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DOI: 10.21776/ub.jpacr.2018.007.02.393 Synthesis and Characterization of Fe3O4 Nanoparticles Modified with Polyethylene Glycol as Antibacterial Material Poedji Loekitowati Hariani1*, Desnelli1, Fatma1, Rizki Indah Putri1, and Salni2 1Department

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Inderalaya OI, Indonesia *Corresponding author: puji_lukitowati@mipa.unsri

.ac.id Received day month year; Accepted day month year (will be given) ABSTRACT 1

The iron oxide (Fe3O4) nanoparticles modified with polyethylene glycol (PEG) was synthesized by co-precipitation methods using ferric and ferrous ions as the precursors. Further, the antibacterial activity was performed against gram-positive and gram-negative bacteria. The Fe3O4-PEG was characterized using

X-Ray Diffraction (XRD), Fourier Transform Infra Red (FTIR), Scanning Electron Microscopy (SEM) with **energy dispersive X-Ray** analysis (EDAX) **and Vibrating Sample Magnetometer (VSM).** 6

The particle size of Fe3O4-PEG calculated using XRD is 46.2 nm. The study confirmed that Fe3O4-PEG is superparamagnetic and has a saturation magnetization of 56.43 emu/g. The prepared Fe3O4-PEG gives the effect of both gram-positive and gram-negative pathogenic bacterial strains hence this material has potential utilization in the field of pharmaceutical and biomedical in the future. Keyword: Fe3O4, nanoparticle, PEG, antibacterial activity INTRODUCTION In recent years, spinel ferrite nanoparticles have been the subject of developed research. The dimension of nanoparticle is between bulk materials and atoms or molecules. The spinel ferrite has a structural formula MFe2O4, where M is a divalent metal with a cubic spinel crystal structure. In addition, the spinel ferrite has magnetic properties. Magnetic nanoparticles can be used in various applications such as adsorbent [1,2], magnetic storage, ferrofluids, biomedical applications [3,4] and gas sensor [5]. Various methods can be used to prepare magnetic nanoparticle such as co-precipitation [6,7], hydrothermal [8], microemulsion [9], electrochemical route [10,11] and sol-gel [12]. One Tof the magnetic nanoparticles is Fe3O4 (magnetite). The materials exhibit superparamagnetic behavior, low toxicity, biocompatibility, easier surface modification [13,14]. Another study reported that Fe3O4 magnetic nanoparticles have antibacterial activity properties. Fe3O4 nanoparticles showed strong antibacterial activity, the antibacterial activity caused by the presence of reactive oxygen species (ROS) [4]. The Fe3O4 nanoparticle has the greatest antibacterial effect to pathogenic bacteria of Pseudomonas aeroginosa than Escherichia coli and Staphylococcus aureus [15]. In addition, Fe3O4 nanoparticle has a zone of inhibition consideration to Ag nanoparticle for topical use [14].

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Materials in the form of small particle sizes are easy to agglomerate and made it larger particle size, reducing surface area and magnetic properties. Agglomeration can be prevented by coating nanoparticles with organic polymeric materials. The coating process also prevents nanoparticles from oxidation processes, reduces toxicity and increases chemical stability [16,17]. Some research on coating nanoparticles with organic materials such as CoFe2O4- alginate [18], Fe3O4-polypropylene [8], CoFe2O4-chitosan [19], CoFe2O4-polyvinyl alcohol, gelatin [16]. In this study, we used co-precipitation method for preparing Fe3O4 and coating with polyethylene glycol (PEG)-4000. PEG is long polymer chains with several advantages for coating Fe3O4 of non-toxic in a large extent, non-immunogenic, non-antigenic, and protein- resistant polymer [16,20]. In addition, the incorporation of inorganic and organic particles has combination of the properties of inorganic particles such as thermal, mechanical, magnetic, and the properties of organic particles that is flexibility. Furthermore, the Fe3O4-PEG were evaluated for antibacterial activities. The bacteria used are gram-positive (Staphylococcus aureus) and gram-negative (Escherichia coli).

