Research Article

Nanocomposites Fe/Activated Carbon/PVA for Microwave Absorber: Synthesis and Characterization

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Nanocomposites, activated carbon/polyvinyl alcohol (AC/PVA) filled with Fe to form Fe/AC/PVA, were characterized by using X-ray fluorescence (XRF), X-ray diffraction (XRD), Fourier transform infrared (FTIR), and vector network analyzer (VNA). The crystal orientations from Fe are (104), (110), and (200) and the bonding formations of AC are O-H, C-H, and C=C show existence in nanocomposites, which may be due to the Fe that has been filled the pore of AC via a chemical bond. 20% AC (3 mm in thickness) in nanocomposites shows higher performance absorption for C-band (4.65 GHz) with maximum reflection loss of −32.5 dB.

1. Introduction

Development of modern technology in electronic system and telecommunications has greatly increased the demand for electronic devices using 1–5 GHz range microwave radiation, which is resulting in serious electromagnetic interference (EMI) problems [1–6]. EMI causes device malfunctions, generates false images, and reduces the performance of the system-to-system coupling to a radar system. This has led to a search and creates a new material for electromagnetic (EM) wave absorber with the capability to prevent EMI.

Electromagnetic wave absorber used dielectric materials and magnetic materials that can prevent EMI. Dielectric materials include conducting polymers, carbon, and silicon carbide, which are more lightweight and have high-temperature stability but have poor absorption electromagnetic waves. Magnetic materials such as ferrite and metallic magnetic materials can be used as an electromagnetic absorber; however, the problem of magnetic materials is high conductivity and magnetization induced by electromagnetic waves [7–12]. For these reasons, it is better to use small particle carbonyl iron.

Iron (Fe) and carbon are commonly used as absorber electromagnetic waves [13–16]. Recently, many studies reported on composites such as organic metals Fe/Zn/carbon [1], Fe/carbon [2], and nanocomposite of Co/carbon [3] with reflection loss up to −29.5 dB. Reference [17] was reported to nanocomposite RGO/Fe_3O_4 with additional ZnO that shows EM wave absorption properties the RGO/Fe_3O_4 up to −57 dB at 13.5 GHz and the thickness of 2.0 nm. ZnO is attributed to the interfacial polarization and impedance matching but increases the dielectric losses at higher frequency and magnetic losses at lower frequency due to the unstable bonding at surface composition or valence state. The absorption properties are great challenges which were usually depended on the natural resonance and the eddy current effect of the unstable bonding at the surface composition or valence states. Although there are some works on carbon-based and iron-based materials for the electromagnetic wave absorption, it is still a significant challenge by using the pore of AC as a trap of magnetic particles to achieve high performance by stabilized bonding with surface composition or valence states. By this method, the surface composition or valence state will be more stable in reducing the natural resonance and eddy current effect to improve reflection loss. PVA was used to form continuous network polymer to produce thinner thickness and lighter weight. As long as we know, there is no study reported for absorber materials by

utilizing the pore of activated carbon (AC) as a trap of magnetic particles for stabilizing bonding at the surface composition or valence state.

In this study, PVA could improve the permeability by interfacial adhesion forming a continuous network by utilization of the pore of AC as a trap of metallic magnetic particle (Fe) for stabilizing bonding at surface composition or valence state and will raise polarization relaxation to increase the permittivity [5–10]. The proposed method in the fabrication of nanocomposite Fe/AC/PVA materials in this study is relatively easy-to-handle and economic; the method can be seen clearly in Figure 1.

The nanocomposite materials were characterized by using various techniques (XRD, FTIR, and XRF) to establish the relationship of composition and structural and chemical properties to the electromagnetic wave absorption performance. Electromagnetic wave absorber performance was measured by using vector network analyzer (VNA).

