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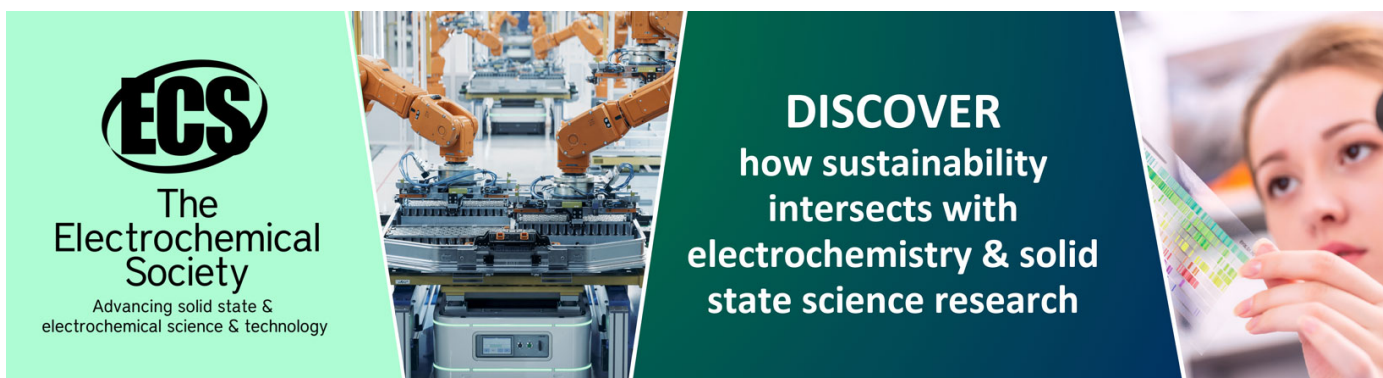
Green synthesis of magnetite (Fe_3O_4) nanoparticles using *Graptophyllum pictum* leaf aqueous extract

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Green synthesis of magnetite (Fe_3O_4) nanoparticles using *Graptophyllum pictum* leaf aqueous extract

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Abstract. Magnetite nanoparticles (MNPs) attracted the attention of many researchers due to their unique properties. In this research, nanoscale magnetite particles have been successfully synthesized through an environmentally friendly method using aqueous extract of *Graptophyllum pictum* leaf (GPLE). In MNPs formation, GPLE acted as a base source and capping agent. Alkaloids in GPLE were hydrolyzed in water and hydroxylated Fe^{2+} to form Fe_3O_4 nanoparticles powder through calcination. After the addition of leaf extract, MNPs formation was observed by color change from pale yellow to dark brown. The synthesized nanoparticles were characterized using UV-Vis spectrophotometer, X-Ray diffraction (XRD), and Fourier transform infra red (FTIR) spectroscopy. The results confirmed that MNPs formation indicated the surface plasmon resonance at a maximum wavelength, λ_{max} 291 nm. The average crystallite size is 23.17 nm. The formed MNPs through green synthesis method promise in various medical applications such as drug carrier and targeted therapy.

1. Introduction

Recently, researches on magnetite nanoparticles (MNPs) have been widely developed. MNPs were reported to have some advantages in biocompatibility, biodegradability [1], and easily encapsulated [2]. MNPs are used in various application such as catalyst [3,4], biosensor [5], magnetic resonance imaging (MRI) [6,7] and targeted drug delivery [8,9].

Several conventional methods including co-precipitation and thermal decomposition are widely employed for MNPs synthesis. Co-precipitation method is used due to its simplicity [10] whereas thermal decomposition is a good method in controlling nanoparticles size and morphology [11]. Those methods use chemicals as reagent which are not environmentally friendly. In contrast with the conventional method, green synthesis is more friendly to environment because it utilizes plants extract as the chemicals substitute.

There are several successful studies in synthesizing of MNPs using plant extract, for instance, MNPs synthesis using banana peel ash and *Colocasia esculanta* leaves [12], soya bean sprout [13], and *Datura Innoxia* leaves [14] extract. Unfortunately, some of them still used chemicals such as sodium hydroxide and ammonium hydroxide as a base source. From research, there is limited report of MNPs synthesis using Indonesian medicinal plants.

