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Microwave Absorption Properties of Fe_2O_3 and Fe_2O_3/CNTs Nanocomposites

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ABSTRACT

The dielectric loss properties of recycled Fe_2O_3 (iron oxide nanoparticles, hematite) can be enhanced by using it as a catalyst for synthesizing carbon nanotubes (CNTs) from waste cooking oil. Hematite was successfully processed using the high-energy ball milling (HEBM) method for 12 h and sintered at 600 °C for 4 h. This work aimed to improve the electromagnetic (EM) wave absorption characteristics in the frequency range of 8−18 GHz by increasing interfacial polarization and dielectric loss resulting from the growth of CNTs. Our results indicated that Fe_2O_3/CNTs showed the highest microwave absorption, exceeding 99.9%, with a minimum reflection loss of –15.05 dB and an absorption bandwidth of 1.68 GHz. The microwave absorption properties of these nanocomposites were attributed to the combined effects of dielectric loss from CNTs and magnetic loss from hematite.

Keywords: hematite (Fe_2O_3); carbon nanotubes (CNTs); reflection loss (RL); microwave absorption properties

INTRODUCTION

With the advancement of microwave technology, the high emission of electromagnetic (EM) waves into the atmosphere poses a severe health risk, which is harmful to people's health and affects the normal operation of electronic equipment. Therefore, EM protection and detection have garnered great attention [1,2], motivating the development of novel EM wave absorbers with low thickness, high performance, and a broad effective absorption band. Because of their good permeability and higher saturated magnetization, iron oxide ferrites can be an effective microwave absorber (MA) used to limit skin effects in the high frequency EM bands. Among these ferrites, Fe_2O_3 has been discovered to be one of the low-cost materials that possesses a magnetic properties, which is being explored as a viable solution to tackle these issues[3, 4]. The raw absorbent materials used in this study were magnetic materials that were easily derived from industrial waste via milling techniques. These materials were explored as effective microwave absorbing materials. High-purity hematite was initially reduced to micron-sized particles using conventional milling, then further refined to nanoparticles via high-energy ball milling (HEBM). An earlier report aimed to investigate the effect of microstructural variations on the microwave absorption properties of recycled α-Fe_2O_3, following the application of a high-energy ball milling technique to modify particles into nanopowders [3]. According to their results, a positive correlation was identified that connects microstructural variations with relatively complex permittivity and power loss. It could be exploited to tune the microwave absorption properties of the recycled α-Fe_2O_3 particles. In addition, synthesized from mill scale shavings, α-Fe_2O_3 particles were reduced to 16.2 nm through high-energy ball milling. Recycled α-Fe_2O_3/PCL nanocomposites, made via melt-blending were analyzed for their complex permittivity and power loss. Nanocomposites with higher α-Fe_2O_3 content exhibited superior permittivity and power loss (11.25–16.37 dB), showing promise for microwave noise reduction due to their cost-effectiveness, and low density. Moreover, recycled α-Fe_2O_3 surpassed commercial α-Fe_2O_3 in complex permittivity at 1–4 GHz and 8–12 GHz, suggesting potential appliction as fillers in polymeric composites to reduce costs while maintaining absorption efficiency [5].

From a different aspect, the hematite materials alone may not meet all the requirements of an MA. Consequently, we aspire to improve the MA properties by carbon nanotubes (CNTs) growth on ferrite materials [6,7]. The aim was to obtain a wide absorption bandwidth or reduce both the thickness and the loading percentage. Furthermore, the carbon source also plays an important role for the growth of CNTs, where it is important to use carbon sources which are cheaper, easily accessible, and available in large amount like waste cooking oil. In this context, a recent study focused on the development of hematite (Fe_2O_3)@carbon nanotubes (CNTs)/polyacrylamide hydrogel composites with favorable flexibility and biocompatibility. These composites demonstrated exceptional microwave absorption performance, even at low load ratios of Fe_2O_3@CNTs [8]. Therefore, the objective of this work was to prepare Fe_2O_3/CNTs composites with complementary dielectric and magnetic loss properties to utilize their synergistic effect and enhance the microwave absorption (MA) properties of the two-component composites.