2. Experiments

2.1. Materials. PVA (polyvinyl alcohol) purity 99.5% was purchased from Merck. Iron powder (Fe) was purchased from Sigma-Aldrich in a form of carbonyl-iron powder, low in magnesium and manganese compounds, $\geq 99.5\%$, resistivity $9.71\,\mu\Omega\,cm$, impurities $\leq 0.01\%$ total nitrogen (N), and grain size $5–9\,\mu m$. Activated carbon (AC) was supplied from the local company PT. Cahaya Indo Abadi in Indonesia with a size of an average diameter $< 10\,\mu m$, purity $> 95\%$, and surface area > 240 m/g.

2.2. Sample Preparation. Activated carbon (AC) was crushed into powder by using a mortar and sieved with 200 meshes. For homogeneous nanocomposites, the Fe was mixed with AC by Retsch MM for 30 minutes at a frequency 10 Hz, which was controlled by XRF. The final mass of each sample is 15 grams in nanocomposite that consists of 10%, 20%, and 25% of AC.

2.3. Compaction Process. The samples were added 5 ml in 2% concentration of PVA for each sample and poured into the beaker. Samples were stirred at 50°C for 30 minutes by using a magnetic stirrer to obtain slurry form. The samples were pelleted (thickness 2 mm, 3 mm, and 4 mm) by using hydraulic compactor at a pressure of 50 kPa and then were cooled naturally for 5–7 minutes. Samples were annealed in a furnace at 80°C for 5 hours. Figure 1 shows the fabrication processes of nanocomposites for the microwave absorber.

2.4. Characterization. Crystallite size of the samples was determined from the X-ray diffraction (XRD) data. Spectra were collected on XRD spectroscopy (Shimadzu 7000) with CuKα radiation ($\lambda = 1.5405$ Å) over the angular range $15° \leq 2\theta \leq 80°$, operating at 30 kV and 10 mA. The elemental composition of the samples was determined by X-ray fluorescence (XRF) spectroscopy, a Thermo ARL QUANT'X EDXRF model. Fourier transform infrared (FTIR) spectroscopy was carried out on a IRPrestige-21 FTIR spectrometer (Shimadzu Corp.) equipped with a bright ceramic light source, a KBr beamsplitter, and a deuterated triglycine sulfate doped with L-alanine (DLATGS) detector. The measurements of the samples were collected over the range of $4000–600\,cm^{-1}$ and 16 coadded scans. All spectra of FTIR were in transmittance units. Electromagnetic wave absorber performance was measured by using vector network analyzer (VNA) (Rohde & Schwarz, ZVHB) with the frequency range from 2.5 GHz to 8 GHz.

3. Results and Discussion

Table 1 shows the chemical composition of iron powder (Fe), AC, and nanocomposites determined by using XRF spectroscopy. The amount of chemical oxide for pure AC is 35.47%, 38.41%, 12.18%, 6.98%, and 2.36%, respectively, for Fe_2O_3, SiO_2, K_2O, CaO, and TiO_2. Nanocomposites show Fe increase with the increasing amount AC, as we predict due to the highest amount of Fe_2O_3 in pure AC. Chemical composition in Table 1 indicated homogenates of nanocomposite in this study.

For nanocomposites, the aspect ratios of PVA as a matrix for composites (AC and Fe) and AC as a second matrix for Fe as fillers are important parameters and offer a large flexibility for design and property control. Nanocomposite with ~10% AC appears the element MgO probably comes from the PVA. AC is more attractive bonding due to the surface area, so for low AC content, not all Fe atoms filled the pore of AC and formed covalent bonding; by adding the PVA, some of these Fe atoms outside the pore of AC by interfacial adhesion form small area continuous network. For >10% AC, the amount of MgO decreases with increasing the amount of AC that may due to the amount of Fe particles that has enough filled the pore of AC and by using PVA as an interfacial adhesion forming uniform and strong continuous network bonding with AC and Fe particles. Figure 2 shows schematic illustrations of the pore of AC filled with Fe particles and PVA as a matrix for composites to form interfacial adhesion which could improve chemical bonding between Fe and AC and also supporting to reduce conductivity [12, 13]. The interfacial adhesion between matrix and filler in nanocomposites Fe/AC/PVA is also a great influence on the properties of composites: absorption capacity and structural properties. The continuous network bonding is beneficial to EMI shielding of nanocomposite as reported by Hoang A. S. (2011) [18] and Kuester et al. (2016) [19].

