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To cite this article: A Taufiq *et al* 2017 *IOP Conf. Ser.: Mater. Sci. Eng.* **202** 012008

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IOP Conf. Series: Materials Science and Engineering **202** (2017) 012008 doi:10.1088/1757-899X/202/1/012008

Fabrication of Magnetite Nanoparticles Dispersed in Olive Oil and Their Structural and Magnetic Investigations

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Abstract: In this work, the iron sand taken from Wedi Ireng Beach in Banyuwangi, Indonesia, was employed as the main precursor in fabricating magnetite nanoparticles. The magnetite nanoparticles were then functionalized in preparing magnetic fluids coated by oleic acid as a surfactant and dispersed in olive oil as a liquid carrier. The phase purity, crystallite size and crystal structure of the dried magnetic fluids were characterized by using X-Ray Diffractometer. Meanwhile, the functional groups of the magnetic fluids were investigated by means of Fourier Transform Infra-Red (FTIR) spectroscopy. The particle size and morphology of the magnetite particles were also investigated by using Transmission Electron Microscopy (TEM). The magnetic behaviors of the magnetic fluids were determined by using Vibrating Sample Magnetometer (VSM). Based on the XRD data analysis, the magnetite particles crystallized in the spinel structure without the presence of any other phases. The FTIR spectra showed that the functional groups of the magnetic fluids were referring to the magnetite, oleic acid, and olive oil. The TEM image presented that the magnetite particle was formed in a nanometric size. Finally, the saturation magnetization of the magnetic fluids varied in the mass composition and particle size of the magnetite nanoparticles.

Keywords: Magnetite, magnetic fluid, olive oil, structure, magnetization

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1. Introduction

Magnetic fluids or ferrofluids, colloidal suspensions containing magnetic nanoparticles in a single domain and dispersed in appropriate media, become a group of smart magnetic materials that play an essential role in many advanced applications. Due to their unique properties, the magnetic fluids can be applied in several fields, such as for rotating seal and rotating damper [1], as contrast agent in magnetic resonance imaging [2], for magnetic sensing [3], and so forth.

In 2016, Borin and co-workers successfully prepared magnetite-based magnetic fluids by employing alkenyl succinic anhydride, stearic acid, and oleic acid as combined surfactants [4]. In 2015, El-Boubbou and co-workers successfully synthesized the magnetite nanocolloids by using fatty acid as a stabilizer [5]. Furthermore, Lobato *et al*. successfully prepared magnetic fluids by employing oleic acid as appropriate surfactant and produced the magnetic with average particle size of about 10 nm [6]. Therefore, it can be pointed out that oleic acid becomes one of the best choices to be used as an essential surfactant in surfacing the magnetite particles in magnetic fluids

It was reported that the magnetic fluids surfaced by oleic acid as a surfactant were prepared using several methods to produce them in various sizes and morphologies. In general, the preparation of magnetic fluids was started by preparing the magnetic nanoparticles using several methods such as coprecipitation [7,8], sonochemistry [9], hydrothermal [10], one-pot synthesis [11], and so on. Moreover, the magnetic fluids were also prepared by dispersing the magnetic nanoparticles in various liquid carriers such as water [7], dialkyldiphenyl and polyethylsiloxane [4], transformer oil [12], and others. However, several problems of the utilization of the mentioned liquid carriers still appeared and needed to be overcome by suitable strategies. The strategies can be conducted such as by lowering the particle size of the magnetic and reducing the agglomeration using appropriate liquid carrier and surfactant.

In this work, we develop synthesis method of the magnetic fluids by introducing a combination of oleic acid and olive oil as a stabilizer and as a dispersing media of the magnetic nanoparticles, respectively. The oleic acid and olive oil were selected because they have good performances related to their similar physical properties. Therefore, we believed that olive oil could play an essential role as a suitable liquid carrier to produce the magnetic fluids with a homogeneous magnetic particle size. So far, based on our knowledge, there is no a report of the utilization of oleic acid as a surfactant and olive oil as a liquid carrier in preparing the magnetic fluids from iron sand becomes a novelty of this work. In order to cut off the preparation cost, we propose the use of an inexpensive raw material in preparing the magnetic fluids by exploring the natural iron sand from Indonesia. Furthermore, this paper also presents the structural and magnetic behaviors of the magnetic fluids.

