Structural and bonding properties of honeycomb structure of composite nanoparticles Fe₃O₄ and activated carbon

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Abstract. Magnetite nanocomposite synthesis and activated carbon with the addition of polyvinyl alcohol (PVA) as an absorber of electromagnetic waves have been successfully carried out with variations in the thickness of 4 mm, 6 mm, 8 mm, and 10 mm. Characterization was carried out with XRD (X-Ray Diffraction), XRF (X-Ray Fluorescence), FTIR (Fourier Transform Infrared) and VNA (Vector Network Analyzer) with a working frequency of 3 GHz - 7.5 GHz. XRD characterization controls the formation of magnetite (Fe₃O₄) and determines the crystal size. The diffraction peaks of the absorber at (221), (331) and (440) with a size of crystal on 10.46 nm. Characterization of XRF shows the chemical composition of the absorber which indicates the formation of nanocomposites, FT-IR spectrum obtained by C = C alkene bond and C-H alkane derived from activated carbon, honeycomb paper and magnetite (Fe₃O₄). According to the data, it contributes to the formation of molecular bonds from nanocomposite absorber. Characterization of VNA was obtained by the best absorber with reflection loss value of -16.24 dB, the frequency of 5.77 GHz at a thickness of 10 mm.

1. Introduction

Utilization of electromagnetic waves (EM) in electronic devices such as telephones, cell phones, local area networks (LAN), and intelligent transport systems (ITS) results in increased radiation of electromagnetic waves to the environment [1-5]. Electromagnetic wave absorbent material technology is one of the technologies produced to handle electromagnetic interference (EMI) [6-8]. This technology has created a new material like radar absorbing material (RAM) [4-10]. This material absorbs electromagnetic waves. Until now iron as composite materials is materials that can be used as magnetic absorbers in electromagnetic waves [9-11].

Developing and Innovation of RAM continues to improve the quality of absorbance from the material. The characteristics of materials with good wave absorption are like the previous report by Qiang, R. et al. (2015), using Fe / carbon organic metal material with synthetic and carbonization methods [10]. Liu, Q. et al. (2016), using the same method using Fe / carbon organic metal Zn [9], and Lu, Y et al. (2015), Co / C nanocomposite material as an absorber [2].

Material structure engineering to find material that corresponds to EMI such as a sandwich structure [12], double-layer [13], pyramid structure [14] and others. Weight and thickness of materials are problems unavoidable factors, so that the application of material limited to certain applications. The honeycomb structure is widely used as an absorber structure with high strength and low density...
for dampers [1,6-7]. In this research, absorber material with the honeycomb structure, synthesis of magnetite nanoparticles (Fe₃O₄) by using the coprecipitation method will create a honeycomb absorber structure with frequency to handle EMI problems.

2. Experiment
2.1 Material
FeSO₄.7H₂O (Merck), FeCl₃.6H₂O (Merck), NH₄OH (Merck) 10 M purchased from Sigma-Aldrich, Purity of PVA (Polyvinyl alcohol) 99.5% (Brand), Aquabides, Paper honeycomb, Ethanol, Activated Carbon (AC) provided from local company PT. Indo Light in Indonesia with an average diameter <10 μm, purity> 95%, and a surface area> 240 m / g and pH specialized indicators (pH range 0-14).

2.2 Material Preparation
2.2.1 Synthesis of magnetite (Fe₃O₄)

![Figure 1. Synthesis of magnetite (Fe₃O₄)](image)

The magnetite synthesis process is shown in Figure 1. Synthesis Fe₃O₄ with coprecipitation method from FeSO₄.7H₂O and FeCl₃ 6H₂O materials. The ingredients are weighed, 2 grams of FeSO₄.7H₂O and 4 grams of FeCl₃, 6H₂O, respectively. Mix the ingredients into a 100 ml Erlenmeyer then dissolve with 30 ml of aquabides while stirring using a magnetic stirrer and hot plate. The solution was heated to 80°C and slowly added 60 ml of 10 M NH₄OH solution for 60 minutes. The use of high concentrated solutions will produce smaller magnetite sizes. After that, the black deposits obtained are filtered and washed using ethanol so that PH is neutral. The sediment was dried using a furnace for 1 hour at 78°C. Characterizing the XRD against the black powder to prove that the synthesis process was successful in obtaining Fe₃O₄ by matching the XRD images with the JCPDS standard (Joint committee on powder Diffraction Standard) and supported by several research results [11].
2.2.2 Manufacture of composite absorber magnetite nanoparticles and activated carbon

The composite absorber is made using magnetite (Fe₃O₄) and activated carbon as the main ingredients. Fe₃O₄ material was obtained from previous synthesis results. Activated carbon is crushed first to get finer results, then weighed as much as 4 grams, then dissolved with concentration 2% of PVA as much as 12 ml. After that, dip honeycomb paper which varies in thickness from 4 mm, 6 mm, 8 mm, and 10 mm into an activated carbon solution. Dry honeycomb paper using a furnace at 80°C for 1 hour. Mix 2 grams of Fe₃O₄ with 6 ml aquabides and then sterilize for 3 minutes. Pouring the solution on a honeycomb paper mold that contains activated carbon then heated with a temperature of 60°C for 5 hours. This heating is done to remove the water content on the honeycomb paper mold. The process of making composite absorber magnetite nanoparticles and activated carbon is shown in Figure 2.

