



Graphene@Ni_{0.5}Co_{0.5}Fe₂O₄ hybrid framework with enhanced interfacial polarization for electromagnetic wave absorption

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ABSTRACT

Fabrication of smart materials for electromagnetic (EM) wave absorption has been propounded as efficient EM interference and pollution mitigation method. Herein, a porous lightweight graphene@Ni_{0.5}Co_{0.5}Fe₂O₄ composite was prepared via a coprecipitation method. The results show that Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticles are homogeneously dispersed and anchored on the graphene flakes. Investigation of the EM waves absorption properties of the material at different filling in paraffin reveals that at 15 wt% loading, the composite absorbs large percent of the EM waves at minimal thickness. The composite attains optimum reflection loss peak −44.7 dB at 17.45 GHz, with 1.5 mm thickness. This enhanced EM wave absorption performance (at lesser thickness than Ni_{0.5}Co_{0.5}Fe₂O₄ in literature) could be ascribed to interfacial polarization and a good impedance match arising from unique pore configuration of the dielectric (graphene) and magnetic (Ni_{0.5}Co_{0.5}Fe₂O₄) composite. These results indicate that the lightweight G@Ni_{0.5}Co_{0.5}Fe₂O₄ composites with strong absorption at reduced thickness is an efficient absorber for high-frequency EM wave attenuation.

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1. Introduction

The demand for smart electronics, wireless communication, and the proliferation of other advanced technologies utilizing electromagnetic (EM) waves are rapidly increasing and progressing. Though these technologies are convenient and abundant, their massive use has resulted in anthropogenic EM noise that affects human health, causing serious diseases like cancer and brain tumors [1]. According to some recent studies, the risk of exposure to EM radiation is high in pregnant women and children. Another consequence of the incessant use of EM applications is EM interference (EMI) that affect the proper functioning of electronic

appliances as a result of radiation from nearby EM power sources [2]. In this case, the effect could be fatal if the harm is caused to a life-supporting device, such as in hospitals. One effective way to suppress EMI is the use of EM wave absorbing material that can dissipate this unwanted EM radiation. Generally, EM wave absorbing materials are of two types, dielectric and magnetic. The use of the traditional EM shielding materials such as aluminum and silver are restricted by their bulkiness, rigidity, high corrosion rate, and difficult processing [3]. Also, they have high conductivity which favors surface reflection of EM waves, which results in secondary EM pollution [2]. Hence, recent advances in EM wave absorption applications have focused on the use of carbon, ceramic, and polymer matrix to achieve a set of desired properties which include strong and broad absorption range at low density and minimal thickness.

Accordingly, carbonaceous materials like carbon nanotubes, graphene, and carbon fiber [4–6] have shown superior properties because of their proper conductivity, vast surface area, and abundance. Among these allotropes, graphene which is composed

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of two-dimensional graphite layers has superior properties like remarkable lightweight, increased charge carriers mobility, excellent electrical conductivity, and unique porous morphology, which makes it a potential EM wave absorber [7,8]. Some studies have utilized several ways to prepare graphene and its derivatives like graphene oxide (GO) and reduced graphene oxide (rGO) as EM waves absorber. According to a report, the EM wave absorption of chemically rGO could reach -6.9 dB at 7 GHz [9]. Another study by Sudeep et al. [10] shows that the EM wave absorption capacity of fluorinated graphene oxide could reach a minimum reflection loss (RL) of -37 dB at 3.2 GHz, however, the thickness is as large as 6.5 mm. Since the EM wave absorption ability demonstrated by these sole graphene materials are not satisfactory, lots of research groups have reported composite of graphene with magnetic and/or dielectric fillers with improved EM wave absorption ability [11,12]. For instance, Chang et al. [13] synthesized rGO decorated zinc oxide nanowires (rGO/ZnO_{nws}) via a hydrothermal and freeze-drying method. The absorber attained minimum RL of -27.8 dB at 9.57 GHz with a 4.8 mm absorber thickness. Wang et al. [14] presented a NiO@graphene composite synthesized via hydrothermal route. At an absorber loading of 25 wt percent (wt%), they reported a minimum RL of -59.6 dB at 14.16 GHz with 1.7 mm matching thickness. In another report by Wei et al. [15] RGO/ZnO nanocrystals attained a minimum RL of -54.2 dB with 2.4 mm absorber thickness. Motivated by these studies, we report a novel graphene nickel-cobalt ferrites composites (G@Ni_{0.5}Co_{0.5}Fe₂O₄) prepared by a simple in-situ coprecipitation method. This study aims to increase the impedance matching and interfacial polarization on the graphene nanoflakes with the inclusion of Ni_{0.5}Co_{0.5}Fe₂O₄. The presence of the Ni_{0.5}Co_{0.5}Fe₂O₄ on the graphene surface increases the number of active sites for interface polarization and favors dielectric relaxation. In addition to the porous structure exhibited by the composite, the homogenous distribution of the magnetic Ni_{0.5}Co_{0.5}Fe₂O₄ on graphene surface improves impedance matching of the composite to achieve satisfying EM wave absorption. A composite of 15 wt% G@Ni_{0.5}Co_{0.5}Fe₂O₄ in paraffin displays superior EM wave absorption performance. This route is easy, cheap, and environmentally friendly making the reported composites a promising candidate for broadband EM wave absorption applications.

2. Experimental

2.1. Synthesis of G@Ni_{0.5}Co_{0.5}Fe₂O₄ composites

Graphene flakes, nickel (II) nitrate hexahydrate ((Ni(NO₃)₂)·6H₂O), iron (III) nitrate nonahydrate ((Fe(NO₃)₃)·9H₂O), cobalt nitrate (II) hexahydrate ((Co(NO₃)₂)·6H₂O) and ammonium hydroxide (NH₄OH, 30 wt%) were purchased from R&M chemicals. To synthesize the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composites, a facile coprecipitation method was utilized followed by high-temperature calcination. The graphene flakes (0.5 g) were first dispersed into 80 ml of deionized (DI) water through ultrasonication treatment for 2 h to produce layered graphene nanosheets. A separate solution of the ferrite precursors was prepared by dissolving Fe(NO₃)₃·9H₂O, Co(NO₃)₂·6H₂O, and Ni(NO₃)₂·6H₂O (with a proportion of $4:1:1$) into 50 ml DI water. Subsequently, the ferrite solution was mixed with the graphene suspension and further ultrasonicated for 2 h. Afterward, the solution temperature was raised to 80 °C, and 5 ml of NH₄OH was gradually poured into the solution under constant magnetic stirring. Subsequently, the solution was stirred continuously for 2 h at 80 °C before collecting the sample and cleaned several times using ethanol and DI water. The black product was finally calcined at 450 °C for 2.5 h under nitrogen (N₂) gas in a tube furnace leading to the final formation of G@Ni_{0.5}Co_{0.5}Fe₂O₄ composites.

