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Magnetic Properties of Manganese Ferrite (MnFe2O4) by Co-Precipitation Method with Different pH Concentration

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DETERMINATION OF THE MAGNETIC PROPERTIES OF MANGANESE FERRITE BY THE COPRECIPITATION METHOD AT DIFFERENT pH CONCENTRATIONS

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Manganese ferrite ($MnFe_2O_4$) as a magnetic material was greatly affected by several parameters formed during the synthesis process. One of the parameters necessary in the coprecipitation synthesis process is the pH concentration in the titration process. The pH variations can affect the magnetic properties of $MnFe_2O_4$, and it was very interesting to review the effect of pH on its magnetic properties of . Manganese ferrite was synthesized by using the coprecipitation method with pH being equal to 8, 10, and 12 and sintered at a temperature of 1000°C for 3 h. Manganese ferrite nanopowder that had been synthesized was analyzed by using XRD, SEM, and VSM. The characterization process was conducted with both sintered and unsintered manganese ferrite. The XRD results indicated that the unsintered sample at all values of pH was amorphous, while the sintered samples at all pH values showed that a single phase was formed in a lattice with dimensions 59.48 nm, 41.60 nm, and 29.72 nm for pH equal to 8, 10, and 12. The SEM testing showed that the structure of all samples was homogeneous. Although the bulk size of unsintered samples was smaller and more homogeneous than that of sintered samples, the results showed that all unsintered samples had similar values and superparamagnetic properties. However, unlike the sintered sample, which was much more homogeneous, there was transition from the superparamagnetic properties to paramagnetic properties at pH equal to 12. Thus, it was found that the higher the pH level, the smaller the crystallite size.

KEY WORDS: *manganese ferrite, magnetic nanomaterial, coprecipitation, acidity, phase, morphology, and magnetic properties*

1. INTRODUCTION

Researching into nanomaterials is sufficiently interesting for scientists and engineers. Magnetic nanomaterials arosed interest due to their properties to be applied in various fields, such as sensors, magnetic fluid in storage media, catalysts, pigments, magnetic recording media, retrieval of information of magnetic resonance imaging (MRI), and others (Akhtar and Younas, 2012).

The nanostructure of oxide magnetic transition with spinel structure MFe_2O_4 (M = Mn, Fe, Co, Ni) has unique magnetic properties, such as medium strong magnetization, high coercivity, single domain effect, superparamagnetism, and others that led to their application in industry and biology. As an important part of ferrite group, MFe_2O_4 had attracted the researcher's attention due to its high magnetic and electromagnetic properties. MFe_2O_4 showed different properties in the form of nanoparticles, nanostructures, and thin films, such as constant anisotropy, magnetization saturation dependent on the size, superparamagnetism, and its high Curie temperature (Zipare et al., 2015).

The synthesis method which was quite effective and relatively simple was the coprecipitation method (Zhang et al., 2007). This method was one of the synthesis methods of inorganic compounds based on sedimentation of more than one substance when passing through the saturation point. Another advantage of this method was that the process could be conducted at room temperature, and the particle size could be controlled easily, so that this process took shorter time. Therefore, the coprecipitation method in this study was used to synthesize magnetic nanoparticles of manganese ferrite (MFe_2O_4) (Zhang et al., 2007).

During the synthesis process of the coprecipitation method, the pH in a solution needs to be adjusted (Ramadan et al., 2011). The adjustment of pH in the process of synthesizing magnetic nanoparticles by the coprecipitation method would affects the size and structure of particles. Thus, the adjustment of pH was one of the decisive factors to determine the size of magnetic nanoparticles obtained by the coprecipitation method. Thus, in this study, the effect of different pH concentrations during titration process will be observed.

2. METHODOLOGY

This study relates to experimental research to obtain descriptive data on the characteristics of phase, morphology, and magnetic properties of manganese ferrite powders using coprecipitation method. The materials used in this study were manganese oxide and iron oxide, as well as ethylene glycol used as a solvent. Independent variables in this study were pH values varying by adding sodium hydroxide pH 5M to reach pH equal to 8, 10, and 12, and the sintering process was conducted at a temperature of 1000ºC for 3 h. After the synthesis process, samples were tested using different methods of analysis: X-ray difraction (XRD) [X'pert Pro, PANalytical], scanning electron microscopy (SEM) [Phenom], and vibrating sample magnetometer (VSM) [OXFORD, type 1.2H]. Moreover, the Scherrer equation was used to get the value of crystallite size of manganese ferrite nanoparticles (Yahya et al., 2012; Yahya and Puspitasari, 2012, 2013):

$$d = \frac{K\lambda}{\beta \cos\theta},$$

where d is the crystallite diameter (Å), K is constant equal to 0.89, λ is the wave length equal to 1.5406 Å, and β is the FWHM (rad).

