The Influence of Alternating Magnetic Field Frequency on Magneto-Thermal Behavior of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI Material

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Abstract. Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material has been successfully synthesized using in-situ polymerization method. Meanwhile, Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles based on local sand was synthesized using the coprecipitation method. The Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI sample was characterized using the instrument of X-Ray Diffraction (XRD), Fourier Transform Infrared (FTIR), Transmission Electron Microscopy (TEM), and Magneto-Thermal Test to know the structure of the material, functional group of the material, size morphology of the material, and the specific absorption rate (SAR) value. The XRD pattern of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material was dominated by an amorphous structural pattern. The domination of PANI polymer in the Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material implicated to the decrease in the intensity of Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticle peak. The existence of PANI polymer also impacted on the amount of aggregation of the Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles so that the size of the Mn$_{0.25}$Fe$_{2.75}$O$_4$ particles increased from 8.4 nm (without PANI) to 30.4 nm (with PANI). Moreover, the test result using magneto-thermal instrument showed that the bigger the frequency of the alternating magnetic field, the obtained SAR value of the Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material would be bigger as well. For the frequency of 483 and 553 Hz, the SAR values of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material was 0.81 and 3.65 W/g, respectively. Based on the SAR results, the Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material has the potential to be the media of drug delivery for cancer therapy.

Keywords: Mn$_{0.25}$Fe$_{2.75}$O$_4$, PANI, SAR, frequency

1. Introduction
Magnetite (Fe$_3$O$_4$) is one of the materials that can be synthesized from the local iron sand and the combination of ferrous chloride (FeCl$_2$) solution and ferric chloride FeCl$_3$ solution through coprecipitation method [1], hydrothermal [2], sonochemistry [3], sol-gel [4], microwave [5] and the others. Many pieces of research have been reported so far regarding the development of magnetic material based on Fe$_3$O$_4$ [5–8]. The use and development of Fe$_3$O$_4$ material can be applied in many fields such as a biomedical field. In the biomedical field, Fe$_3$O$_4$ material can be modified from powder to liquid magnetic [10] and gel magnetic [11]. From the modification of such synthesis and fabrication,
the Fe₃O₄ material has the potential as the material for drug delivery system, magnetic resonance imaging (MRI), and hyperthermia therapy [12].

To know the characteristic of a certain material as the main candidate for hyperthermia therapy, the calculation of specific absorption rate (SAR) of that material is necessary through its magneto-thermal behavior [12–14]. Some researchers have successfully characterized SAR of a certain material B through some variations of frequencies of the alternating magnetic field. Kekalo et al. [15] reported that the SAR values of gamma-Fe₂O₃ particles were in the range of 22–200 W/g with the alternating magnetic field of about 100–500 Oe and in the frequency of 160 kHz. Meanwhile, Sathya et al. [16] successfully characterized Fe₃O₄ nanocluster magnetic through microwave method and obtained the SAR value of 215 W/g with the frequency of alternating magnetic field of 126 kHz. Meanwhile, Shete et al. [17] conducted a comparison of the SAR values obtained between the Fe₃O₄ nanoparticles without a coating and the Fe₃O₄ nanoparticles with chitosan coating. The result showed that the SAR value of Fe₃O₄ nanoparticles with chitosan coating (118.85 W/g) was higher than Fe₃O₄ nanoparticles without (79.32 W/g). That magneto-thermal characterization used magnetic field induction in the range of 167.6–335.2 Oe. However, the study on magneto-thermal of the Fe₃O₄ nanoparticles doped with Mn as well as coated with PANI polymer is rarely reported. Thereby, the study on the magneto-thermal behavior Mn₀₂₅Fe₂₇₅O₄@PANI material needs to be conducted.

The substitution of Mn atom in the Fe₃O₄ material was done to obtain better magnetic properties with the optimal saturation magnetization value such as the research reported by Taufiq et al. [18]. They doped Mn in the Mn₀ₓFe₃₋ₓO₄ material with the composition of x = 0, 0.25, 0.5, 0.75, and 1. From that study, the highest saturation magnetization value was in the composition of x = 0.25 namely 41.31 emu/gr. Therefore, Mn₀₂₅Fe₂₇₅O₄ nanoparticles were chosen as the raw material in this work. The further study on the SAR values of the Mn₀₂₅Fe₂₇₅O₄-PANI material with the variation of the frequencies will also be discussed and reported.