Structural properties from XRD spectra for 2θ from 10° to 65° can be seen clearly in Figure 3, for pure Fe, for pure AC, and nanocomposites for 10%, 20%, and 25% of AC. Several crystal orientations appear for pure Fe are (104), (110), and (200). For nanocomposites in Figure 3, the peaks are small shifted from pure Fe position to lower 2θ positions due to bonding formation changed in the composites [1, 8]. Contribution from Fe and AC in nanocomposites can be seen in Figure 3 by start symbol and solid circle symbol, respectively. The broad diffraction peak of 18° to 28° indicated the disordered carbon form which is a good characteristic to store electrical energy and may correspond with function of the pore of AC as a trap Fe particles [1, 9, 15]. PVA was used as interfacial adhesion which could improve chemical
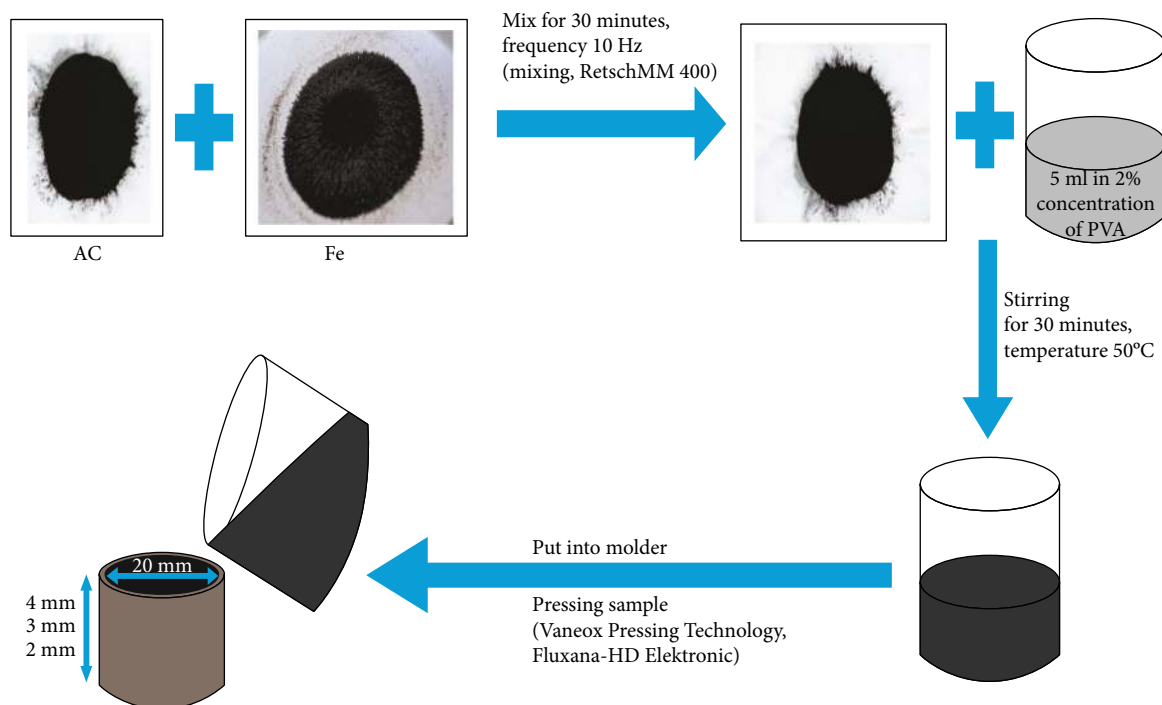
FIGURE 1: Fabrication processes of Fe/AC/PVA nanocomposites for electromagnetic wave absorber.

TABLE 1: Chemical compounds for pure Fe, AC, and nanocomposites determined by XRF spectroscopy. The total amount of chemical compounds is 100% with loss on ignition (LOI) by XRF spectroscopy which is up to 3.9%.