2.2. Characterization

Malvern Panalytical X-ray diffraction spectrometer (XRD) operating with Cu K α radiation ($\lambda = 0.15406$ nm, 40 mA, and 45 kV) was used to study the crystalline structure of the sample. To visualize the morphology of the sample Field Emission Scanning Electron Microscopy (FESEM) characterization was done on a Zeiss Supra 55VP. Saturation magnetization and coercivity of the sample were measured using a vibrating sample magnetometer (VSM 648 series, Lakeshore) at room temperature. X-ray photoelectron spectroscopy was done on a Thermo scientific K-alpha X-ray photoelectron spectrometer to investigate the chemical composition of the sample. Structural analysis was done using transmission electron microscopy (TEM) (JEOL JEM 2010F at 200 kV). To investigate the type of carbon in the sample, the Raman spectrum was recorded with Horiba Jobin Yvon HR800. Keysite E5071C vector network analyzer (VNA) was used to measure the EM properties of the sample at X-band (8.2 – 12.4 GHz) and Ku-band (12.4 – 18 GHz) frequency range. In preparing the sample for EM properties measurement, the G@Ni_{0.5}Co_{0.5}Fe₂O₄ was homogeneously dispersed in paraffin wax at different loading of 5 wt%, 10 wt%, and 15 wt% designated as GNC@5, GNC@10, and GNC@15, respectively. Each sample was compacted into a rectangular shape with dimensions of 1.0×2.3 cm (X-band) and 0.8×1.60 cm (Ku-band) and snugly fitted into the WR90 and WR-62 waveguides for EM measurement.

3. Results and discussion

3.1. Structural and compositional analysis

Fig. 1 displays the X-ray diffraction pattern of the as-prepared and calcined samples. The XRD spectrum of the sample before calcination shows the graphene peaks, whereas the ferrite peaks are still concealed due to low ferrite crystallinity before calcination. A characteristic peak at $2\theta = 11^\circ$ is allotted to the (001) plane of the graphite phase. Another observed diffraction peak at 26.0° shown in the XRD pattern can be assigned to the reflection of the plane (002) and matches well with a report (ICDD 00-003-0864) in the crystallographic database. Whereas the XRD pattern of the calcined sample displays high ferrite crystallinity, with the crystal reflection overshadowing the graphene pattern. The XRD reflection pattern of G@Ni_{0.5}Co_{0.5}Fe₂O₄ composites show crystalline peaks at 18.3 , 30.5 , 36.0 , 43.6 , 57.8 , 63.5 , and 73.9° , which are indexed to crystal planes (111), (220), (311), (400), (511), (440), and (533) of typical cubic spinel phase (JCPDS 10-0325). The average crystallite size of the Ni_{0.5}Co_{0.5}Fe₂O₄ particles was calculated from the XRD result as 9.5 nm, using the Scherrer equation $D = k\lambda/\beta \cos \theta$. Where D is the crystallite size, k (0.94) is Scherrer constant relating to the particle shape, λ (0.15406 nm) is the wavelength of the X-ray source, β is full width at half the maximum peak, and θ is the angle correlating to the highest diffraction peak. The peak shifts to lower 2θ around 35° and 60° observed after calcination is due to strain-induced by crystal distortion after the oxidation of the ferrites incorporated within the graphene system. The average lattice strain $\Delta\xi/\xi$ is calculated as 0.4 using the formula [16].

$$\frac{\Delta\xi}{\xi} = \frac{\beta}{\tan \theta} \quad (1)$$

FT-IR spectra of the as-synthesized and G@Ni_{0.5}Co_{0.5}Fe₂O₄ composites samples are presented in Fig. 1b. In the un-calcined sample, a strong peak at 1383 cm⁻¹ is attributed to C–H symmetric formation [17]. The vibration band at 1621 cm⁻¹ is traceable to the C=C stretch or the vibration of existing aromatic domains. Meanwhile, a small shoulder peak at 1727 cm⁻¹ suggests the

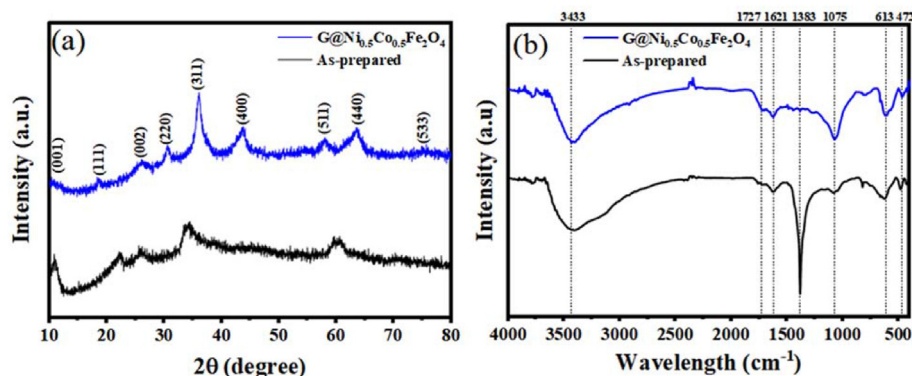


Fig. 1. X-ray diffraction pattern (a), and FT-IR spectra (b) of the as-synthesized sample and G@Ni_{0.5}Co_{0.5}Fe₂O₄ composites.

existence of a little C=O carbonyl group. The vibration band at 1075 cm⁻¹ seen in both materials can be allocated to the stretch vibration of the C–O. Whereas, the vibrating band associated with O–H stretching is evident with a broad peak around 3433 cm⁻¹ [18]. This peak shrank after the calcination process indicating the evaporation of adsorbed surface water during the calcination process. The obvious peak at 613 cm⁻¹ and a shoulder peak at 472 cm⁻¹ are ascribed to the tetrahedral and octahedral Fe–O or Ni–O stretching of the spinel ferrite phase of the sample [18,19]. These analyses confirm the successful synthesis of G@Ni_{0.5}Co_{0.5}Fe₂O₄ composites.