2. Methods

2.1. Materials
The materials used in this research were iron sand from Sine beach, Tulungagung, Indonesia, HCl 12.063 M PA 99.9% (Merck), NH₄OH 6.5 M PA 99.9% (Merck), MnCl₂·4H₂O PA 99.9% (Merck), Aniline PA 99.9% (Merck), Polyvinyl Alcohol (PVA) Mw~60.000 (Merck), Ammonium Peroxide Sulfate (APS) PA 99.9% (Merck), and distilled water.

2.2. Synthesis of Mn₀₂₅Fe₂₇₅O₄ nanoparticles
First, the iron sand was dispersed using a permanent magnet. The Fe₃O₄ powder as the result of separation was dissolved in the HCl 12.063 M PA 99.9% (Merck) and stirred using hot plate magnetic stirrer for 30 minutes in a room temperature. Subsequently, the solution was filtered with a certain mess. The solution as the result of the filtration was mixed with MnCl₂·4H₂O (PA 99.9% (Merck)) and dropped with the NH₄OH (6.5 M, PA 99.9% (Merck)) solution. The next step was filtering the solution using filter paper and washed using distilled water until the pH of the solution normal. The precipitate as the result of washing was ready to be characterized and coated with PANI polymer.

2.3. Synthesis of Mn₀₂₅Fe₂₇₅O₄@PANI material
The Mn₀₂₅Fe₂₇₅O₄ powder was dissolved in the distilled water and PVA while stirred using hot plate magnetic stirrer for 20 minutes. The solution was then sonicated for 20 minutes at the room temperature and it was then called solution A. Meanwhile, the aniline was dissolved in 0.2 M HCl and this solution was then called solution B. APS was dissolved in the distilled water that was then called solution C. PANI polymer was formed from the aniline polymerized due to the presence of APS. Solution A and B were then mixed and stirred on the hot plate magnetic stirrer for one hour at the room temperature. The solution of that mixture was then mixed again with solution C and stirred using hot plate magnetic stirrer for one hour at the room temperature. After the solution was completely mixed, the solution was
sonicated for 10 minutes and followed by precipitation for ±24 hours. The precipitated sample was then washed using distilled water and ethanol until the pH normal. Finally, the last precipitate in the form of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material was ready to be characterized.

2.4. Characterization
The characterization of the sample was done using the instrument of XRD X’Pert Pro Cu-Kα 1.540 Å, FTIR Shimadzu, TEM, and magneto-thermal. In XRD test, the Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles and Mn$_{0.25}$Fe$_{2.75}$O$_4$-PANI material were run in the range of 20 angle from 10° to 70°. The data of XRD result was then analyzed using Rietica and Origin software. In FTIR test, the Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles and Mn$_{0.25}$Fe$_{2.75}$O$_4$-PANI material was dispersed in the form of powder and radiated in the range of the wavelength from 450 to 4000 cm$^{-1}$. For the characterization using TEM, the morphology data of the Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles and Mn$_{0.25}$Fe$_{2.75}$O$_4$-PANI material was analyzed using ImageJ software. Meanwhile, the determination of SAR values of the Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles and Mn$_{0.25}$Fe$_{2.75}$O$_4$-PANI material was carried out by using a magneto-thermal instrument. In the magneto-thermal characterization, the sample was placed in the coils of alternating magnetic field and given the frequency with the variation of 483 and 553 Hz. The increase in the temperature of the sample was measured using a thermometer as the function of the time. The data of the measurement results were then analyzed to calculate the SAR value of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material.