![Figure 2. Making composite absorber](image)

2.3. Characterization

The elemental composition of the samples was determined by X-ray fluorescence (XRF) spectroscopy, a Thermo ARL QUANT'X EDXRF model. Crystallite size, bonding characteristics, and absorber properties of the sample in this study were determined by using X-ray diffraction (XRD), Fourier transform infrared (FTIR), and a vector network analyzer (VNA). For more detail about characterization by using XRD, FTIR, and VNA, see our previous published paper [15-16].
3. Results and Discussion
The sample characterization was carried out by XRD to determine that the synthesis process was carried out successfully to obtain Fe₃O₄ nanoparticles and to determine the crystal size in the sample. Characterization was carried out before and after making the absorber composite with an angular range 10° ≤ 2θ ≤ 70° as the XRD test results in Figure 3.

![XRD test results](image)

**Figure 3.** XRD test results

**Figure 3 (a), (b) and (c) shows the results of XRD test material before the composite absorber is made. Figure 3 (c) diffraction peak (221) for the Fe₃O₄ sample at 32.29° is the hkl index for α- Fe₂O₃ (hematite) which is indicated by the symbol *. This indicates that oxidation occurs at each synthesis process which produces Fe₂O₃ and Fe₃O₄. Qualitatively it can be seen that the α- Fe₂O₃ phase is relatively small compared to Fe₃O₄. The diffraction peaks in Figure 3 (c) are matched with the JCPDS standard (Joint committee on powder Diffraction Standard) and supported by several results of previous studies [11], it was identified that the sample synthesis process succeeded in obtaining Fe₃O₄. The diffraction peaks for the absorber are sequentially (221) angles 32.29°, (331) angles 35.48° and (440) angles 62.67° as reported in the study of F. Yazdani, et. al. (2010) [17] and H. C. Ling, et. al. (2013) [11]. In Figure 3 (d) there is an increase in the intensity of the diffraction peak due to the addition of Fe material from activated carbon (Table 1). The average crystal size of the magnetite composite absorber by using the Scherrer equation is 10.46 nm.

XRF characterization was carried out to determine the content of the elements contained in the study sample. Table 1 shows the chemical composition of the sample in making absorber, which obtained a very significant percentage increase in absorber due to the addition of Fe from the...
magnetite Fe₃O₄ material, and the activated carbon and honeycomb paper. From these results indicate that the achievement of the synthesis is carried out.

**Table 1. Chemical Composition of XRF Test Results**

| No | Element   | Chemical Composition (%) |
|----|-----------|--------------------------|
|    |           | Honeycomb Paper | Activated Carbon | Absorber Composite |
| 1  | Fe₂O₃     | 4.59           | 35.47           | 96.8               |
| 2  | CaO       | 61.22          | 6.98            | 0.674              |
| 3  | SiO₂      | 24.39          | 38.41           | -                  |
| 4  | TiO₂      | 2.69           | 2.36            | -                  |
| 5  | Al₂O₃     | 6.17           | -               | -                  |
| 6  | K₂O       | -              | 12.18           | 0.291              |
| 7  | LOI       | 0.94           | 4.6             | 0.41               |

**Figure 4. Spectrum of FT-IR test results**

FT-IR characterization is carried out before and after the manufacture of the composite absorber. **Figure 4** shows the FT-IR spectrum the functional groups. **Figure 4 (a)** and (b) shows the same type of bond. For the activated carbon, forming an O-H bond of hydrogen alcohol which intensity changing and widening. These are one of the characteristics of carbon produced by natural materials. N-H Amina bond at wavenumber 3417.86 cm⁻¹, C = C alkene bond at wavenumber 1641.42 cm⁻¹, C-H alkane bond at wavenumber 1427.32 cm⁻¹, C-O alcohol bond at wavenumber 1056 cm⁻¹, and C-H bond of the aromatic ring at a wavenumber of 500-750 cm⁻¹.
The magnetite characterization of \( \text{Fe}_3\text{O}_4 \) and absorber shows the same type of functional group bond as shown in Figure 4 (c) and (d). In the range of wave numbers 500-750 cm\(^{-1}\), Fe-O bonds are formed as a result of the successful synthesis process as shown into with the XRD and XRF results. In Figure 4 (d) shows that the absorber properties that are successfully created are the effects of the composite that has been integrated in the previous process. The shift from the characteristic of the absorption stretch to the absorber is caused by hydrogen bonds between hydroxyl groups in PVA as reported by A. Bualkar et. al. (2018) [8].