The surface chemical makeup of the elements of the sample was studied by XPS measurements, with results displayed in Fig. 2. The full XPS spectrum presented in Fig. 2a reveals that the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite is fully composed of C, Fe, Ni, Co, and O element with photoelectron peaks at binding energies (BE) of 285.3, 711.5, 855.6, 780.1, and 529.3 eV, respectively. The deconvoluted C 1s peak in Fig. 2b reveals few peaks having BE of 284.6, 285.5, and 288.3 eV that can be ascribed to the C–C, C–OH, and the C=O, respectively [20]. High-resolution Fe 2p peak presented in Fig. 2c reveals two peaks at 710.9 and 724.4 eV relating to the Fe

2p_{3/2} and Fe 2p_{1/2}, respectively. A satellite peak associated with Fe 2p_{3/2} can be seen at BE at 712.9 eV which shows the contribution of Fe³⁺ ions in the octahedral sites. The deconvoluted Ni 2p peak in Fig. 2d reveals a Ni 2p_{1/2} peak at a BE of 888 eV while the Ni 2p_{3/2} spectrum match well with a single peak at 855.5 eV with a characteristic satellite peak at 861.5 eV, which can be attributed to a shake-up type peak of Ni at the high binding energy side of the Ni 2p_{3/2} [21]. The two peaks of BE at 780.6 and 795.4 eV shown in Fig. 2e also correspond to the Co 2p_{3/2} and Co 2p_{1/2}, respectively. All binding energies are referenced: showing the Fe³⁺ & Fe²⁺, Ni³⁺ & Ni²⁺, and Co³⁺ & Co²⁺ states respectively, confirming the spinel ferrite phase of the sample. Whereas, the two peaks at BE of 530 and 531.5 eV seen in Fig. 2f are related to the O²⁻ and O–H group due to the presence of oxygen [18].

Raman spectrum of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite is displayed in Fig. 3. The Raman fingerprint of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite shows the typical intense G-band and D-band peaks of graphene at 1581 cm⁻¹ and 1349 cm⁻¹, respectively [22]. The G-mode arises from the C–C stretching of the graphene sheet, while the D-mode emerges from disordered sp² of the hybridized G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite [23]. Generally, the D-mode intensity

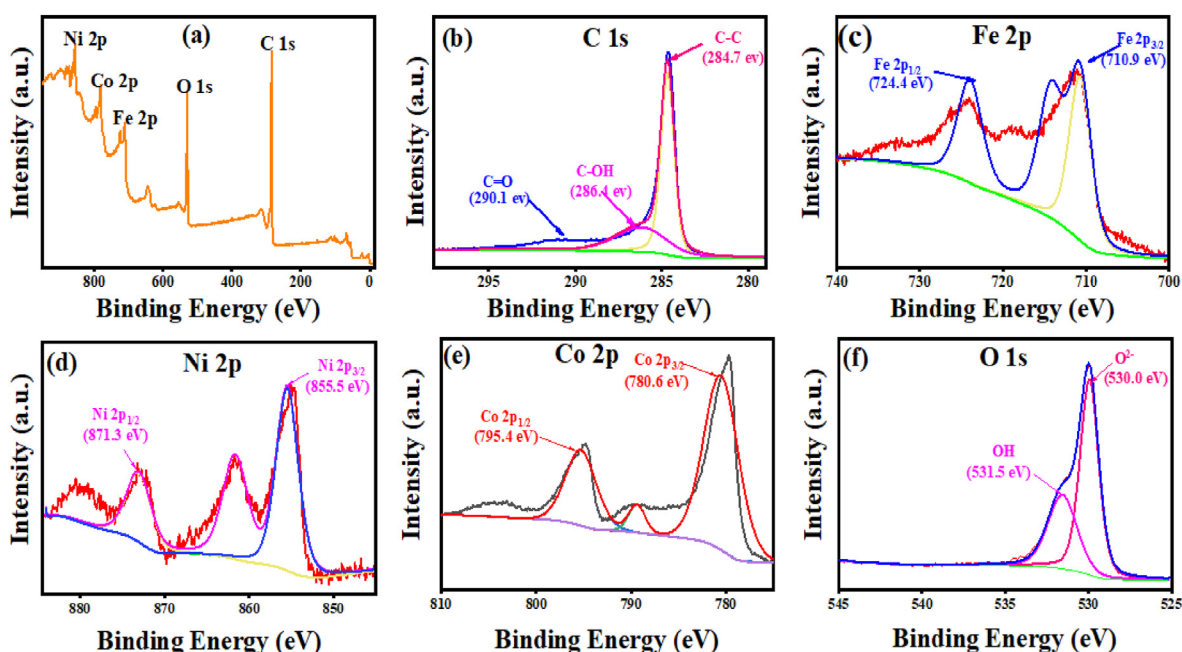


Fig. 2. XPS survey spectrum of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite (a), deconvoluted C 1s (b), Fe 2p spectrum (c), Ni 2p spectrum (d), Co 2p spectrum (e), and O 1s region (f).

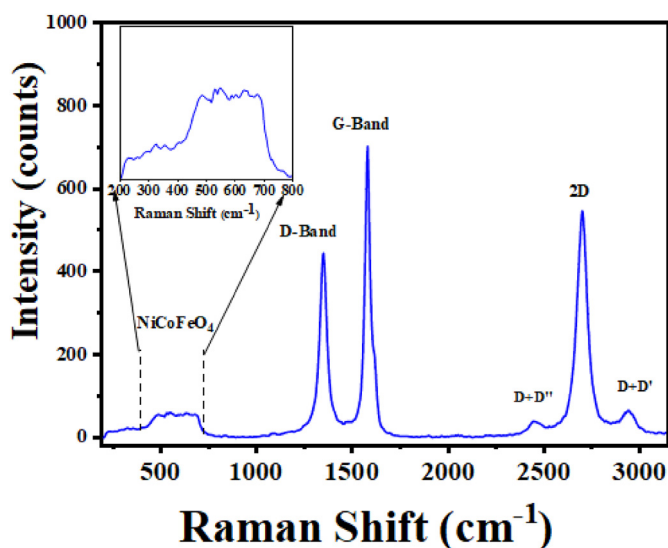


Fig. 3. Raman spectra of G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite, with inset showing the magnified spectrum of Ni_{0.5}Co_{0.5}Fe₂O₄ spinel peaks.