EXPERIMENT Chemicals and instrumentation The chemical reagents **used in this study were** 11

FeCl3.6H2O, FeCl2.4H2O, NaOH, HCl and PEG-4000, nutrient agar from Merck.

The bacteria species **Staphylococcus aureus ATTC 25923 and Escherichia coli ATCC 25922** 13

from PT Bio Farma. Synthesis Fe3O4 -PEG 4000 The Fe3O4 magnetic nanoparticles prepared by co-precipitation. Initially,

5.41 g of FeCl3.6H2O and 1.99 g of FeCl2.4H2O were added into 20 **mL of distilled** deionized **water.** Into **the** mixture, aqueous **solution** 7

NaOH 1 M is added dropwise until pH 11 while flowing N2 gas at room temperature and stirring at 200 rpm [21,22]. The magnetic nanoparticles are black precipitates, which can be separated from the solution using magnet permanent. The powder was washed using aquadest until neutral and then washed using ethanol. The product was dried under vacuum for 3 h at 60 0C. The reaction synthesis of Fe3O4 by coprecipitation method is as follows: Fe3+ + Fe2+ + 8OH- → Fe3O4↓ + 4H2O (1) The next step was dissolving PEG (2.50 g) in 5 mL deionized water. The solution was stirred for 30 minutes until homogeneous. Then, 0.25 g of Fe3O4

was added to the suspension. **The mixture was stirred under nitrogen** atmosphere **for** 12

10 h at 450C. Fe3O4 coated PEG product were separated from the solution by centrifugation. Finally, the product washed with ethanol to dissolve the remaining of PEG. The crystal structure of Fe3O4-PEG was obtained by XRD (Shimadzu XD-3H) with Cu Kα (λ = 1.5406 Å) radiation, magnetic properties were determined by VSM (Lake Shore 7410), in

an external field (temperature in the range 10-400 K), functional group was analyzed by FTIR Shimadzu 5400 in the range 4.000-400 cm-1, morphology and element composition of Fe3O4-PEG were studied by SEM-EDX JEOL-JSM-6510 LV. Screening of antibacterial activities In this study, the test of antibacterial activity was conducted using diffusion disc method [23]. The bacteria (Staphylococcus aureus and Escherichia coli) were inoculated to Petri dish with Nutrient Agar (NA) medium, then paper disc with 6 mm diameter were used to inoculated test organism. Fe3O4-PEG 10 μL was instilled

with different concentration **(0; 12.5; 25; 50; 100 and 200 μg/mL).** 9

The Petri dish were wrapped by parafilm tape and then all of Petri dish

were incubated at 37°C for 24 hours. The **antibacterial** activities were determined **by** measurement **the** zone inhibition **diameter** 14

in millimeters. RESULT AND DISCUSSION Characteristic of Fe3O4-PEG 4000 The XRD pattern of Fe3O4 and Fe3O4-PEG shown in figure 1. The product has a cubic spinel structure in accordance with JCPDS 19-0629. The main peak of Fe3O4 that appears on 2θ corresponds to the reflection planes (220), (311), (400), (422), (511) and (440). The Fe3O4-PEG spectra decreased in intensity due to the addition PEG which has amorphous properties.

Using the Scherrer formula (D = **0.89λ/β cos θ)** one can estimate **the average size** 10