Chemical compound	Sample (composition (%))				
	Fe	Activated carbon (AC)	10% AC	20% AC	25% AC
Fe_2O_3	97.98	35.47	82.87	88.28	89.89
TiO_2	0.67	2.36	1.71	2.15	2.51
MnO	0.56	0.749	1.27	1.57	1.59
CaO	0.297	6.98	0.526	0.709	0.85
P_2O_5	0.356	—	0.27	—	—
K_2O	—	12.18	0.526	1.40	1.91
SiO_2	—	38.41	4.12	5.02	5.15
MgO	—	—	8.73	1.27	0.11

bonding with AC and Fe and will raise polarization relaxation to increase the permittivity [19–24].

We determined crystallite sizes from the XRD spectra by using Scherrer equation with the average crystallite sizes for each absorber, which are 15 nm for 10% AC, 24 nm for 20% AC, and 12 nm for 25% AC in nanocomposites Fe/AC/PVA.

FTIR spectra as shown in Figure 4 were recorded in the wavelength range of 4000 to 500 cm^{-1}. The FTIR results for Fe and AC prove that there is the same type of bond from the functional group. The intensity of bonding formation from Fe is increased with increasing the amount of AC in the nanocomposites as confirmed by the XRF results. In Figure 4(a), Fe shows the presence of Fe-O vibration at 583.6 cm^{-1}, C-N amine bonds at 1028.06 cm^{-}, C-OH bonds at 1554.63 cm^{-1}, and O-H bond at 3439.04 cm^{-1}. In Figure 4(b), AC shows the small Fe-O vibration bonds at 551.64 cm^{-1} and C-N amine shows the strongest peak at 1023.47 cm^{-1}. C-H vibration bond appears at 2879.72 cm^{-1} and the C=C aromatic ring is at 1681.93 cm^{-1}. The broad absorption peak for O-H bond is at 2526.75 cm^{-1} and the small peak at 3394.72 cm^{-1}. For nanocomposites in Figures 4(c)–4(e), the bonding formation is influenced by the Fe and AC as can be seen clearly for O-H, C-N, C=C, and Fe-O intensities, which are increased with increasing the amount of AC. Bonding formation of O-H that was shifted from 3439.04 cm^{-1} (Fe) to 3392 cm^{-1} (20% AC) and Fe-O from 583.6 cm^{-1} (Fe) to 586.4 cm^{-1} (20% AC) may correspond to functional of the pore of AC as a trap that has been filled with Fe particle via covalent bond, which is similar with that of reported by Zhang et al. (2017) [23]. The active functional group (C-N) at the surface will improve the dispersion of nanocomposites in PVA; thereby, Fe/AC/PVA possess resulting better magnetic network which provided multiple reflections and offered sufficient transmission path for PVA network to attenuate EM wave [23, 24]. AC was the dielectric loss of Fe/AC/PVA network, and the presence of PVA could further increase the dielectric loss due to dipole relaxation and interface relaxation [18–23]. AC was facilitating charge transfer and was removing the effect of natural resonance due to the strong bonding at the surface composition or valence state which increased the magnetic loss of the nanocomposites.