3. Results and Discussion

3.1. The XRD characterization
The X-ray diffraction pattern of the Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles was shown in Figure 1 (a). After doing qualitative analysis by search and match, the X-ray diffraction pattern of Mn$_{0.25}$Fe$_{2.75}$O$_4$ corresponded to AMCSD 0002763 model data. All diffraction peaks were detected that they fulfilled the magnetite model without the other phases. From the diffraction pattern, it can be seen that the highest crystal diffraction peak occurred at the angle of 2-theta 35.45° and it corresponded to hkl (311). The quantitative analysis using Origin software with Gaussian modelling resulted in the crystal size of the Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles of 10.08 nm with the FWHM of 0.92°. this analysis was carried out using the Debye-Scherrer equation (Equation 1).

\[
D = \frac{K\lambda}{B_0 \cos \theta}
\]

where $D$ is the crystal size, $K$ is the constant, $B_0$ is the width of the half of the peak (FWHM), $\lambda$ is the wavelength of the ray used in the characterization, and $\theta$ is the difference in the angle of the coming light and reflection light.

Meanwhile, Figure 1(b) is X-Ray diffraction pattern of Mn$_{0.25}$Fe$_{2.75}$O$_4$ @PANI material. By qualitative analysis, the X-Ray diffraction pattern of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI corresponded to AMCSD 0002763 model data and amorphous diffraction peak was also found. The half of diffraction peaks were detected that they fulfilled the magnetite model and the others were in the form of an amorphous phase. The quantitative analysis taken from the magnetite phase obtained the crystal size of the Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material was 8.09 nm with the FWHM of 1.14° in the angle of 2 thetas 35.35°. This analysis was undertaken in the peak on hkl (311) as done in Figure 1(a), where this peak is the crystalline peak of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material. Another peak in the diffraction profile of the Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material in the form of amorphous phase appearing in the angle of 2 thetas 20.08° was also found. This case is the representation of the amorphous diffraction peak of PANI material. This result is in line with the research conducted by Alam et al. [19]. They reported that the amorphous diffraction peak of PANI appeared in the angle of 2 theta which was about 20-30°.
Figure 1. X-ray diffraction pattern of (a) Mn$_{0.25}$Fe$_{2.75}$O$_4$ and (b) Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI

3.2. The FTIR characterization

Figure 2(a) shows the functional group of Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles by using FTIR instrument. Based on the result of characterization using FTIR, it can be seen that the vibration due to Metal-O bond occurred at the wavenumber of 419-531 cm$^{-1}$ where that metal was detected as Mn element. The vibration of this Mn element bond is in line with the research conducted by [20]. The vibration due to the O-H group bond happened at the wavelength of 1629 and 3426 cm$^{-1}$ that became the representation of the presence of water content in the Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles. This group stretching pattern corresponds the previous report [21,22]. Besides, the absorption at the wavenumber of 2310 cm$^{-1}$ corresponded the vibration due to the C-O bond [21].

Meanwhile, Figure 2(b) is the characterization of the functional group of the Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material. All vibration peaks detected in Figure 2(a) were detected again in Figure 2 (b) and they were the parts of the elements of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material. This characterization also resulted in functional group characterizing the PANI material by the appearance of the C-N bond. In line with the finding of Kim et al. [23] stating that the observation peak of PANI polymer was represented in the wavelength of 1100–1140 cm$^{-1}$. In this research, the C-N bond also occurred at the wavelength of 1152 cm$^{-1}$. Thereby, it can be said that the Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material has been well formed shown by the appearance of the vibration of the bonds characterizing the Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material.
Figure 2. The results of FTIR of (a) Mn$_{0.25}$Fe$_{2.75}$O$_4$ dan (b) Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI

3.3. The Morphology characterization by using TEM instrument

Figure 3 is the morphology of Mn$_{0.25}$Fe$_{2.75}$O$_4$ material that was successfully recorded using TEM with the scale bar of 50 nm. Visually, the Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles underwent aggregation due to an electromagnetic attraction of each particle because the main element of the Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles is magnetite that has remanent magnetization above 40 emu/g [18]. By using ImageJ software, the distribution of the size of Mn$_{0.25}$Fe$_{2.75}$O$_4$ particles was 8.96 nm. This result was well confirmed by data analysis using XRD that has been discussed previously namely about 10.08 nm.