of crystals of nanoparticles,

where β is the full width at half maxima **(FWHM).** Calculation **of** 15

crystal size based on the peak is highest in reflection plane of the (311) peak. It was found that the Fe3O4-PEG crystal size larger than Fe3O4. The crystallite sizes is estimated 46.2 and 35.7 nm, respectively. The similar results that Fe3O4 nanoparticles coated using polyethyleneimine (PEI) has larger crystal size than Fe3O4 without coating [11]. Other studies show that CoFe2O4 nanoparticles have a smaller particle size than CoFe2O4 that is coated using PEG [24]. 6000 5000 4000 3000 Intensity (311) Fe3O4 Fe3O4-PEG 4000 (220) (440) 2000 (400) (422) (511) 1000 0 0 10 20 30 40 50 60 70 80 90 100 2 Theta (deg) Figure 1. XRD pattern of Fe3O4 and Fe3O4-PEG 4000. The presence of PEG on the surface of Fe3O4 nanoparticles was evaluated by FTIR. Figure 2 displays the FTIR spectra of Fe3O4 and Fe3O4-PEG 4000 at range 400-4000 cm-1. The peaks that appeared at the wave number 3390.6 cm-1 on the Fe3O4 and 3392.5 cm-1 on Fe3O4-PEG 4000 showed the absorption band of O-H groups from the adsorbed H2O onto materials. The peaks of Fe3O4 and Fe3O4-PEG 4000 at the 1626.7 and 1627.8 cm-1 indicate the bending vibrations of O-H. The characteristic peaks of PEG showed at 2862.2 and 1458.1 cm-1 assigned stretching vibration and bending vibration of C–H in –CH2. The band at 1470 cm-1 is C-C vibration stretching PEG. The peak that appears at 1110.9 cm-1 is to the bond stretching vibrating of C-O [8]. The peak is characteristic of PEG, that does not appear on the Fe3O4 spectra. The wave numbers are characteristic of Fe-O bonds shown with a strong peak at 584.4 and 532.3 cm-1 on Fe3O4 and Fe3O4-PEG, respectively. The shift of wave numbers of Fe-O bond on Fe3O4-PEG spectra shows the interaction between Fe3O4 and PEG. Several studies have shown that the wave number of the Fe-O bond appears at 569 cm-1 [20], 581 cm- 1 [25], and 567.12 cm-1 [10]. % T 110 100 90 80 1110.9 70 60 532.3 50 40 20 30 Fe3O4-PEG 4000 Fe3O4 584.4 4400 4000 3600 3200 2800 2400 2000 1600 1200 800 400 cm-1 Figure 2. Spectra FTIR of Fe3O4-PEG 4000. Figure 3 shows the magnetic curves of Fe3O4 and Fe3O4-PEG. It reveals that Fe3O4 has high saturation magnetization than Fe3O4-PEG. The addition of organic polymers (PEG) causes a small reduction in magnetic properties. Fe3O4-PEG is classified as superparamagnetic which in this research shows saturation magnetization of 56.43 emu/g while Fe3O4 is 74.33 emu/g. The changes in magnetic properties due to the effect of surface modification of Fe3O4 by large polymer molecules. The greater the concentration of PEG is added, the lesser of the saturation magnetization where the polymer coat the nanoparticles so that giving a protection effect from the magnetic field [24]. The saturation magnetization value is similar to the other reference [20,25]. Their study reported Fe3O4 modified with sodium citrate and oleic acid with various concentrations of Fe3O4 showed magnetization saturation of 50.61 – 61.36 emu/g [25]. 100 Momen Magnet (emu/g) 80 60 40 20 0 -1 -0,5 -20 -40 -60 -80 -100 0 0,5 Fe3O4-PEG 4000 Fe3O4 1 Field (tesla) Figure 3. The saturation magnetization of Fe3O4 and Fe3O4-PEG 4000. The morphology of Fe3O4 and Fe3O4-PEG 4000 and its constituent elements were analyzed using SEM-EDX. SEM image of Fe3O4 and Fe3O4-PEG 4000 are displayed in figure 4. The image shows a clear difference between Fe3O4 before and after modification with PEG. The morphology of Fe3O4 appears to be agglomerated while modified with PEG appears to be dispersed on PEG surface. a b Figure 4. SEM image of (a) Fe3O4 and (b) Fe3O4-PEG 4000 Table 1 shows the constituent elements of Fe3O4 and Fe3O4-PEG 4000. It can be seen that the addition of carbon elements in Fe3O4-PEG 4000 indicates that modification of Fe3O4-PEG is successful. The main elements of Fe3O4 are Fe and O while modified of Fe3O4 with PEG affects as the percentage of C element increases. Table 1. The data of elements Fe3O4 and Fe3O4-PEG 4000 Materials Mass (%) Fe O C Fe3O4 43.36 56.98 - Fe3O4-PEG 4000 26.80 38.93 34.27 Antibacterial activity Antibacterial activities in this study show in figure 5 and table 2. In the figure, we can see that zone of inhibition of Fe3O4-PEG 4000 to Staphylococcus aureus and Escherichia coli. The different concentrations of Fe3O4-PEG 4000 give different diameter of inhibition zone. The size of the inhibitory zone depends on the type of bacteria, the size, and concentration of the nanoparticles [26]. The antibacterial activity of Fe3O4 can be explained that Fe3O4 is positively charged while the bacterium is negatively charged, so there is an interesting attraction between Fe3O4 and bacteria. Bacteria is oxidized and dies [4]. In this study, it appears that gram-positive bacteria has a smaller diameter of inhibition zone than that in gram-negative bacteria. Another study, also suggest that the gram-negative bacteria are more sensitive compare to gram-positive bacteria 5