MATERIALS AND METHODS

The methods of this work were divided into two stages. First, the starting raw powder materials used to produce Fe_2O_3 were industrial waste. The powder was milled using HEBM and purified using a magnetic separation technique, following similar routes as in previous reports [9-11]. Secondly, the Fe_2O_3 powder acted as a catalyst, while waste cooking oil served as a carbon source and argon as a carrier gas to grow carbon nanotubes (CNTs) via the modified chemical vapor deposition (CVD) process, following the method of Ismail et al. [12].

The microwave-absorbing properties of all prepared composites were investigated by using vector network analyzers (VNA) in a continuous frequency range of 8–12 GHz within the X-band frequency range and 12–18 GHz within the Ku-band frequency range. The Fe_2O_3 sample were prepared for MA measurement by incorporating 70 wt. % Fe_2O_3 into 30 wt. % epoxy resin. While the hybrid nanocomposites Fe_2O_3/CNTs were prepared by blending Fe_2O_3/CNTs powders with an epoxy resin in a 12:88 weight ratio. The mixtures obtained by mixing the Fe_2O_3 and Fe_2O_3/CNTs particles, epoxy resin, and hardener were immediately poured into a standard waveguide rectangular mould for the X and Ku bands. The mould was designed to have specific inner dimensions, which were

23 × 10 mm^2 for the X band and 15 × 7 mm^2 for the Ku band. This work was done by testing the composite materials at a thickness of 2 mm. The XRD instrument used in this work for the analysis of phrase and structural analysis of prepared composite samples is a Philips X'' pert Diffractometer model 7602 EA Almelo, which emits CuK_α radiation source with a wavelength of 1.5418 Å within the range of $2\theta = 20°–90°$. The obtained XRD spectra were interpreted using the database of the JCPDS data. Panalytical Highscore Plus was used to characterize and interpret the X-ray diffraction (XRD) data. The microstructures of the prepared samples were examined using an FEI NOVA NanoSEM230 (FESEM) with an energy-dispersive X-ray (EDX) system.

RESULTS AND DISCUSSION

Figure 1(a) shows the XRD patterns of hematite, clearly showing only the Fe_2O_3 phase with the rhombohedral reflections, indicating the crystalline nature of the sample and that no other phase transformation has occurred. Fe_2O_3 samples detected in the patterns were matched with the diffraction spectrum of 98-004-0476 with the peak label at 104 (33.02°), 110 (35.46°), 113 (40.82°), 116 (53.08°), 214 (62.25°), 030 (63.54°). Figure 1(b) displays the XRD pattern for Fe_2O_3/CNTs which shows the iron carbide reflection peaks. All diffraction peaks were identified and found to correspond to the planes of the orthorhombic structure of Fe_3C. The diffraction peaks at 2θ=26.27 and 39.75 are respectively assigned to the (111) and (002) crystal planes of the graphitized carbon structure of CNTs. In contrast, other diffraction peaks in the range of 2θ = 37.56°, 42.79°, 44.54°, 45.75°, 48.99°, 51.81°, 54.30°, 57.84°, 70.76° and 74.91° matches well with the (121), (211), (220), (112), (221), (022), (301), (231), (232), and (420) reflections of the standard fcc phase of iron carbide (space group: Pnma (62), JCPDS No. 07-9759). No peaks corresponding to impurities are detected.