is much smaller than the G-mode. However, the D-mode in this sample is relatively higher (with $I_D/I_G \approx 0.8$) than that of some reported pure graphene owing to the Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticle interactions with the graphene sheets which results in more defects. The appearance of another intense peak at around 2705 cm⁻¹ (2D band) resulting from second-order two-phonon Raman scattering near the Brillouin zone boundary, combined with the G-band peak indicates the graphitic sp² signature. The high symmetry of the 2D-mode clearly distinguishes the graphene sheet of the sample from graphite [23]. The formation of two faint symmetric peaks at 2458 cm⁻¹ (D + D'') and 2947 (D + D') around the 2D-band indicates the formation of sublayers of the graphene sheets [22]. The observed peaks between 200 cm⁻¹ and 690 cm⁻¹ (Fig. 3 inset) can be attributed to metal ions groups corresponding to symmetric and antisymmetric bending in metal-oxygen bonds. These Raman active modes ($A_{1g} + E_g + B_{2g}$) confirms the spinel structure of the Ni_{0.5}Co_{0.5}Fe₂O₄ within the graphene sheets [24].

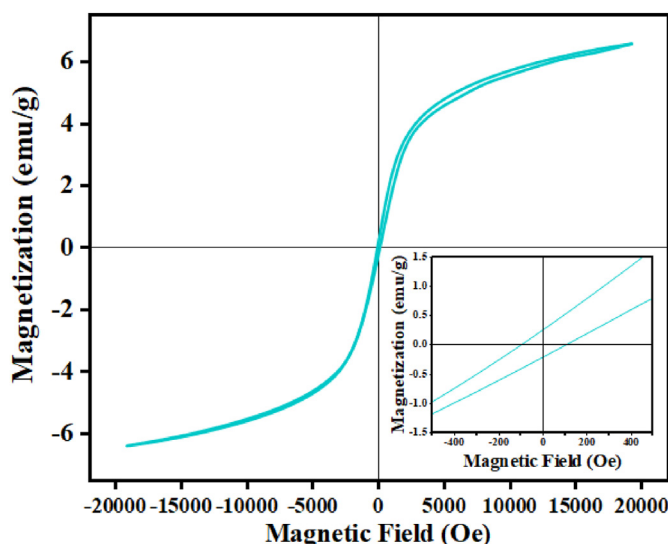


Fig. 4. Hysteresis Loop of G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite.

3.2. Magnetic properties

Fig. 4 shows the room temperature magnetic hysteresis curve of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite. The saturation magnetization value (M_s) of the composite is 6.5 emu/g. The low M_s value can be traced to the superparamagnetic nature (average particle size 9.5 nm) of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticles and the presence of non-magnetic graphene in the composites. The sample possesses a coercivity value of 105.56 Oe reflected in the magnified view of the low magnetic field (inset of Fig. 4). It has been reported that H_c is directly linked to the crystallite size and shape as well as magnetocrystalline anisotropy. Here, the large surface of the graphene flakes produced large surface anisotropic energy that results in the large H_c value [25].

3.3. Morphology analysis

The morphology of the as-received graphene as investigated by FESEM is presented in Fig. 5a which clearly shows the formation of layers of the graphene nanoflakes. The FESEM image in Fig. 5b–d shows the morphology of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite. High magnification of the FESEM image in Fig. 5c and d reveal that the Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticles are well distributed and anchored within the graphene layers with the existence of the interconnected porous network. FTIR and XPS analysis suggest that C–C, C=O and C–OH play part in the interconnect porous network. The porous structure would ensure that G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite has low density, indicating its potential as a lightweight absorber. Another benefit resulting from this porous structure is the multiple transmission routes for the transmission and dissipation of the EM wave [26]. Whereas, the high distribution of the Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticles on the graphene flakes would yield an abundance interface for interfacial polarization that would increase the dielectric loss of the microwave. The EDS spectrum (Fig. 5e) confirms the purity of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite indicated by the non-existence of other peaks in the spectrum. The elements identified in the spectrum show consistency with the XPS results revealing large percent of carbon (C) and oxygen (O), with lesser iron (Fe), cobalt (Co), and nickel (Ni) percent, which relates to the G@Ni_{0.5}Co_{0.5}Fe₂O₄ stoichiometric ratio. The EDS elemental mapping showing the homogenous distribution of elements in the sample is presented in Fig. S1.

The structure of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite was further examined by TEM. As displayed in Fig. 6a, a connected porous structure decorated with homogenous distribution of Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticles was detected, consistent with the FESEM analysis. It is worthy to note that Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticles remain tightly anchored to the graphene flakes (Fig. 6b) without detachment and agglomeration despite hours of sonication in preparing the sample for the TEM analysis. High-resolution TEM image (HR-TEM) displayed in Fig. 6c presents the crystallinity of the composite. The interplanar lattice fringes 0.41 nm and 0.24 nm correspond to the (002) and (113) planes of graphene. Whereas, an interplanar lattice fringe of 0.26 nm corresponds to the (311) plane of typical spinel showing consistency with our XRD results. The selected area diffraction (SAED) pattern (Fig. 6c inset) presents a diffraction ring of a poly-nano-crystalline structure typical of the ferrite phase. These analyses confirm the successful synthesis and structure of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite.