[4]. (a) (b) ii i ii i iii vi iii vi iv v iv v Figure 5. Antibacterial activity of Fe3O4-PEG 4000 with concentration (i)0

(ii)12.5 (iii)25 (iv)50 (v)100 and (vi)200 µg/mL 8

to (a) Staphylococcus aureus and (b) Escherichia coli (give information number in image; the number refer to concentration sample in ppm or other) The same result was reported previously, that gram-negative have higher susceptibility than gram-positive bacteria. The killing rate of Escherichia coli is higher than that in Staphylococcus aureus [27]. The differences in susceptibility are caused by differences in cell wall structures, cell physiology and metabolism [15,28]. PEG was also reported to have antibacterial activity. The hydrophilic properties of PEG inhibit bacterial growth. Water is very important for bacteria for growth and multiplication [29]. The MIC (Minimum Inhibitory Concentration) for Staphylococcus aureus is 12.5 µg/mL with an average of inhibitory diameter 6.6 mm, and this result is smaller than that in Escherichia coli with an average of inhibitory diameter 6.3 mm at concentration 25 µg/mL. Table 2. The diameter of inhibition zone for Fe3O4-PEG 4000 Concentration Average inhibitory diameter (mm) (µg/mL) Staphylococcus aureus Escherichia coli 200 11.0 10.2 100 11.1 11.3 50 7.3 8.4 25 6.3 7.2 12.5 0 6.6 0 0 0 CONCLUSION Fe3O4 nanoparticles modified with PEG could be used as an antibacterial material. The Fe3O4-PEG showed

antibacterial properties on gram-positive and gram-negative bacterial strains. The antibacterial effect of 5

Fe3O4-PEG on Escherichia coli is stronger than Staphylococcus aureus. The MIC value of Fe3O4-PEG for Escherichia coli is 12.5 µg/mL whilest for Staphylococcus aureus is 25 µg/mL. These results suggest that the Fe3O4-PEG has a potential applications, and further research has to be undertaken for toxicity evaluation in animal model or human. ACKNOWLEDGMENT The financial support of the research by Sriwijaya University for Unggulan Kompetitif Universitas Sriwijaya Research Grant No. 988/UN9.3.1/PP/2017. REFERENCES [1] Sharma, Y.C., and Srivastava, V., J. Chem. Eng. Data, 2011, 56, 819–825. [2] Shahriari, T., Bidhendi, G.B., Mehrdadi, N., Int. J. Environ. Sci. Technol., 2014, 11, 349–356. [3] Sam, S., and Nesaraj, A.S., Int. J. Appl. Sci. Eng., 2011, 9(4), 223–239. [4] Prabhu, Y. T., and Rao, K, V., Int. Nano Lett., 2015, 5, 85–92. [5] Vignesh, R. H., Sankar, K.V., Amaresh, S., Lee, Y.S., Selvan, R.K., Sensor Actuat B- Chem., 2015, 220, 50–58. [6] Iwasaki, T., Mizutani, N., Watano, S., Yanagida, T., Kawai, T., J. Exp. Nano Sci., 2010, 5(3), 251–

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