Figure 3. Morphology of Mn$_{0.25}$Fe$_{2.75}$O$_4$ material by using TEM instrument
Figure 4. Morphology of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material by using TEM instrument

The morphology of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material recorded using TEM and the scale bar of 20 nm is presented in Figure 4. Visually, the aggregation of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material was getting bigger compared to the visualization of the Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles without PANI template. This case is, of course, related to the presence of PANI template in the Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles. The Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles were included in PANI template so that it reduced the movement of the nanoparticles to expand and interact with the others. By using ImageJ image, the distribution of the size of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI particles was big enough that was 30.40 nm. If it is compared to the result of characterization using TEM in Figure 3 and 4, the increase in the size of Mn$_{0.25}$Fe$_{2.75}$O$_4$ particles became Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI was big enough that was from 8.96 to 30.40 nm. This phenomenon was also reported by Han et al. [24]. They reported that the increase in PANI template in the Fe$_3$O$_4$ particles caused the increase in aggregation namely from the Fe$_3$O$_4$ size of 250 to 310 nm when they changed into Fe$_3$O$_6$@PANI [24].

3.4. The Magneto-thermal characterization

Figure 5 is the result of magneto-thermal characterization to the Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI sample using the frequency of alternating magnetic field of (a) 483 and (b) 553 Hz. Through the magneto-thermal characterization, the SAR (specific absorption rate) value of the Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI sample could be found using Equation 2 [25].

$$\text{SAR} = c\left(\frac{\Delta T}{\Delta t}\right)$$

(2)

where $T$ is the temperature of the result of characterization using magneto-thermal and $t$ is a time interval during the measurement. Meanwhile, $c$ is the constant of the magnetic material heat capacity used.

In more detailed, the SAR calculation results of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material using equation 2 are presented in Table 1. Referring to Table 1, the bigger the frequency of the alternating magnetic field used, the SAR values of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material was getting higher as well. This case means that the bigger the frequency used, the interaction of magnetic spin in the Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI nanoparticles was getting bigger as well so that the movement of the magnetic particles in the sample
changed impacting on the increase in the temperature of the sample. From Equation 2, it can be clearly seen that the change in the temperature of the sample was in line with the SAR value obtained from that sample. This result correlates to the previous research result. The SAR values of gamma-Fe$_2$O$_3$ particles were in the range of 22–200 W/g with the strength of the alternating magnetic field of about 100–500 Oe and at the frequency of 160 kHz [15]. Meanwhile, Shete et al. [17] also reported that Fe$_3$O$_4$ nanoparticles without coating had the smaller SAR value that was 79.32 W/g compared to the SAR value of Fe$_3$O$_4$ nanoparticles with chitosan coating that was 118.85 W/g in the magnetic field induction of 167.6 - 335.2 Oe. Thereby, the Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material has the potential to be the material in cancer therapy that can change the temperature of the sample in the range of 15-18 °C. However, further repairment is necessary to increase the change in the temperature of that sample.

**Figure 5.** The magneto-thermal characterization of the Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI sample with the frequency of (a) 483 and (b) 553 Hz

**Table 1.** The SAR values of some measurements with certain frequencies

| Frequency (Hz) | SAR (W/g) |
|---------------|-----------|
| 483           | 0.81      |
| 553           | 3.65      |

4. Conclusion
This research has successfully synthesized Mn$_{0.25}$Fe$_{2.75}$O$_4$ nanoparticles using coprecipitation method and Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material using in-situ polymerization. Based on the characterization using XRD, the crystal size of Mn$_{0.25}$Fe$_{2.75}$O$_4$ material was about 10.08 nm and the formed Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI had the crystal size of 8.09 nm. Through the magneto-thermal characterization, the higher the frequency of the alternating magnetic field used, the SAR value of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material obtained would be bigger as well. In the frequency of 483 Hz, the SAR value of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material was 0.81 W/g while in the frequency of 553 Hz, the SAR value was 3.65 W/g. From this behavior of magneto-thermal characteristic of Mn$_{0.25}$Fe$_{2.75}$O$_4$@PANI material, this particular material has the potential to be a material of cancer therapy.
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