2. Experimental Method

The materials used were iron sand taken from Wedi Ireng Beach in Banyuwangi, Indonesia, HCl, NH_4OH, distilled water, oleic acid, and olive oil. The iron sand was washed by using distilled water for several times and followed by natural drying process under sunlight. The iron sand was then extracted by using a magnetic separator to obtain the magnetite powder [13]. The powder was reacted with HCl as a dissolving mediator by using the method proposed in our previous work [14]. The product of the previous process was then reacted with NH_4OH using a magnetic stirrer to achieve a black precipitate sample [15]. The process was continued by washing the precipitate sample for several times to get magnetite particles. The prepared magnetite particles were then coated with oleic acid as a surfactant and dispersed in olive oil as a liquid carrier. The three samples were prepared by varying the mass compositions of the magnetite particles in the magnetic fluids to 0.32, 0.38, and 0.41 g. The samples were coded by MF1, MF2, and MF3 for the magnetite particles' compositions of 0.32, 0.32, and 0.41, respectively.

For characterization processes, X-Ray Diffractometer (XRD) was employed to study the phase purity, crystallite size and crystal structure of the dried magnetite fluids. Fourier Transform Infra Red (FTIR) spectroscopy was used to characterize the functional groups of the magnetic fluids. Transmission Electron Microscopy (TEM) was utilized to investigate the particle size and morphology

of the magnetite particles. Finally, Vibrating Sample Magnetometer (VSM) was employed to study the magnetic behaviors of the magnetic fluids.

3. Results and Discussion

The X-ray diffraction pattern of the dried magnetic fluid prepared from iron sand taken from Wedi Ireng Beach in Banyuwangi, Indonesia, is presented in Figure 1. In the figure, the circle and solid line represent the diffraction data and refinement model, respectively.

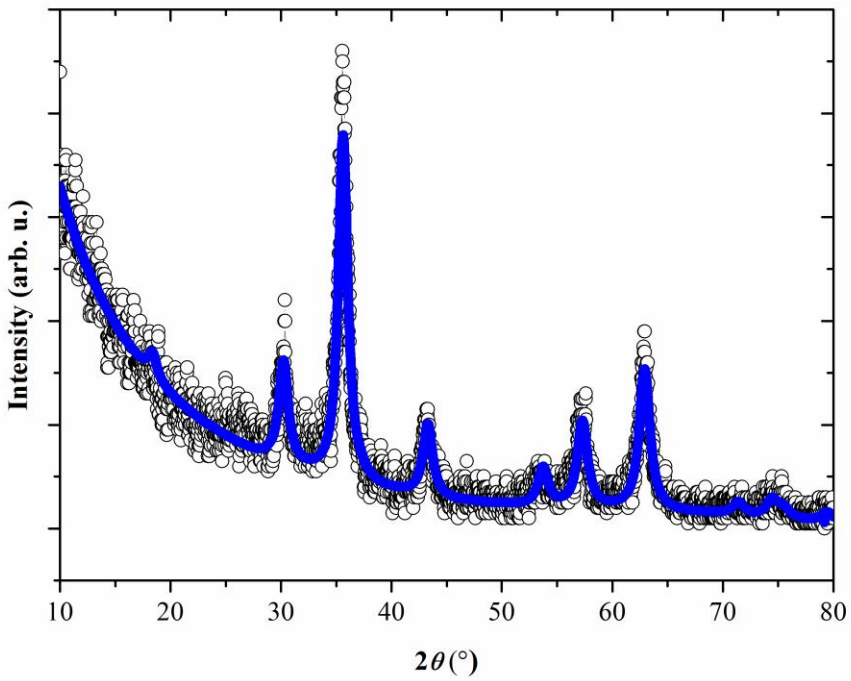

Figure 1. X-ray diffraction pattern of the dried magnetic fluid using Cu-Kα radiation 1.5406 Å.