3. RESULTS AND DISCUSSION

3.1 X-Ray Diffraction Analysis

The result of the XRD analysis of manganese ferrite nanoparticles with different values of pH in sintered and unsintered conditions is given in Fig. 1. According to Fig. 1, the unsintered samples did not have any peak, which meant that they are amorphous, while the sintered samples had several peaks, with the highest spinel cubic-shaped one located at 311, which means a single phase is formed during the sintering process (Caballero et al., 2008). The height of the main peak is found in the sintered sample at pH 8, which had the highest intensity and differed greatly from other samples. This indicated that the sintered sample at pH 8 had the highest crystallinity degree. Where the height of the crystallinity degree was affected by the nucleation process the crystal grew rapidly and simultaneously as the result of the high saturation degree of the solution with a reducing pH in the synthesis process.

Table 1 presents the results of the XRD analysis at the 311 peak at pH equal to 8, 10, and 12. After calculating the XRD results by using the Scherrer equation it was obtained that the

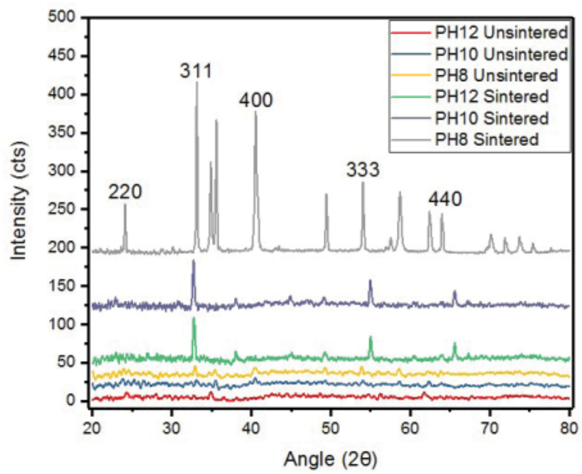

FIG. 1: X-ray diffractogram of $MnFe_2O_4$

TABLE 1: XRD analysis

No.	pH	Pos. [° 2Th.]	Height [Cts]	FWHM [° 2Th.]	*d*-Spacing [Å]	Crystallite Size (nm)
1	8	33.1157	270.70	0.1378	2.70520	59.48
2	10	32.7036	67.38	0.1968	2.73834	41.60
3	12	32.7505	49.64	0.2755	2.73453	29.72

sample at pH 8 had a crystallite size of 59.48 nm, the sample at pH 10 had a crystallite size of 41.60 nm, and the sample at pH 12 had a crystallite size of 29.72 nm. Thus, based on the the information presented in the table it can be inferred that the higher the pH value, the smaller is the crystallite obtained.

There were other peaks found in epy sintered sample at pH 8 besides the peak which was characteristic of $MnFe_2O_4$, because of the presence of impurities as a result of the oxidation reaction between the sample ions and oxygen ions.

3.2 Scanning Electron Microscopy Analysis

Figure 2 shows the results of the SEM analysis of all samples. SEM testing is aimed at analyzing and comparing the morphology of each manganese ferrite sample. The magnification used in this test was 50,000×. Figure 2A presents the result of analysis for an unsintered sample at pH 8, Fig. 2B—for an unsintered sample at pH 10, Fig. 2C—for an unsintered sample at pH 12, while Fig. 2D presents the result for a sintered sample at pH 8, Fig. 2E—for a sintered sample at pH 10, and Fig. 2F—for a sintered sample at pH 12.

Figure 2 shows that the unsintered samples had a smaller crystal size than the sintered ones. During the process of sintering, the particles are rearranged to bind with one another and form