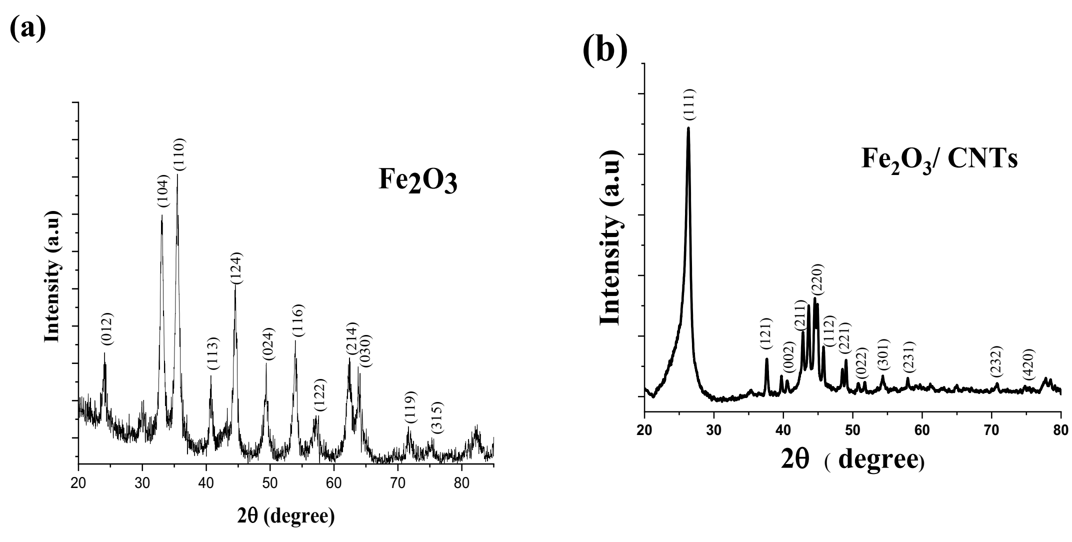
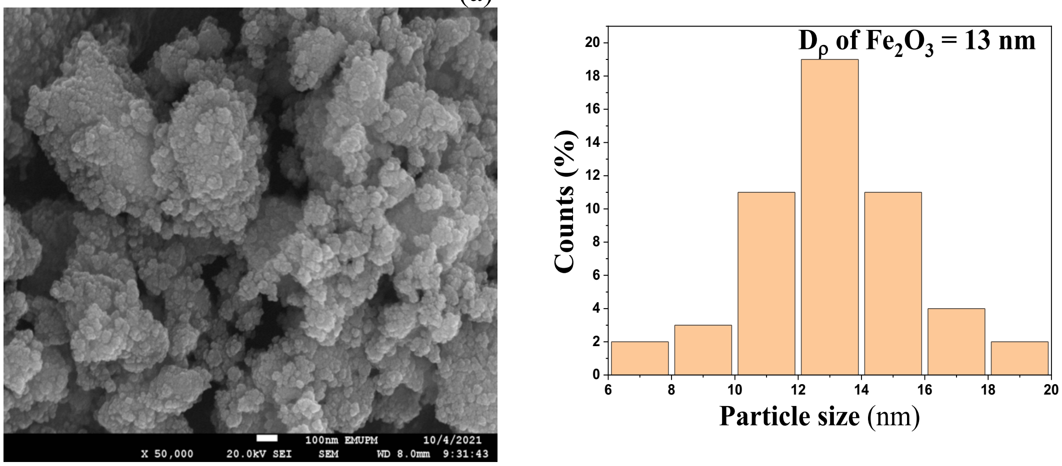


Figure 1. The XRD pattern of (a) Fe_2O_3 (b)and Fe_2O_3/CNTs

The microstructure of the Fe_2O_3 nanoparticles in Figure 2(a) indicates that the grains are quite similar to spherical particles. Since the Fe_2O_3 powder was repeatedly flattened, cold-welded, fractured, and re-welded throughout the rigorous work-hardening process, certain grains showed highly agglomerated particles and irregular shapes with an average size of 13 nm. Figure 2(b) shows the overall surface morphology of the as-grown CNTs, where the area density of the deposited CNTs was high and entangled, and the CNT structures had a randomly oriented, spaghetti-like morphology. Elemental composition of the Fe_2O_3/CNTs nanocomposites obtained from EDX analysis confirmed the presence of Fe, C, and O. Figure 2(b) indicated the presence of C, O, and Fe as the major elemental components throughout the entire region of the Fe_2O_3/CNTs sample.