The quantitative analysis for the X-ray diffraction pattern exhibited a single phase of magnetite without any impurities. The refinement parameters such as *Rp, Rwp,* and *GoF* were 13.64, 18.91, and 0.58, respectively. These values indicated that the experimental data were well refined by the theoretical model. Furthermore, the lattice parameter and crystal volume of the dried magnetic fluid obtained from Rietveld analysis were 8.354 Å and 583.02 $Å^3$, respectively. The sample was crystallized in a cubic crystal structure. In this structure, the Fe^{2+} and Fe^{3+} ions randomly placed tetrahedral and octahedral sites. Furthermore, the dried magnetic fluid formed on a nanometric scale with the size of about 9 nm. This result corresponds to the size of secondary particles constructed by primary particles as building blocks [13]. In order to confirm the particle size of the sample, we also characterized the dried magnetic fluid by using TEM as presented in the following figure.

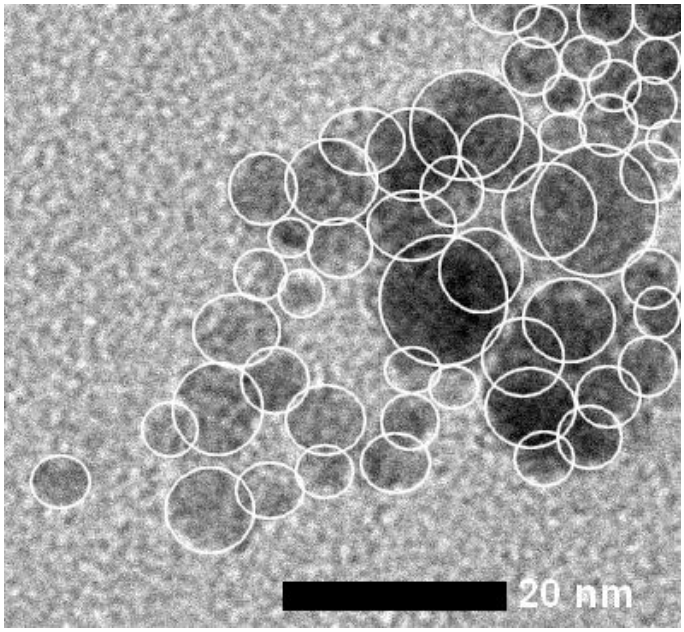

Figure 2. TEM image of the dried magnetic fluid.

The TEM image in Figure 2 shows that the dried magnetic fluid tended to agglomerate in a nanometric size. Theoretically, the agglomeration phenomenon in the magnetite occurs during nucleation process of the magnetic nanoparticles. Quantitatively, the average particle size of the sample was approximately 9.3 nm. This result is quite similar to the particle size obtained by a calculation using Debye Scherer's formula for the XRD data. Another work reported by Song and co-workers has successfully employed a co-precipitation method in preparing magnetite nanoparticles with the size of about 12 nm from commercial precursors [16]. Furthermore, Liu et al. reported that they successfully synthesized the magnetite nanoparticles by using a rapid microwave-assisted hydrothermal method with the particle size of about 20-25 nm, also from commercial precursors [17]. Therefore, the use of iron sand in this work was effective as a raw material in producing the magnetic fluids with the magnetite's particle size below 10 nm.