Graptophyllum pictum is a traditional medicine plants from Papua, Indonesia [15] containing alkaloids [16]. Its secondary metabolite contents act as an alternative base source and capping agent in MNPs synthesis as hazardous chemicals substitute. In previous study, *Graptophyllum pictum* leaf extract (GPLE) was reported contains alkaloids and flavonoids [16]. Alkaloids as a weak base are needed in MNPs preparation, and flavonoids have hydroxyl group to prevent MNPs agglomeration



[11]. Both of functional materials are contained in GPLE as base source and capping agents. Therefore, this research is important in MNPs synthesis, which is not reported in previous study yet.

2. Experimental

2.1. Materials

Iron (II) sulphate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) was purchased from Merck as precursor of MNPs synthesis. *Graptophyllum pictum* leaves (GPL), were obtained from Cimanggu, Bogor, Indonesia. All the aqueous solutions were prepared by MilliQ water ($18.2 \Omega\text{cm}^{-1}$).

2.2. Preparation of GPLE

GPL were cleaned and dried in room temperature for five days. 100 g of leaf powder were soaked in 500 distilled water for 10 days under stirring. The extract was filtered as GPLE using whatman No. 41. The presence of flavonoids, alkaloids, saponin, tannin and terpenoids were tested by general phytochemical method [16].

2.3. MNPs synthesis using GPLE

Synthesis of GPLE-capped MNPs was conducted by mixing 6 mM Fe^{2+} solution with GPLE under stirring at 45°C for three hours until the solution change to dark brown. The mixture was aged for two hours at room temperature, dried in oven at 100°C for one hour and at 70°C for three hours to form MNPs-GPLE powders.

2.4. Characterization

The functional groups presence in active compounds of GPLE and MNPs-GPLE were investigated using FTIR Spectrophotometer (IRPrestige-21 Shimadzu) at range of $400 - 4000 \text{ cm}^{-1}$. The UV-Vis absorption spectrum of MNPs-GPLE was examined using UV-Vis spectrophotometer (Shimadzu 2600) at range $200 - 800 \text{ nm}$. The MNPs band gap was determined using UV-Vis DRS (Shimadzu 2450). The XRD pattern of MNPs-GPLE was observed at 20 mA and 45 kV with Cu $K\alpha$ radiation ($\lambda = 1.54 \text{ \AA}$) at 2θ range of $10 - 80^\circ$.

3. Result and discussion

The UV-Vis absorption spectra of Fe^{2+} solution, GPLE and MNPs-GPLE colloids are shown in Figure 1.

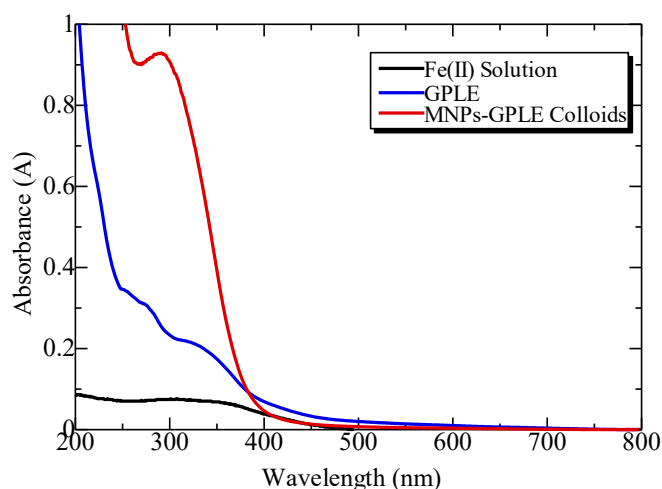


Figure 1. UV-Vis absorption spectra of Fe^{2+} solution, GPLE, and MNPs-GPLE colloids.

The presence of surface plasmon resonance in UV-Vis absorption spectrum at a maximum wavelength, $\lambda_{\max}$ 291 nm indicates the MNPs formation. This $\lambda_{\max}$ is suitable with the previous result of MNPs synthesis using *Datura innoxia* leaf extract [14] and conventional chemical method [17]. Fig. 2 shows linear correlation between $F(R)$ against E_g to determine MNPs-GPLE band gap. Based on graphic equation $y = 8.954x - 13.953$, the band gap is 1.56 eV.