(a)



(b)

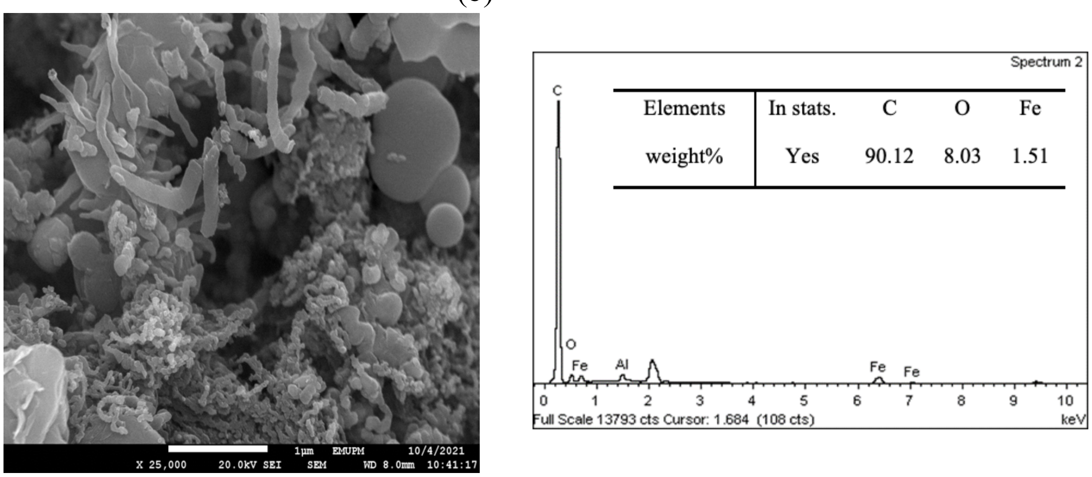


Figure 2. FESEM micrographs of (a) Fe_2O_3 and (b) Fe_2O_3/ CNTs

The absorption properties are depicted as plots of the relationship between reflection loss (RL) and microwave frequency, measured between 8 and 18 GHz (X- and Ku-band) at a thickness of 2 mm (Figure 3). The absorbing performance of the prepared samples (Fe_2O_3 nanoparticles and Fe_2O_3/CNT hybrids), with frequency bandwidth (GHz) (RL<−10 dB) and absorption (%), are displayed in Figure 4. The transmission line theory was used to clarify the electromagnetic wave absorption properties, and RL characteristics were calculated according to equations (1) and (2):

$$Z_{in} = Z_0 \sqrt{\mu_r / \varepsilon_r} \, tanh \left[j \left(2\pi ft / c\sqrt{\varepsilon_r / \mu_r} \right) \right] \quad (1)$$

$$R_L = 20 \log \left| \frac{(Z_{in} - Z_0)}{(Z_{in} + Z_0)} \right| \quad (2)$$

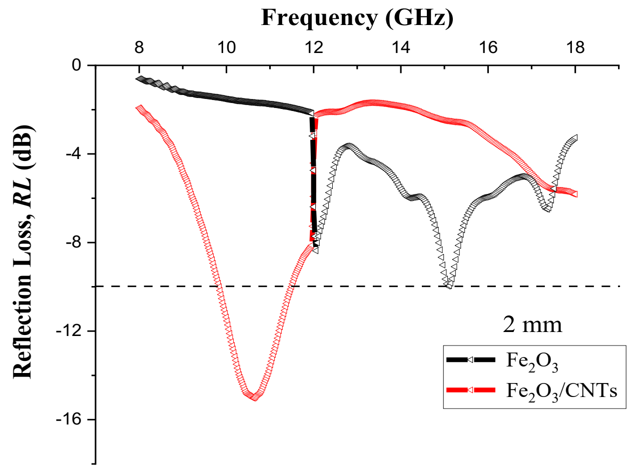


Figure 3. The behavior of the microwave reflection loss vs frequency of Fe_2O_3 nanoparticles and Fe_2O_3/ CNTs