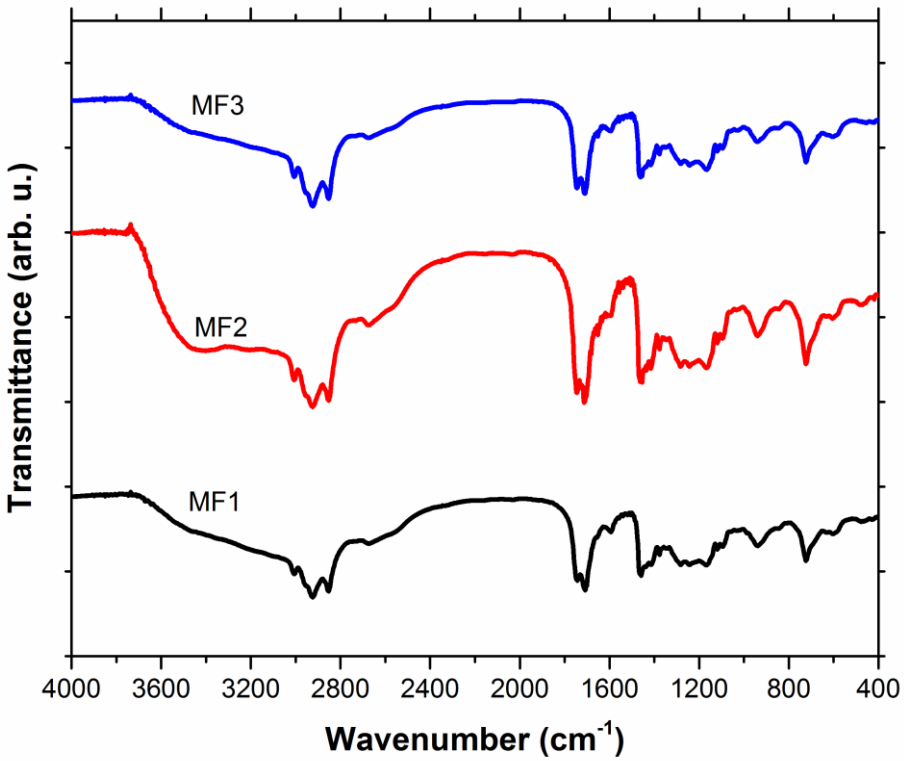

Figure 3. FTIR spectra of the magnetic fluids.

Based on Figure 3, it can be seen that there are several peaks indicating the functional groups of the samples. The magnetite particles can be identified at the wavenumbers of around 590 cm^{-1} and 600 cm^{-1}. These peaks represented the intrinsic stretching vibrations of Fe^{2+}-O and Fe^{3+}-O that randomly placed in the octahedral and tetrahedral positions. In a good agreement with this experiment, other groups reported that vibrations of C-H could be detected at around 1112 cm^{-1}, stretching vibrations of C=C in oleic acid could be detected at around 1384 cm^{-1}, stretching vibrations at 3404 cm^{-1} and 1636 cm^{-1} were originated from -CH_3 and C=C groups, respectively [18]. Furthermore, the presence of oleic acid could be detected by the stretching vibrations of -CH_2 groups at the bands around 2921 cm^{-1} and 1852 cm^{-1} [19]. The bands at around 1430 cm^{-1} and 1590 cm^{-1} represented the vibrations of a oleat (-COO) [20]. Furthermore, the peak observed at around 1744 cm^{-1} represented the vibration of C=O, while the peaks at around 3005 cm^{-1} and 1171 cm^{-1} represented C-H and C-O, respectively, as band characters of olive oil. These results are in good agreement with the results reported by another group [21]. Therefore, the FTIR data showed the present oleic acid that successfully covered the magnetite nanoparticles of the fluids dispersed in the olive oil. As a final remark, in this work, the olive oil plays an essential role as a good liquid carrier of the magnetic fluids.