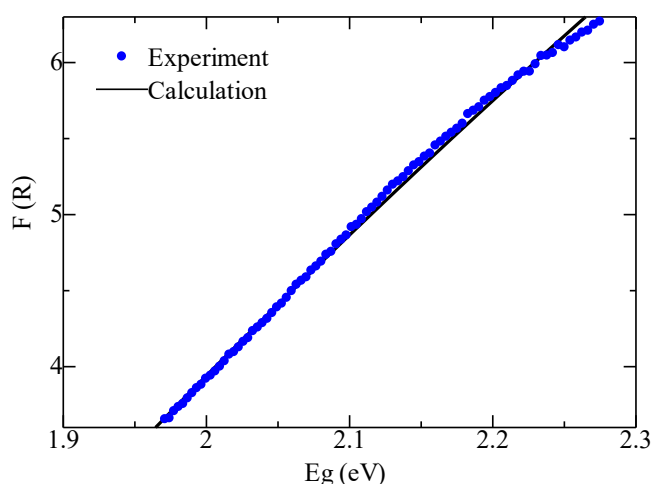


Figure 2. Linear correlation between $F(R)$ against E_g to determine MNPs-GPLE band gap.

FTIR characterization was conducted to study the interaction between functional groups of GPLE and MNPs. Figure 3 showed the wavenumber shifts of GPLE before and after reaction with Fe^{2+} . The band at 3412.0 cm^{-1} of -OH stretch [14] in GPLE shifted to 3051.5 cm^{-1} and occurred the decreasing of % transmittance due to MNPs-GPLE formation. The band at 1419.7 cm^{-1} of -NH stretch [14] in GPLE shifted to 1393.6 cm^{-1} . The band at 1609.7 cm^{-1} of -CO- stretch shifted to 1604.8 cm^{-1} due to interaction Fe in MNPs with GPLE [18]. These results are supported from phytochemical test of flavonoids and alkaloids. The presence of Fe-O was confirmed at 620.0 cm^{-1} [14,17] which was not formed before.

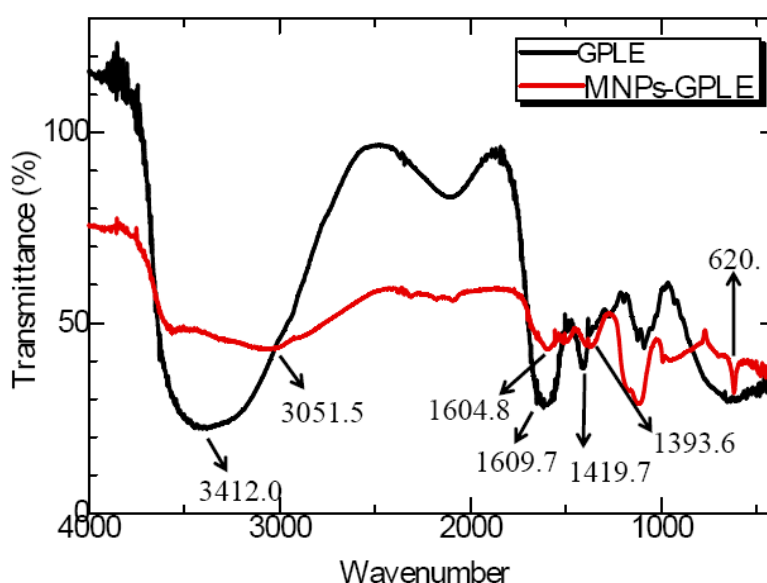


Figure 3. FTIR spectra of GPLE and MNPs-GPLE.

Figure 4 shows XRD pattern of synthesized MNPs-GPLE at $2\theta = 17.99^\circ; 30.11^\circ; 35.35^\circ; 43.48^\circ; 53.55^\circ; 55.78^\circ; \text{ and } 62.18^\circ$. These values are adjusted with American Mineralogist Crystal Structure Database (AMCSD) entry number 96-900-5841 (magnetite), 96-901-2693 (maghemite) and 96-101-1170 (wuestite) obtained crystal planes (111), (202), (311), (400), (422), (333), and (404) of magnetite with face-centered cubic (FCC) phase. The crystallite size of MNPs was calculated as 23.17 nm through Debye-Scherrer equation.