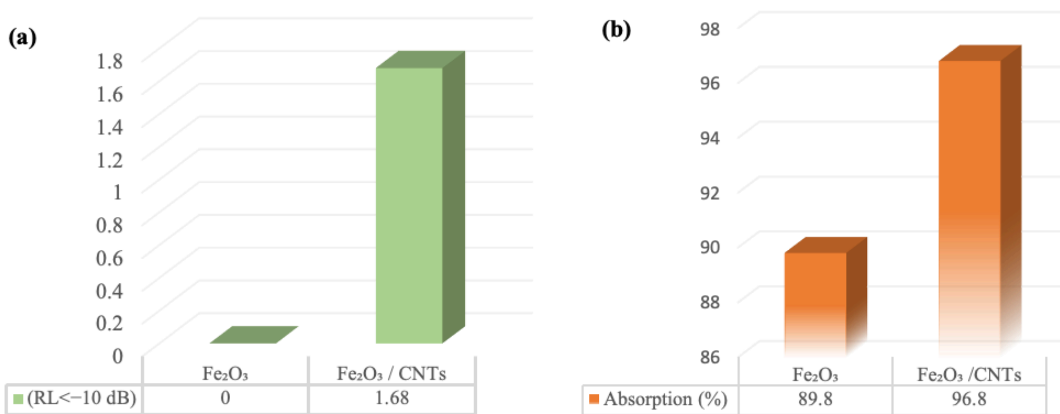


Figure 4. MA properties of synthesized (a) Fe_2O_3 and (b) Fe_2O_3/ CNTs nanocomposites

The results clearly showed that a lower value of RL was obtained for the Fe_2O_3 powder. However, with the 2 mm thickness sample of Fe_2O_3/CNTs, the reflection loss peaks shifted to a lower frequency, and the sample showed the strongest absorption, with a minimum reflection loss of −15.05 dB at 10.64 GHz and the broadest bandwidth at <−10 dB among all samples, around 1.68 GHz. These results indicate that the effective absorption bandwidth and excellent electromagnetic absorption performance allow Fe_2O_3/CNTs to meet the requirements of a high-performance absorbing material. The enhancement of the microwave absorption property of these samples was due to the increase in the dielectric and magnetic losses and was attributed to the impedance matching in the investigated frequency and thickness range. The synergy between hematite extracted from mill scale waste and CNTs synthesized from cooking oil waste enhanced and provided a more reliable and consistent composition for microwave absorption. The Fe_2O_3/CNTs nanocomposite leverages the magnetic properties of ferrites, and the dielectric loss properties of CNTs catalyzed by waste materials to give exceptional microwave absorption performance reach to 96.8% in an ultra-broad bandwidth. The absorption parameters in Figure 4 were calculated by the following equation:

$$Absorption\,(\%) = (1 - |S_{11}|^2) \times 100 \quad (3)$$

where S_{11} parameter is the measured scattering response of an electromagnetic signal directed back towards the radiation source via the signal port.

CONCLUSIONS

The novel Fe_2O_3/CNTs nanocomposites have been successfully fabricated via a modified chemical vapor deposition method with enhanced microwave absorption properties. The Fe_2O_3 hybrid material exhibited considerable electromagnetic absorbing ability between 8 and 18 GHz frequency. The minimum RL reaches −15.05 dB at 10.64 GHz with a layer thickness of 2 mm, and the absorption bandwidth with the RL values below −10 dB was about 1.68 GHz. The introduction of CNTs resulted in the increase of dielectric loss and improved the impedance matching of the composite. The results clearly showed that a lower value of RL was obtained for the Fe_2O_3 powder. However, with the 2 mm thick sample of Fe2O3/CNTs, the reflection loss peaks shifted to a lower frequency, and the sample exhibited the strongest absorption, with a minimum reflection loss of –15.05 dB at 10.64 GHz and the broadest bandwidth at <−10 dB among all samples, around 1.68 GHz. These results indicate that the effective absorption bandwidth and excellent electromagnetic absorption performance allow Fe_2O_3/CNTs to meet the requirements of a high-performance absorbing material.

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