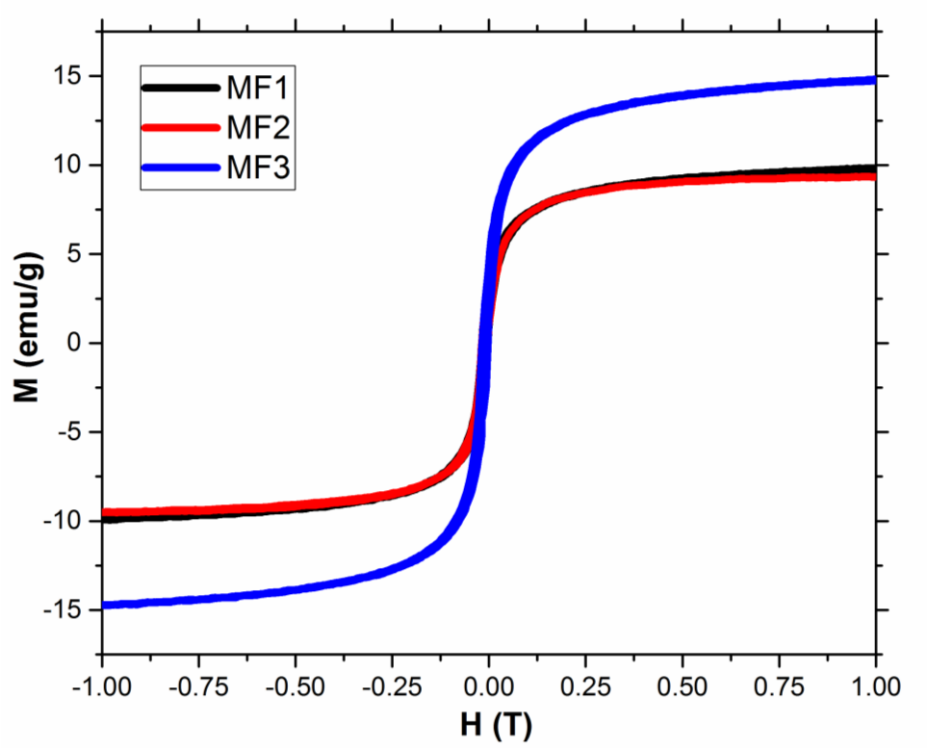

Figure 4. Magnetization curves of the magnetic fluids.

Figure 4 shows the magnetization curve of the magnetic fluids prepared from the iron sand. From the figure, MF1 and MF2 had similar saturation magnetization values, while MF3 had the highest one. The particle size obtained from the quantitative data analysis for magnetization curve and saturation magnetization of the magnetic fluids are presented in Table 1.

Table 1. Saturation magnetization (M_s), and particle size of the magnetic fluids

Sample	M_s (emu/g)	Particle size (nm)
MF1	9.81	8.0
MF2	9.76	7.3
MF3	14.81	8.4

Visually, all samples exhibited a superparamagnetic character at room temperature because they had an *S* shape and their coercive field and remanent magnetization were almost negligible. From Table 1, all of the samples had the particle size of less than 10 nm which indicated a single domain character of the magnetic fluids and exhibited a superparamagnetic character. In a single domain state, the magnetic particles are uniformly magnetized and alignment of all the spins are in the same direction [22]. It can be seen from Figure 4 and Table 1 that the MF1 and MF2 samples have a similar saturation magnetization value of around 9.8 emu/g, while MF3 sample has the highest one of around 14.8 emu/g. Quantitatively, the saturation magnetization (M_s) values of all samples were smaller than that of bulk magnetite [23]. The decreasing saturation magnetization was originated from the decreasing particle size of the sample. In more detail, the saturation magnetization values of MF1 and MF2 were quite similar. Theoretically, the saturation magnetization of MF2 should increase because its magnetite particle size was bigger than that of MF1. This phenomenon was originated from the increasing magnetite content in the magnetic fluid. Furthermore, MF3 has the highest saturation magnetization value than those of other samples originating from its increasing mass composition and

particle size of the magnetite simultaneously. These results are in good agreement with the results reported by another group [24].

4. Conclusion

Based on the above discussion, we conclude that the magnetite particles from iron sand were successfully prepared as magnetic fluids using oleic acid as a surfactant and olive oil as a liquid carrier. All magnetic particles were crystallized in a cubic structure in a single phase. The magnetic particle sized in a nanometric scale of below 10 nm obtained by XRD, TEM, and VSM characterizations. The functional groups obtained by FTIR characterization showed the presence of the magnetite, oleic acid, and olive oil in the magnetic fluids. Moreover, the saturation magnetization of the magnetic fluids had a function of the mass composition and particle size of magnetite nanoparticles.

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Acknowledgements
The authors would like to thank DRPM, Kemenristekdikti, Republic of Indonesia, for supporting a "research grant" for AT.