To analyze the porous nature of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite, N₂ adsorption-desorption investigation was implemented. As indicated in Fig. 7, the sample displayed a characteristic type-IV isotherm having a clear hysteresis loop with relative pressure ranging from 0.14 to 1.0, suggesting the presence of mesopores in the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite. Using the BET calculation

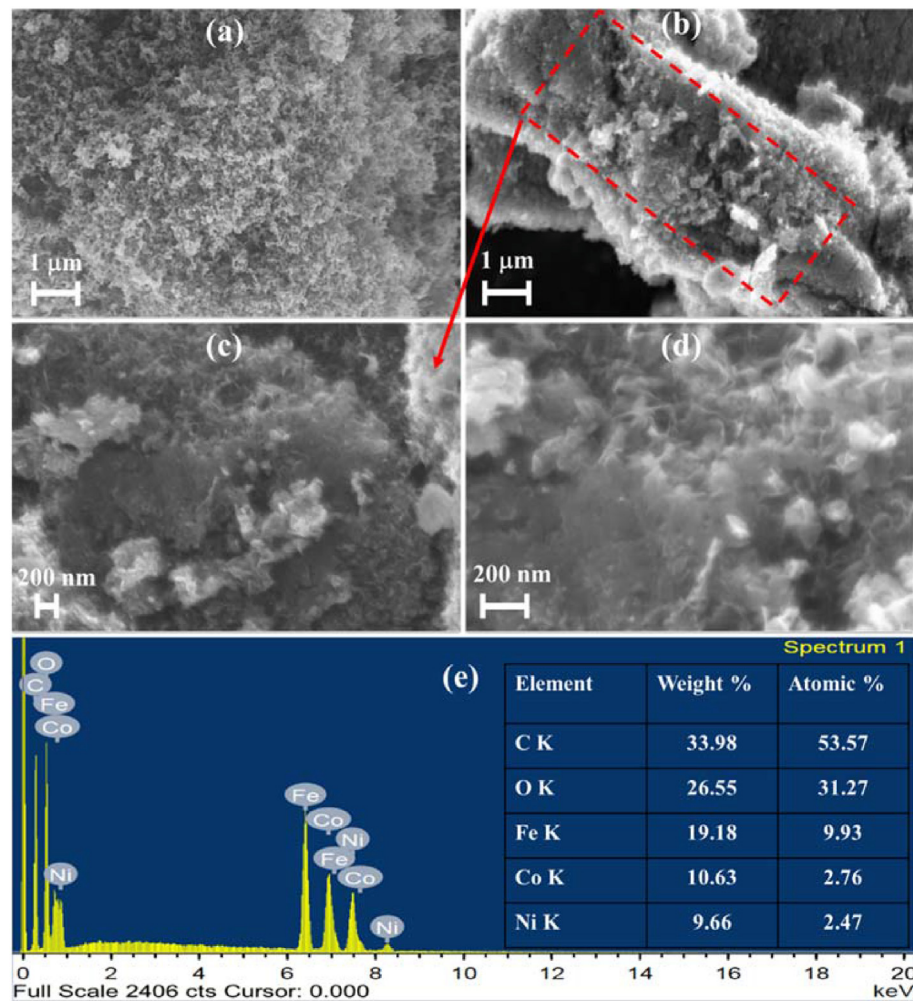


Fig. 5. FESEM images of pure graphene nanoflakes (a), the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite (b), higher magnification images of a selected area (c, d), and EDX spectrum of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite (e).

method, the sample exhibits a high specific surface area ($155.6 \text{ m}^2\text{g}^{-1}$), with a pore volume of $0.32 \text{ cm}^3\text{g}^{-1}$. The average pore size displayed in the inset (Fig. 7) is calculated as 7.9 nm. It is expected that the existence of Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticles on the graphene sheets will increase the composite's overall polarization. Because the presence of inhomogeneous materials at pore boundaries (i.e. graphene - Ni_{0.5}Co_{0.5}Fe₂O₄ in this case) produces inhomogeneous EM field distribution throughout the composite structure [27]. It is expected that this porosity would reduce the density of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite and favor the absorption of EM wave by providing several interfaces for scattering and rapid attenuation of EM wave [28].

3.4. Electromagnetic waves absorption

EM wave absorption widely represented by the RL of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite filled in paraffin was calculated based on the transmission line theory with the equations

$$RL(\text{dB}) = 20 \log_{10} \left| \frac{Z_{in} - Z_0}{Z_{in} + Z_0} \right| \quad (2)$$

$$Z_{in} = Z_0 \sqrt{\frac{\epsilon_r}{\mu_r}} \tanh \left(j \frac{2\pi f d}{c} \sqrt{\epsilon_r \mu_r} \right) \quad (3)$$

where f represents microwave frequency, c is the free space velocity, the ϵ_r denotes the relative permittivity, d is the absorber thickness, the μ_r is relative permeability, Z_{in} is the absorber characteristic impedance, and Z_0 is the impedance of air. It is widely accepted that RL values of -10 dB correspond to 90% absorption of the incident waves [29]. Thus, the range of frequency where $RL < -10 \text{ dB}$ is deemed as effective absorption bandwidth. With absorber thickness in the range of 1.0–5.0 mm, three-dimensional representations of the frequency-dependent RL curves of the GNC@5, GNC@10 and GNC@15 are displayed in Fig. 8a–c. The optimum RL value achieved by GNC@5 is -46 dB at a 17.65 GHz with a 3.0 mm thickness. Also, Fig. 8b shows that the optimal RL of GNC@10 reached -55.2 dB at 15.25 GHz when the matching thickness was 2.5 mm whereas, the optimal absorption achieved by GNC@15 was achieved at a thin thickness of 1.5 mm with a minimum RL value of -44.7 dB at 17.398 dB. It is observed that the absorption efficiency improved with an increase in the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite loading. Nonetheless, all composites display good EM wave absorption performance.