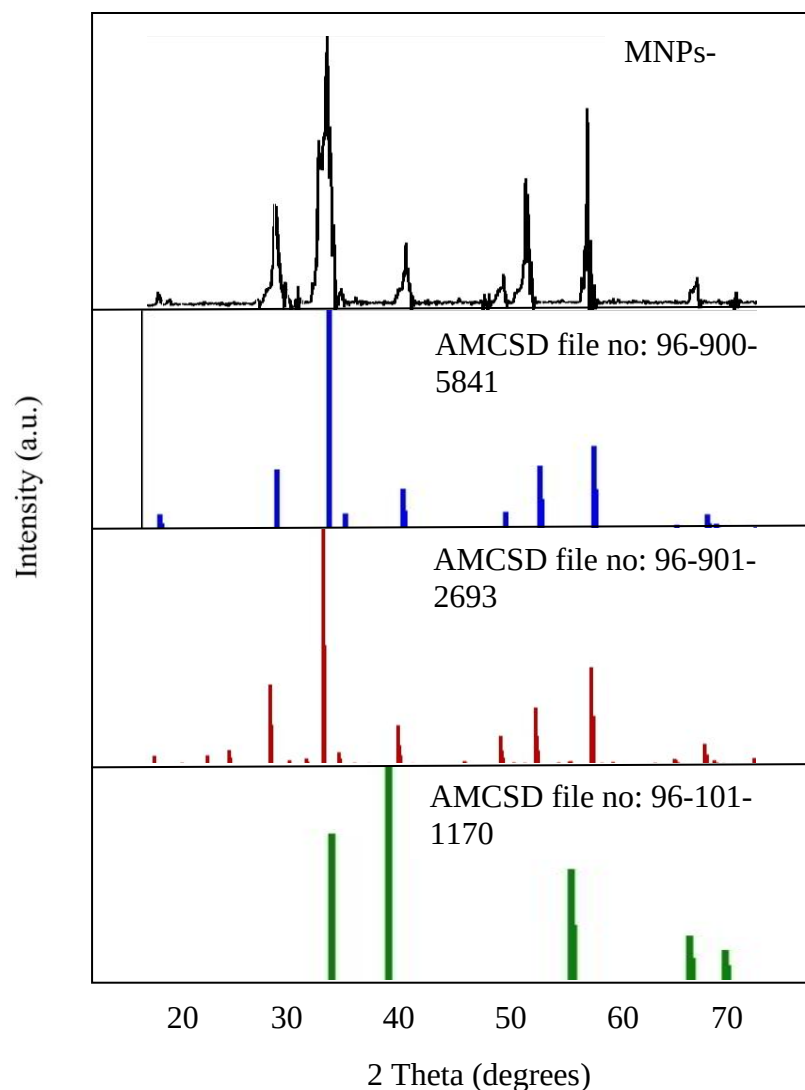


Figure 4. XRD Patterns of MNPS-GPLE, Magnetite, Maghemite, and Wuestite.

The crystalline MNPs were synthesized for the first time using GPLE involved the interactions of -OH and -NH groups in GPLE with MNPs. This result is potential as functional nanoparticles based on environmentally friendly method.

4. Conclusions

Magnetite nanoparticles, Fe_3O_4 (MNPs) were successfully synthesized using *Graptophyllum pictum* leaf extract (GPLE) by a simple and green method. GPLE contains alkaloids as a base source meanwhile flavonoids act as capping agent to prevent the MNPs agglomeration. The XRD analysis confirmed the MNPs crystal size of 23.17 nm. UV-Vis spectroscopy investigated MNPs formation at a maximum wavelength, λ_{max} 291 nm. FTIR spectroscopy showed the presence of biomolecules functional group of GPLE through interaction between -OH and -NH with MNPs. The green

synthesized MNPs contribute in medical fields such as drug carrier and targeted therapy. This green synthesis method is environmentally friendly, cheap and potential to produce other metal oxides.

Acknowledgements

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