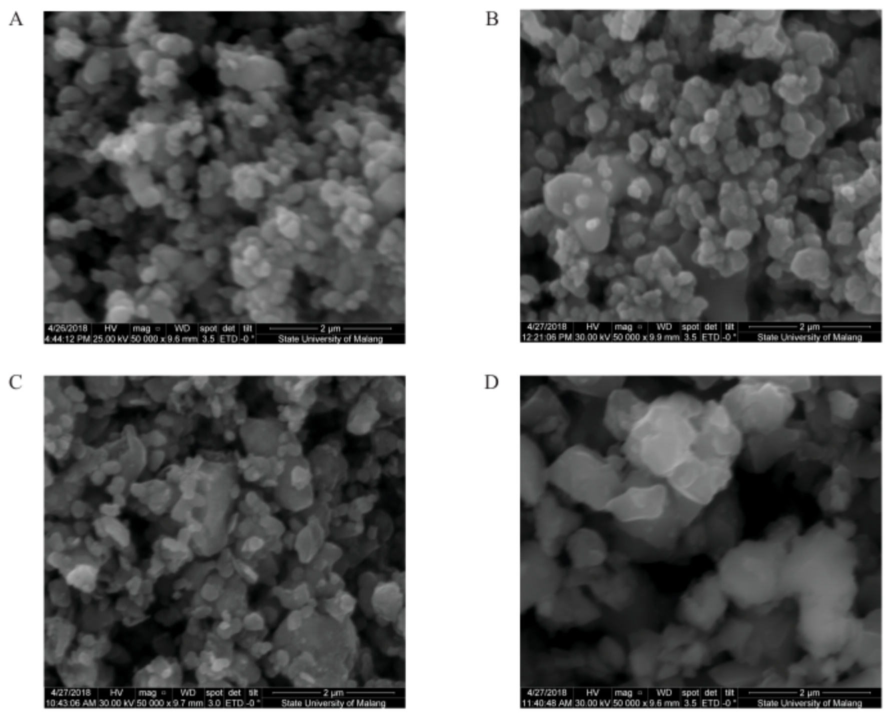

FIG. 2.

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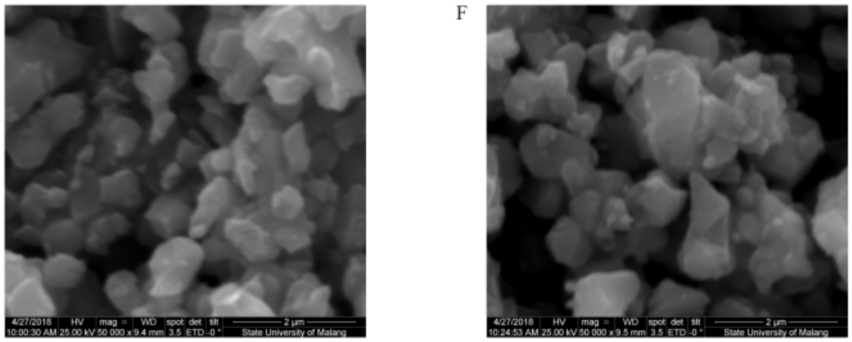

FIG. 2: Morphology of $MnFe_2O_4$ (unsintered and sintered) at different pH values

a single phase in a new lattice structure (Shanmugavel et al., 2014). Besides, the increase of pH value leads to a smaller particle size. Furthermore, it also shows that the unsintered samples were more homogeneous than the sintered samples.

3.3 Vibrating Sample Magnetometer Analysis

According to Fig. 3, it can be inferred that the results of the VSM analysis of unsintered samples at pH 8, 10, and 12, were not significantly different. Based on Ms, Mr, and Mc given

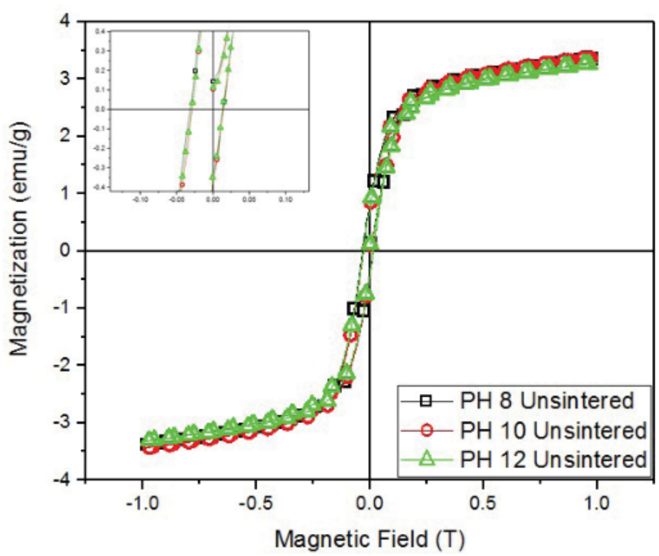

FIG. 3: Hysteresis curve for unsintered samples

in Table 2, it is also shown that all unsintered samples were identical and had the same properties, and their hysteresis loop showed that this material belongs to superparamagnetic materials (Puspitasari, 2016). Thus, variations of pH did not significantly affect unsintered samples. This is due to the fact that, on the one hand, these materials were amorphous, on the other hand, there was no single phase particles and crystallites that formed in the lattice.

