cause the particle size to become clumpy and irregular, while high temperatures cause the particle size to become clumpy and irregular. MnFe₂O₄ nanoparticles have a non-uniform morphology and agglomerate at low temperatures. The nanoparticles have agglomerated or clumped together, causing the particle size to appear irregular and dispersed in SEM images. The results of the SEM test shown in Figure 4 show that agglomeration/clumping causes the particle to appear larger. The joining of small particles into larger structures can cause agglomeration. Because of the high rotational speed, the agglomeration mechanism occurs. Because of the high rotation speed, the kinetic energy of the particles increases, making particle collisions lighter and allowing materials to interact and combine to form larger agglomerations. Agglomeration happens when small particles are joined together to form a larger structure. The agglomeration process occurs as a result of the coprecipitation method's effect on high-speed rotation in the synthesis process [14].

IV. CONCLUSION

The coprecipitation method was used to make nanoparticles, with the addition of NaOH as a precipitate and HCl to speed up the reaction. MnFe₂O₄ nanoparticles with crystallinity and no impurity phases are formed using this process. 220, 311, 222, 400, 422, and 511 were developed as hkl fields. At low temperatures, the synthesis of MnFe₂O₄ nanoparticles is more effective. The crystals formed have a spinel structure and belong to the space group Fd3m in the cubic process. MnFe₂O₄ nanoparticles have a non-uniform morphology and agglomerate at low temperatures.

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REFERENCES

- [1] F. Malega, I.P.T. Indrayana, and E. Suharyadi, "Synthesis and Characterization of the Microstructure and Functional Group Bond of Fe₃O₄ Nanoparticles from Natural Iron Sand in Tobelo North Halmahera," *J. Ilm. Pendidik. Fis. Al-Biruni*, vol. 7, no. 2, p. 129, 2018.
- [2] T. Shanmugavel, S. G. Raj, G. R. Kumar, and G. Rajarajan, "Synthesis and Structural Analysis of Nanocrystalline MnFe₂O₄," *Phys. Procedia*, vol. 54, pp. 159–163, 2014.
- [3] M. Goodarz Naseri, E.B. Saion, and A. Kamali, "An Overview on Nanocrystalline ZnFe₂O₄, MnFe₂O₄, and CoFe₂O₄ Synthesized by a Thermal Treatment Method," *ISRN Nanotechnol.*, vol. 2012, pp. 1–11, 2012.
- [4] S. Xavier, S. Thankachan, B.P. Jacob, and E.M. Mohammed, "Structural and electrical properties of neodymium substituted cobalt ferrite nanoparticles," *IOP Conf. Ser. Mater. Sci. Eng.*, vol. 73, no. 1, pp. 64–67, 2015.
- [5] N.M. Deraz and A. Alarifi, "Controlled synthesis, physicochemical and magnetic properties of nano-crystalline Mn ferrite system," *Int. J. Electrochem. Sci.*, vol. 7, no. 6, pp. 5534–5543, 2012.
- [6] R.D. Tawainella, Y. Riana, R. Fatayati, T. Kato, and S. Iwata, "Sintesis Nanopartikel Manganese Ferrite (MnFe₂O₄) dengan Metode Kopresipitasi dan Karakterisasi Sifat Kemagnetannya," vol. XVIII, no. April, pp. 1–7, 2014.
- [7] D. Nurhayani, "1 Efek Konsetrasi La Terhadap Struktur dan Karakteristik Serapan Gelombang Mikro Sistem MnLa," vol. 4, 2019.
- [8] U. Shankar Sharma, R. Naresh Sharma, and R. Shah, "Physical and Magnetic Properties of Manganese Ferrite Nanoparticles," *J. Eng. Res. Appl. www.ijera.com*, vol. 4, no. 8, pp. 14–17, 2014.
- [9] M. Ahmed, A. Anwar, and S.A. Ahmad, "a Literature Review on Study of Concrete Strength Using Partial Replacement of Cement With Rice Husk Ash and Fine Aggregate With Ceramic Powder," *Int. J. Recent Sci. Res.*, vol. 9, no. 3, pp. 23083–23086, 2018.
- [10] B.B. Dora, S. Kumar, R.K. Kotnala, B.C. Raulo, and M.C. Sahu, "Improved magnetic properties of Ni-Zn nano ferrites by using aloe vera extract solution," *Int. J. Pharm. Sci. Rev. Res.*, vol. 30, no. 1, pp. 294–298, 2015.
- [11] P.M. Tamhankar, A.M. Kulkarni, and S.C. Watawe, "Functionalization of Cobalt Ferrite Nanoparticles with Alginate Coating for Biocompatible Applications," *Mater. Sci. Appl.*, vol. 02, no. 09, pp. 1317–1321, 2011.

- [12] S. Pande, M.M. Islam, S.C. Mohanta, and N. Uddin, "Single-Step Synthesis of Manganese Ferrite Nanoparticles with Enhanced Magnetization via Chemical Co-precipitation Route," *J. Sci. Res.*, vol. 11, no. 2, pp. 225–234, 2019.
- [13] S.H. Kareem, Y.K. Ooi, S.S. Abdulnoor, M. Shamsuddin, and S. L. Lee, "Influence of zinc on the structure and morphology of manganese ferrite nanoparticles," *J. Teknol. (Sciences Eng.)*, vol. 69, no. 5, pp. 103–106, 2014.
- [14] S.D.W.I. Prasetyo, P. Studi, T. Mesin, F. Teknik, and U. M. Surakarta, "Kajian Produksi Nanopartikel dari Arang Bambu Dengan Peningkatan Energi Tumbukan Bola Baja Diameter 5 / 32 Inchi," 2018.