Figure 5. Reflection Loss Absorber Value

Table 2. Results of wave absorption test from composite absorber

| Thickness (mm) | Reflection Loss (dB) | Frequency (GHz) | Bandwith Frequency (GHz) |
|---------------|----------------------|-----------------|--------------------------|
| 4             | -12.55               | 5.72            | 1.20                     |
| 6             | -13.09               | 5.82            | 1.22                     |
| 8             | -15.15               | 5.75            | 1.41                     |
| 10            | -16.24               | 5.77            | 1.57                     |

VNA characterization is used to determine the absorption rate of the waves from the absorber by obtaining the Reflection Loss value. To find the optimal absorber thickness, it is necessary to calculate the maximum reflection loss from the absorber working frequency as reported by A. Bualkar et. al. (2018) [8]. In this study measurement of wave absorption with VNA with a working frequency of 3-7.5 GHz has been carried out as shown in Figure 5. The RL value in the absorber increases with the increase in absorber thickness where the greatest RL value is obtained at a thickness of 10 mm with the absorber working frequency in the range 5.5 -6 GHz, an increase in RL value may be due to the influence of AC [2, 7-11]. The increase in bandwidth frequency is also shown in accordance with the increase in absorber thickness. These results indicate that the absorber obtained from the synthesis of \( \text{Fe}_3\text{O}_4 \) (magnetite) nanoparticles with coprecipitation method is better than the results reported by S. Xie, et. al. (2016) [1] and W. H. Choi, at. al. (2014) [6].

4. Conclusion

Synthesis of composite magnetite nanoparticles and activated carbon with the coprecipitation method was successfully made. XRD results showed that the formation of the magnetite phase at the diffraction peak (221) was also supported by the XRF results, with an average size of 10.46 nm. FT-IR spectrum obtained by C = C alkene bond and C-H alkane derived from activated carbon, honeycomb paper and magnetite (\( \text{Fe}_3\text{O}_4 \)) so that it contributes to the formation of molecular bonds from
nanocomposite absorber. Characterization of VNA was obtained by the best absorber with reflection loss value of -16.24 dB, the frequency of 5.77 GHz at a thickness of 10 mm.

Reference
[1] Xie S, Ji Z, Yang Y, Hou G and Wang J 2016 Journal of Building Engineering 7 217-223
[2] Lu Y, Wang Y, Li H, Lin Y, Jiang Z, Xie Z, Kuang Q and Zheng L 2015 ACS Appl. Mater. Interfaces 7 (24) 13604–13611
[3] Huang Y, Ding X, Li S, Zhang N and Wang J 2016 Ceramics International 14 (5) 17116-17122
[4] Liu S T, Yan K K, Zhang Y H, Jin S D, Ye Y and Chen X G 2015 Journal of Magnetism and Magnetic Materials 394 266-273
[5] Lv J L, Zhai S R, Gao C, Zhou N, An Q D and Zhai B 2016 Chemical Engineering Journal 289 261-269
[6] Choi W H, Shin J H, Song T H, Lee W Y, Lee W J and Kim C G 2014 Electronics Letters 50 (4) 292-293
[7] Liu L, Fan C Z, Zhu N B and Zhao Z Y 2014 Appl. Phys. A 166 (3) 901-905
[8] Abdullah B, Ilyas S and Tahir D 2018 Hindawai Journal of Nanomaterials 9823263 6
[9] Liu Q, Liu X, Feng H, Shui H and Yu R 2016 Chemical Engineering Journal 314 (8) 320
[10] Qiang R, Du Y, Zhao H, Wang Y, Tian C, Li Z, Han X and Xu P 2015 Journal of Materials Chemistry A (25) 9
[11] Ling H C, Hu L T, Kai Z T, Guang L H, Hao L L and Juan Z W 2013 New Carbon Materials. 28(3) 184-190
[12] Bantsis G, Sikalidis C, Betsiou M and Xenos T, 2011 Ceram. Int. 37 3535-3545
[13] Zhang X Z, Sun W 2010 Cem. Concr. Compos 32 (9) 726-730
[14] Laukaitis A, Sinica M, Balevicius S and Levitas B 2008 Acta Phys. Pol. A 113 (3) 1047-1050
[15] Tahir D, Ilyas S, Abdullah B, Armynah B, and Kang H J, 2018 Journal of Electron Spectroscopy and Related Phenomena 229 47-51
[16] Tahir D, Ilyas S, Abdullah B, Armynah B, Kim K, and Kang H J, 2019 Mater. Res. Express 6 (3) 035705
[17] Yazdani F, Edrissi M 2010 Materials Science and Engineering B, 171 80-89