Figure 4 shows the significant increase in the magnetic saturation. The sintered sample at pH 8 (Fig. 5) had Ms value of 52.776 emu/g which was the highest value compared with other samples. The Mr value in this sample was 16.350 emu/g, which was also the highest value among all samples. However, the Mc value in this sample was smaller than for the unsintered sample that was 0.021 T. While the sintered sample at pH 10 (Fig. 6) showed that it had a lower value of magnetic saturation, magnetic retentivity, and magnetic coersivity compared with the sintered sample at pH 8 and all unsintered samples, the values sequentially

TABLE 2: Hysteresis curve analysis for unsintered samples

No.	Sample	Ms (emu/g)	Mr (emu/g)	Mc (T)
1	Unsintered, pH 8	3.370	0.800	0.031
2	Unsintered, pH 10	3.400	0.700	0.030
3	Unsintered, pH 12	3.300	0.710	0.030

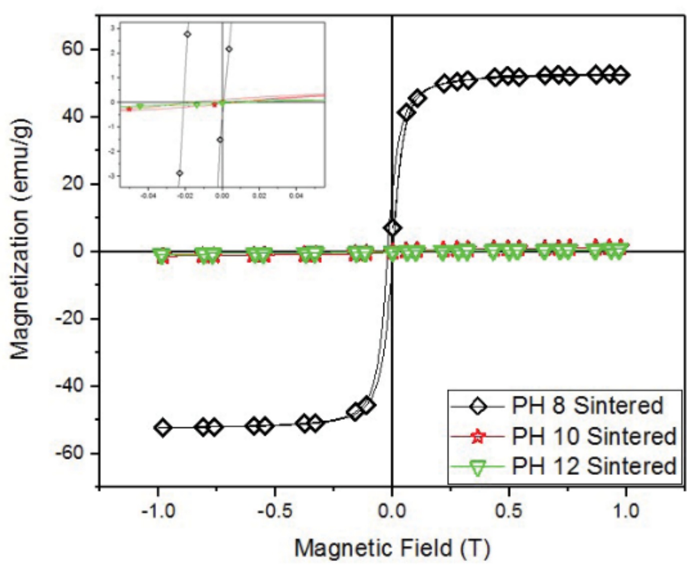

FIG. 4: Hysteresis curve for sintered samples

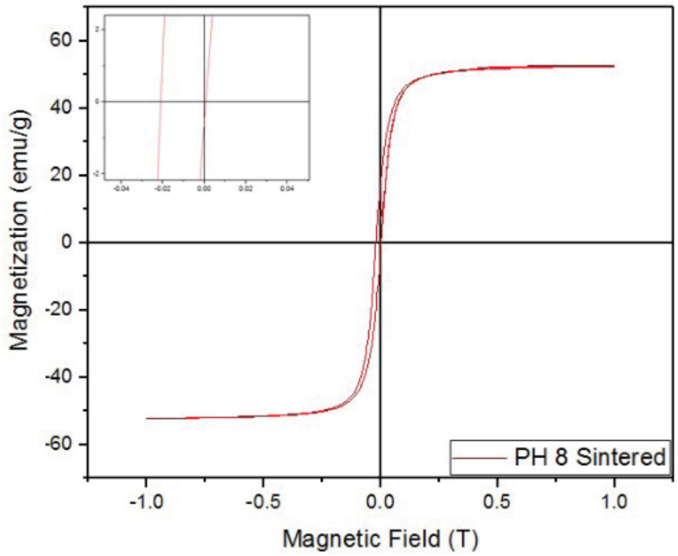

FIG. 5: Hysteresis curve for a sintered sample at pH 8

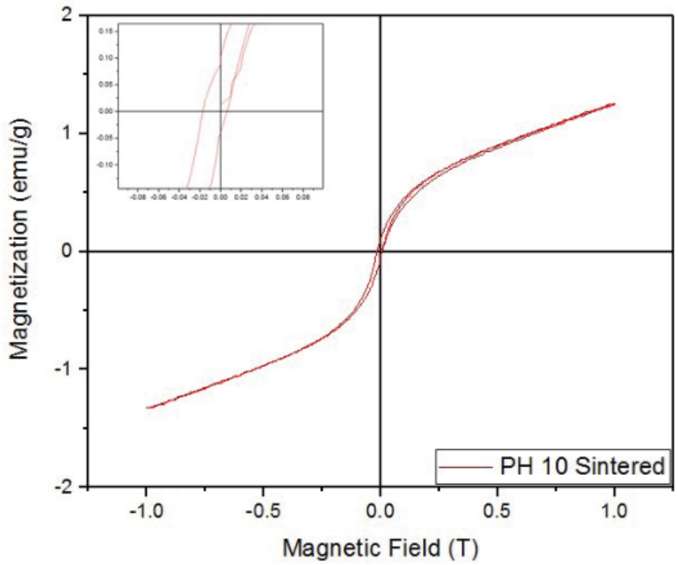

FIG. 6: Hysteresis curve for a sintered sample at pH 10

were 1.260 emu/g, 0.104 emu/g, and 0.017 T. This was due to the fact that its crystallite size was smaller. It also had a very small value when viewed in terms of superparamagnetism. However, it had a high value among paramagnetic materials. The sintered sample at pH 12 (Fig. 7) was a sample which experienced a change in the properties; this was the change from superparamagnetic to paramagnetic properties (Tauxe, 1998). However, the values of magnetic saturation, magnetic retentivity, and magnetic coercivity were much smaller for all the samples, the values sequentially were 0.706 emu/g, 0.022 emu/g, and 0.011 T. This was due to its crystallite size. The higher the pH value determined, the smaller the crystallite size obtained (Tawainella et al., 2014; Razak et al., 2012).