In order to find an optimum thickness of the absorber needed to measure the maximum reflection loss in a broad frequency region, the reflection loss values in different thickness were measured in this study. The ability of absorption has been measured by using VNA for the frequency range 2.5 GHz–8 GHz as shown in Figure 5. The value of reflection
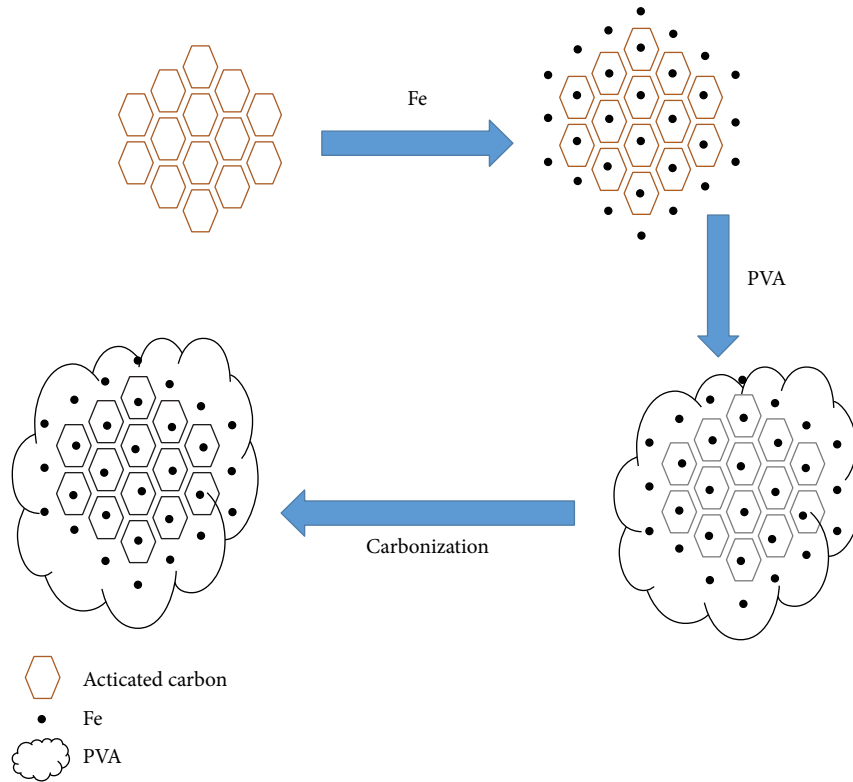
FIGURE 2: Schematic illustrations of the pore of AC filled with Fe and bonding formation of PVA.
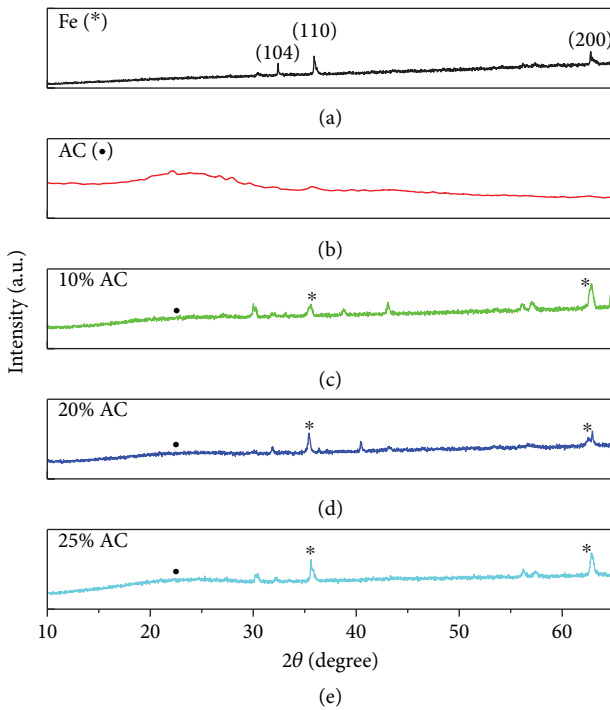

FIGURE 3: XRD spectra in this study: (a) Fe, (b) AC, (c) 10% AC, (d) 20% AC, and (e) 25% AC.(∗ indicated the reflection spectra from Fe and • from AC).

loss (RL) is strongly influenced by the amount of the activated carbon and the thickness of each nanocomposite. Figure 5 shows VNA spectra of the RL for each sample and shows the best absorption frequency in the range of C-band from 4.5 GHz to 5 GHz. The absorption ability (RL) shows an increase with increasing the thickness of the samples from 2 mm to 3 mm and reduces when the thicknesses reach 4 mm. Figure 5 shows that Fe (for 0% AC) shows 2 peaks at 5.7 GHz and 7. 2 GHz and RL about −15 dB to −18 dB. The second peak at 7.2 GHz shows the existence of natural resonance due to the unstable surface composition or valence states. For nanocomposites, the natural resonance is not appearing indicated that the pore of AC was facilitating charge transfer and stabilized the bonding at the surface composition or valence state [17]. 10% AC shows RL increase from −17.5 dB to −27.5 dB and decrease to −20 dB for thicknesses 2 mm, 3 mm, and 4 mm, respectively, similar case for 20% AC and 25% AC in Figure 5.