It has been established that EM wave absorption properties of

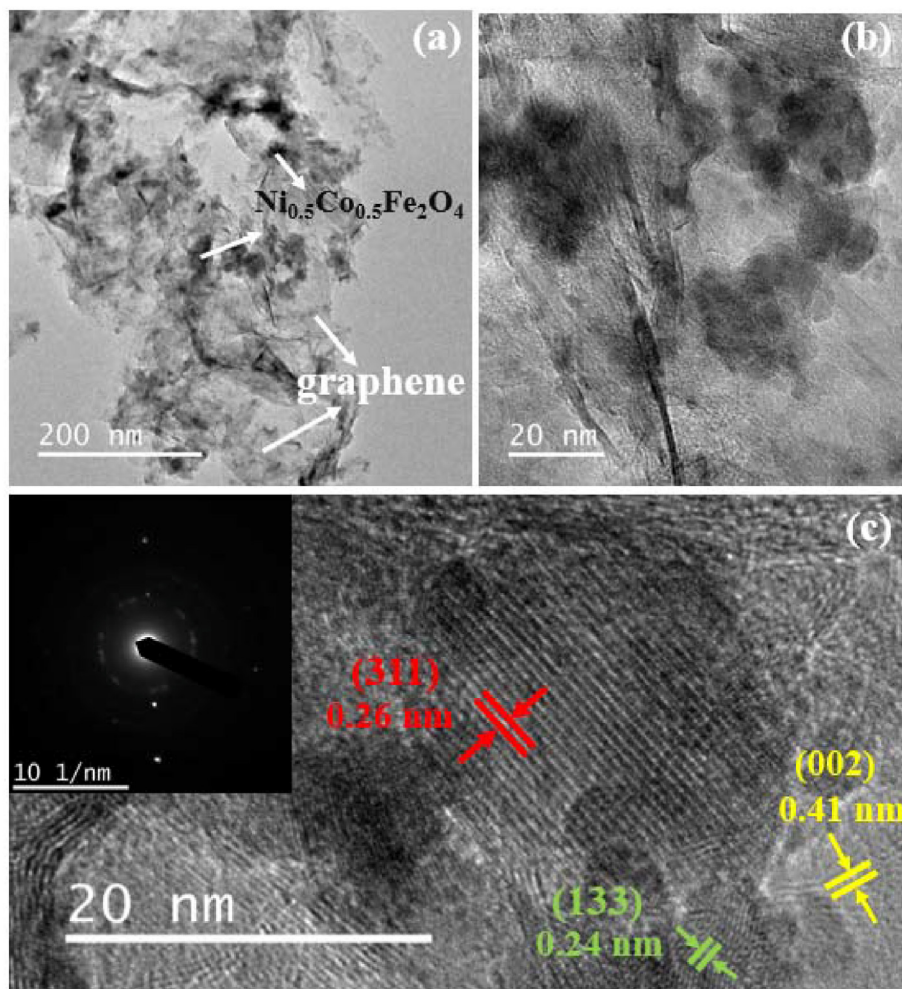


Fig. 6. TEM image G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite (a, b) and HR-TEM image of the nanoparticle, inset is the SAED pattern (c).

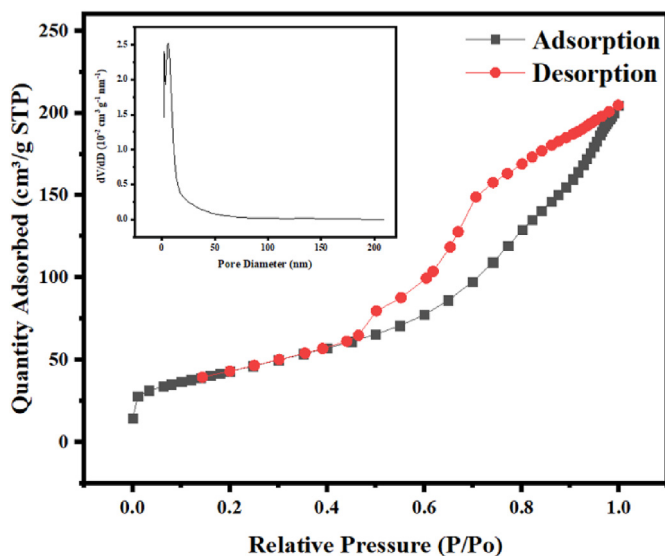


Fig. 7. Nitrogen adsorption/desorption isotherm of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite. Inset shows BJH pore size distribution.

absorbing materials are related to thickness. The RL curve of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite in paraffin with 1.0–5.0 mm absorber thickness is presented in Fig. 9a–c. For all the samples, the effective absorption bandwidth can cover all the X-band and Ku-band frequency by adjusting the thickness from 1.5 to 5.0 mm. The EM wave absorption performance of G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite shows good compliance to the quarter-wavelength rule as the optimal absorption frequency drift towards the lower frequency with increasing thickness [30]. This behavior can be well demonstrated with the quarter wavelength model matching equation defined by equation (4).

$$t_m = n\lambda / 4 = nc / (4f_m \sqrt{|\mu_r||\epsilon_r|}) \quad (4)$$

It suggests that the absorber thickness, t_m varies inversely with the matching frequency, f_m , where $n = 1, 3, 5, 7 \dots$, λ is the EM wavelength, c is the speed of light, μ_r and ϵ_r are the relative permeability and permittivity, respectively. This analysis is presented in Fig. 9 shows the simulated curve of the optimal thickness versus the matching frequency. The blue ovals in this figure are the matching thicknesses equivalent to the optimal absorption frequency in the RL curves. The blue ovals appear on the simulation curve, indicating that the MAP of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite conforms with this model. Aside inherent magnetic and dielectric loss ability of absorbers, the basic prerequisite to achieve EM

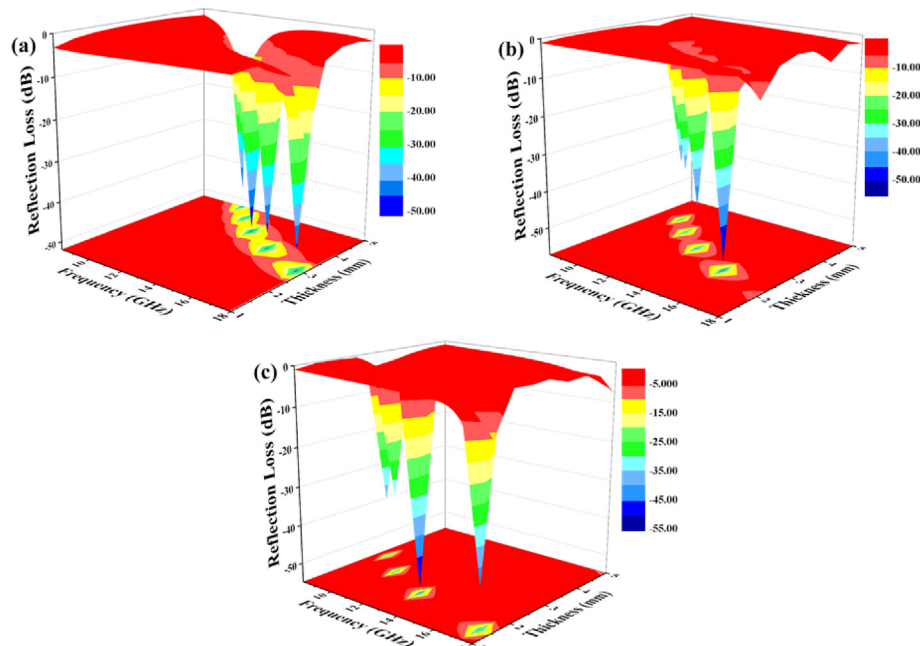


Fig. 8. Three-dimensional representation of RL curves of GNC@5 (a), GNC@10 (b), and GNC@15 (c).