Table 3 shows the value of pH which indicated the reduction in the size of particles. This caused the decrease in the magnetic resistance properties through the magnetic saturation, magnetic retentivity, and magnetic coercivity. It could be concluded that an increase in pH will lead to smaller magnetic resistance properties (Ramadan et al., 2011). There occurred the

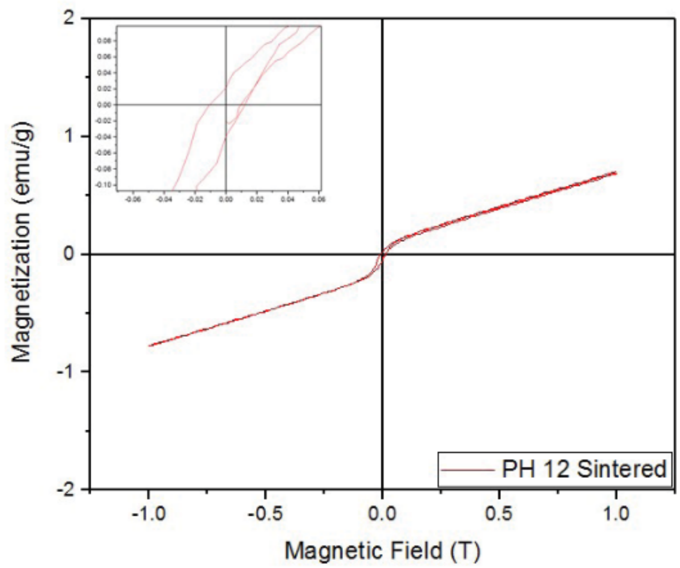

FIG. 7: Hysteresis curve for a sintered sample at pH 12

TABLE 3: VSM analysis for sintered samples

No.	Sample	Crystallite Size (nm)	Ms (emu/g)	Mr (emu/g)	Mc (T)
1	Sintered at pH 8	59.48	52.776	16.350	0.021
2	Sintered at pH 10	41.60	1.260	0.104	0.017
3	Sintered at pH 12	29.72	0.706	0.022	0.011

change in the properties from superparamagnetic ones to paramagnetic in a sintered sample at pH 12 caused by the magnetic resistance which is smaller and the Mn elements are less capable of maintaining the magnetic resistance properties of Fe in very small particles (Ramadan et al., 2011). In addition, the impurities might also be an influential factor for the increasing of pH, the greater amount of impurities was also capable of affecting the test (Tawainella et al., 2014). Furthermore, when comparing sintered samples with unsintered ones, it can be inferred that there is a lot of changes in the samples after they had been sintered. The XRD results showed that the unsintered samples were amorphous in contrast to sintered ones. The sintered samples had varied resistance properties and even there were changes in the properties. The sintering process forms a single phase. In this study, it could be proved that a single phase was formed when Mn and Fe_2O_4 were in a lattice.