Bandwidth frequency corresponding to the reflector loss at −10 dB increased from 1.3 to 2.2 GHz as shown in Figure 5 by lines. AC could effectively improve the electromagnetic wave absorbing properties of the nanocomposites at the frequency range of 4.5 GHz–5 GHz. The effect of thickness of nanocomposites to the electromagnetic wave absorption properties in this study can be seen clearly in Table 2.

From RL in this study shows that the highest RL capacity is −32.5 dB for sample 20% AC with thickness 3 mm in C-band compared with recent results for RL −29.5 dB and − 22.6 dB reported by Liu Q. et al. (2017) [1] and Qiang R.
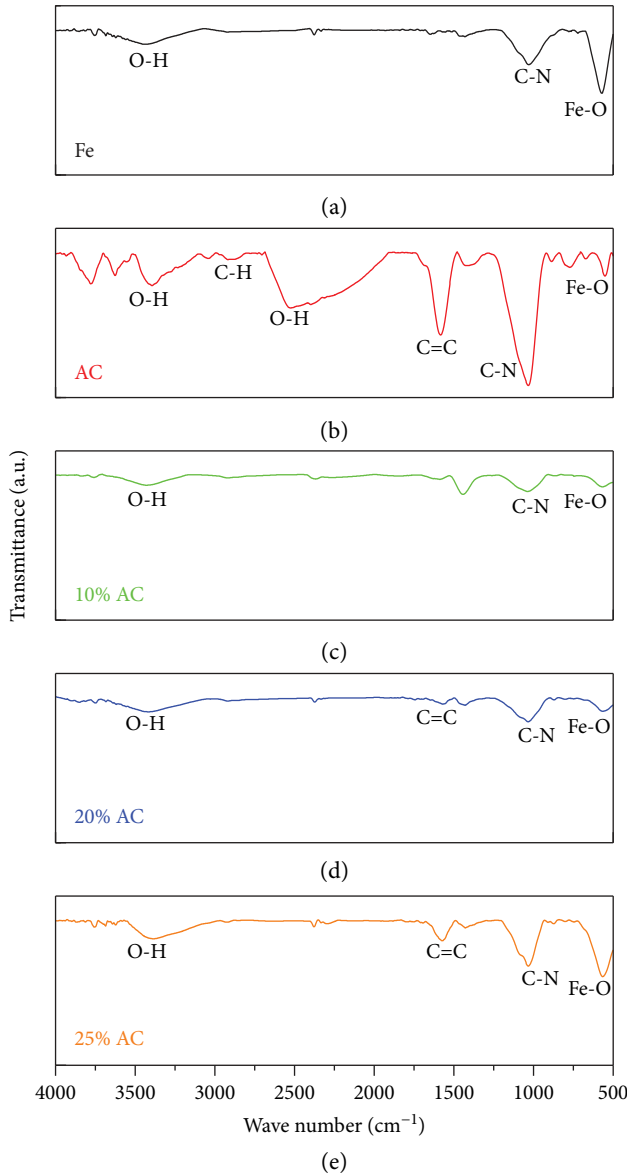
FIGURE 4: FTIR spectra in this study: (a) Fe, (b) AC, (c) 10% AC, (d) 20% AC, and (e) 25% AC.
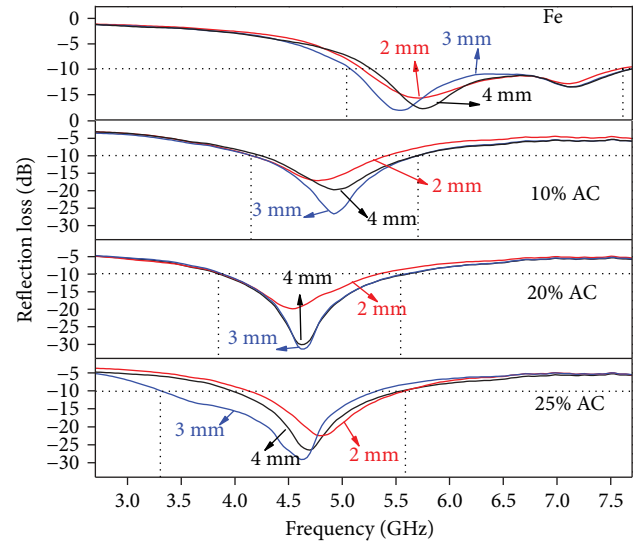