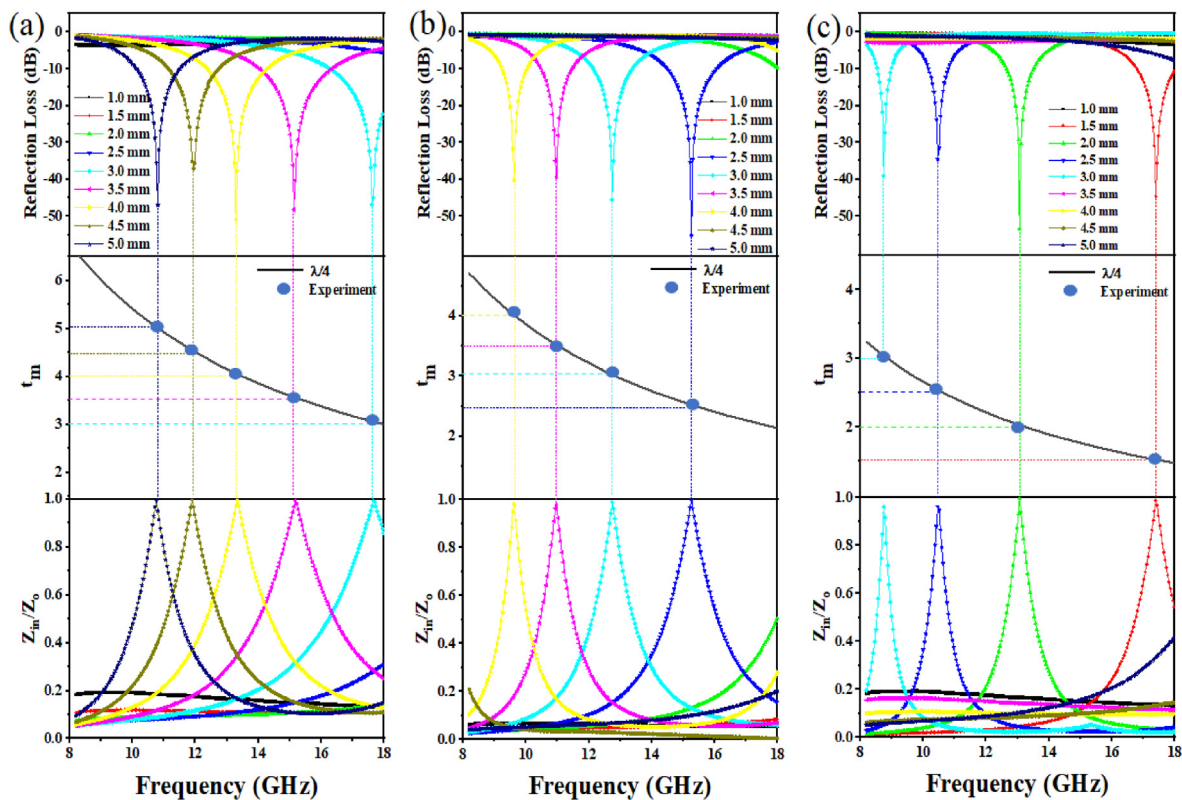


Fig. 9. Frequency-dependent reflection loss, absorber thickness(t_m) against peak frequency (f_m) simulation, and the relationship between Z_{in}/Z_o and microwave frequency of GNC@5 (a), GNC@10 (b) and (c) GNC@15

absorption is an optimal match of impedance. The impedance match is optimal when the $|Z_{in}/Z_o|$ is close or equal to unity [31]. The $|Z_{in}/Z_o|$ values for the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite were analyzed and understood to have values near to 1 at the matching thicknesses and frequencies indicating optimal impedance

matching. Such good impedance matching confirms that a large portion of the EM wave transmits the material interface and results in enhanced EM wave absorption. A thorough comparison between the absorption performance of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite and other earlier reported graphene-magnetic based absorbers is

Table 1
EM waves absorption of related graphene-based absorbers.

Absorbers	wt (%)	RL (dB)	Thickness (mm)	Bandwidth (GHz) (RL < -10 dB)	Ref.
Fe ₃ O ₄ /SiO ₂ /graphene	50	-27.1	1.5	2.5	[48]
Hollow Fe ₃ O ₄ @RGO	50	-41.89	2.5	4.2	[45]
NiFe ₂ O ₄ @MnO ₂ @graphene	30	-47.4	3.0	4.3	[46]
Graphene/Fe	40	-45.0	3.0	4.4	[49]
RGO/NiFe ₂ O ₄	50	-39.7	3.0	5.0	[47]
rGO/ZnO	50	-45.1	2.2	3.3	[50]
graphene@Ni@C	30	-34.2	1.6	3.2	[50]
Graphene/PANI	25	-38.8	3.0	2.3	[51]
Graphene nanosheets/MgO	—	-36.5	1.5	2.0	[6]
G@Ni _{0.5} Co _{0.5} Fe ₂ O ₄	10	-55.2	2.5	1.8	This work
G@Ni _{0.5} Co _{0.5} Fe ₂ O ₄	15	-44.7	1.5	2.8	This work

presented in Table 1. In comparison with graphene-magnetic based absorbers in literature, the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite reported here offers several benefits, like lightweight, strong absorption, and good absorption bandwidth at a minimal absorber thickness.