4. CONCLUSIONS

Based on the phase identification conducted in XRD analysis, it was shown that unsintered samples at various pH values were amorphous, while the sintered samples at various pH values formed a single phase, and there was the correlation that the higher the pH level, the smaller the crystallite size obtained. SEM testing showed that all samples were homogeneous, although the bulk size of unsintered samples were smaller and more homogeneous than sintered samples. This was due to the sintering process that rearranged the particles binding them and then forming a single phase in a new lattice structure. Therefore, it made its bulk size increased.

The VSM analysis showed that all unsintered samples had a similar value and had superparamagnetic properties. However, unlike the sintered samples which had much more various properties, they change from being superparamagnetic to paramagnetic at pH equal to 12. Then, the magnetic resistance of sintered samples would be smaller with increase in the pH of the samples.

REFERENCES

Akhtar, M.J. and Younas, M., Structural and Transport Properties of Nanocrystalline $MnFe_2O_4$ Synthesized by Coprecipitation Method, *Solid State Sci.*, vol. **14**, no. 10, pp. 1536–1542, 2012. DOI: 10.1016/j.solidstatesciences.2012.08.026

Caballero, F.G., Miller, M.K., and Capdevila, C., Phase Transformation Theory: Advanced Steels, *Solid State Phase Transform.*, vol. **60**, no. 12, pp. 16–21, 2018.

Puspitasari, P., Green Ammonia Synthesis using Nanocatalysts in a Novel Magnetic Induction Method (MIM), Thesis, Unpublished, University Technology Petronas, 2016.

Ramadan, W., Kareem, M., Hannoyer, B., and Saha, S., Effect of pH on the Structural and Magnetic Properties of Magnetite Nanoparticles Synthesised by Coprecipitation, *Adv. Mater. Res.*, vol. **324**, pp. 129–132, 2011. DOI: 10.4028/www.scientific.net/AMR.324.129

Razak, J.A., Sufian, S., Ku Shaari, K.Z., Puspitasari, P., Hoe, T.K., and Yahya, N., Synthesis, Characterization and Application of $Y_3Fe_5O_{12}$ Nanocatalyst for Green Production of NH_3 Using Magnetic Induction Method (MIM), in *Proc. AIP Conf.*, vol. **1482**, pp. 633–638, 2012.

Shanmugavel, T., Raj, S.G., Kumar, G.R., and Rajarajan, G., Synthesis and Structural Analysis of Nanocrystalline $MnFe_2O_4$, *Physics Procedia*, vol. **54**, pp. 159–163, 2014. DOI: 10.1016/j.phpro.2014.10.053

Tauxe, L., *Palaeomagnetic Principles and Practice*, Series *Modern Approaches in Geophysics,* vol. **17**, The Netherlands: Springer, 1998.

Tawainella, R.D., Riana, Y., Fatayati, R., Kato, T., and Iwata, S., Sintesis Nanopartikel Manganese Ferrite ($MnFe_2O_4$) dengan Metode Kopresipitasi dan Karakterisasi Sifat Kemagnetannya, vol. **18**, April, pp. 1–7, 2014.

Yahya, N., Puspitasari, P., Koziol, K., and Pavia, G., New Approach to Ammonia Synthesis by Catalysis in Magnetic Field, *J. Nano Res.*, vol. **16**, pp. 119–130, 2012. DOI: 10.4028/www.scientific.net/JNanoR.16.119

Yahya, N. and Puspitasari, P., $Y_3Fe_5O_{12}$ Nanocatalyst for Green Ammonia Production by Using Magnetic Induction Method, *J. Nano Res.*, vol. **21**, pp. 131–137, 2012. DOI: 10.4028/www.scientific.net/JNanoR.21.131

Yahya, N. and Puspitasari, P., Hardness Improvement of Dental Amalgam Using Zinc Oxide and Aluminum Oxide Nanoparticles, *Characteriz. Develop. Biosyst. Biomater.*, vol. **29**, pp. 9–32, 2013. DOI: 10.1007/978-3-642-31470-4

Zhang, B., Tang, G., Yan, Z., Wang, Z., Yang, Q., and Cui, J., Synthesis of Magnetic Manganese Ferrite, *J. Wuhan Univ. Technol., Mater. Sci. Ed.*, vol. **22**, no. 3, pp. 514–517, 2007. DOI: 10.1007/s11595-006-3514-3

Zipare, K., Dhumal, J., Bandgar, S., Mathe, V., and Shahane, G., Superparamagnetic Manganese Ferrite Nanoparticles: Synthesis and Magnetic Properties, *J. Nanosci. Nanoeng.*, vol. **1**, no. 3, pp. 178–182, 2015.

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