FIGURE 5: Electromagnetic (EM) waves absorbing properties in this study for 0% AC (Fe), 10% AC, 20% AC, and 25% AC in Fe/AC/PVA nanocomposites with different thickness: 2 mm, 3 mm, and 4 mm.

TABLE 2: Electromagnetic (EM) wave absorbing properties of Fe/AC/PVA nanocomposites in this study.

Samples	Thickness (mm)	Reflection loss (dB)	Frequency (GHz)	Frequency bandwidth (GHz)
Fe	2	−15.61	5.71	2.41
	3	−18.02	5.52	2.58
	4	−17.69	5.75	2.37
10% AC	2	−17.08	4.76	1.30
	3	−26.61	4.92	1.56
	4	−18.80	5.06	1.45
20% AC	2	−19.85	4.52	1.55
	3	−32.50	4.65	1.76
	4	−31.30	4.62	1.76
25% AC	2	−22.48	4.78	1.46
	3	−29.07	4.62	2.20
	4	−26.44	4.70	1.64

et al. (2015) [2], respectively. For all samples with 3 mm thickness that show high RL may be due to the surface area of AC that has a good ability to store electrical energy and PVA may contribute to the permeability by forming continuous network uniform that makes easy to penetrate the magnetic particle in the nanocomposites which are supporting to reduce natural resonance and conductivity of the magnetic material (Fe) [1, 2, 8, 15]. The above results imply that we can get a higher electromagnetic absorption in the range of C-band from 4.5 GHz–5 GHz by varying the thickness and the content of the AC in Fe/AC/PVA nanocomposites to meet different demands [21]. In summary, the AC in the Fe/AC/PVA nanocomposites plays an important role in the electromagnetic wave absorbing performance. However, the detailed effects of the thickness, the bonding mechanism with Fe particles, conductivity of

nanocomposites, and the composition of AC on the electromagnetic wave absorption properties of Fe/AC/PVA nanocomposites still need deeper investigations.

4. Conclusions

Fe/AC/PVA nanocomposites were successfully synthesized by simple method utilizing the pore of AC as a trap of Fe particle and PVA was used to form continuous network with the thicknesses that were varied: 2 mm, 3 mm, and 4 mm. Fe has been filled the pore of AC via a chemical bond and shows stabilized bonding at the surface composition or valence state as confirmed by the existence of O-H, C-H, and C=C bonding formations in nanocomposites from the FTIR spectra. We demonstrated that Fe/AC/PVA

nanocomposites have very good electromagnetic wave absorption properties for frequency range of 4–6 GHz. The reflection loss value of Fe/AC/PVA nanocomposites for the 20% AC and the thickness 3 mm is −32.5 dB which effectively improves the electromagnetic wave absorbing properties for C-band at the frequency of 4.65 GHz.
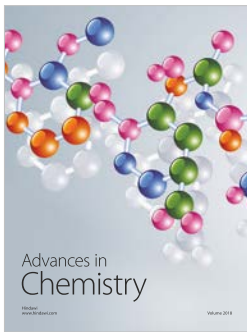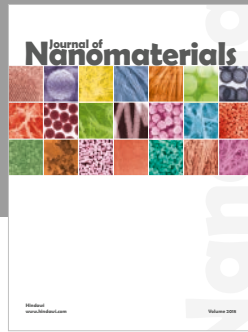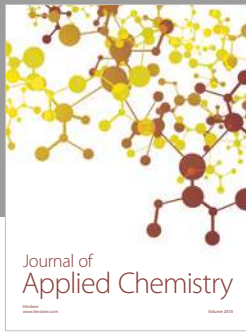
Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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