To ascertain the absorption mechanism, the measured EM parameters of the sample were analyzed. Measured EM parameters include the real and imaginary permittivity ($\epsilon_r = \epsilon' - j\epsilon''$), and also real and imaginary permeability ($\mu_r = \mu' - j\mu''$). The EM parameters

measured at the X-band and Ku-band frequency bands is shown in Fig. 10. The ϵ' values that signify the electric energy storage of the GNC@5, GNC@10, and GNC@15 are in the range of 2.93–6.44, 2.45–9.53, and 2.53–13.22, respectively. Whereas the ϵ'' values representing dissipation of electric energy of these composites are in the range of 1.23–2.73, 0.80–5.11, and 0.26–6.71, respectively. As seen in Fig. 10a and b, the ϵ' and ϵ'' of all composites exhibit the same trend showing a swift decrease in magnitude with an increase in

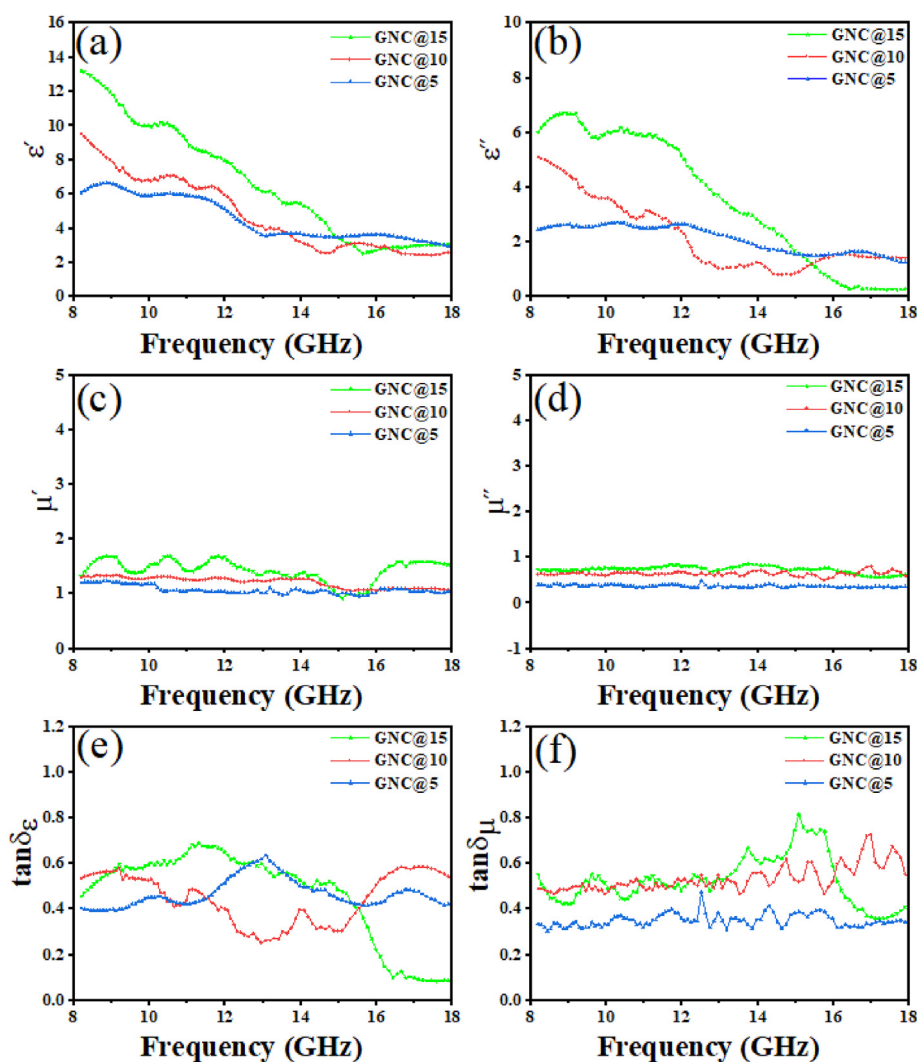


Fig. 10. Real and imaginary permittivity (a, b), real and imaginary permeability (c, d), and dielectric and magnetic loss tangents (e, f).

frequency. This behavior has been reported to be associated with the inability of dipoles to respond quickly to fast alternating frequency [32]. This behavior is also related to a decrease in space charge polarization at the interface of the materials [1,33]. Further, the permittivity curve reveals a rise in the dielectric loss with increase loading of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite which is aligned with the effective medium theory [34]. The measured real and imaginary permeability is presented in Fig. 10c and d. Similarly, the μ' and μ'' characterize the magnetic energy storage and dissipative ability, respectively. As presented in Fig. 10c, the μ' and μ'' values of all the composites have a range of 0.92–1.69, and 0.33–0.86, respectively. These indicate that the samples have good magnetic loss abilities. Both μ' and μ'' var with some resonances at high frequencies. This behavior can be ascribed to the exchange resonance or natural ferromagnetic resonance of the spinel phase and is prominent in the GNC@15, further justifying its superior EM wave absorption ability. These fluctuations and/or variations in the EM parameters observed at high frequency can also be due to interfacial polarization and electron relaxation [35]. To understand the dominant loss mechanism, dielectric tangent loss ($\tan\delta_e$), and the magnetic counterpart ($\tan\delta_\mu$) of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite are displayed in Fig. 10e and f, respectively. The $\tan\delta_e$ values of the composites range within 0.08–0.64 and that of $\tan\delta_\mu$ values range from 0.30 to 0.81 in the X-band and Ku-band frequency bands. Notably, the $\tan\delta_e$ values are higher than the $\tan\delta_\mu$ values at X-band (8.2–12.4 GHz) frequency range, suggesting that dielectric loss is the dominant loss mechanism at this range of frequency. Whereas, the $\tan\delta_\mu$ values can be seen higher than the $\tan\delta_e$ values at Ku-band frequency, indicating a large contribution of magnetic loss to the EM wave attenuation at this frequency band.

Further analysis of the loss mechanism is relevant and presented. It has been established that dielectric losses often originate from the relaxation process and various dielectric polarization such as ionic, electric, dipolar, and interfacial polarization [31]. Previous studies have indicated that electron and ionic polarization occur at quite a higher frequency (10^3 – 10^6 GHz), and thus can be excluded [7]. The Debye relaxation model was used to investigate the resonance behavior seen in the $\tan\delta_e$ curve and ascertain the specific dielectric loss mechanism. This model explains a behavior between ϵ' and ϵ'' and can be defined as:

$$(\epsilon' - \epsilon_\infty)^2 + (\epsilon'')^2 = (\epsilon_s - \epsilon_\infty)^2 \quad (5)$$

in which where $\omega = 2\pi f$, denotes angular frequency, τ is relaxation time, ϵ_s and ϵ_∞ are the static permittivity and the high-frequency relative permittivity. According to the model, a polarization process is confirmed by the appearance of a semi-circle in a Cole-Cole plot (plot of ϵ'' values versus ϵ' values) [36]. As illustrated in Fig. 11a, the Cole-Cole plots of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite exhibited some semi-circles. The semi-circles in the Cole-Cole plots show few distortions suggesting another polarization such as conduction loss contribution to the dielectric loss [37,38]. On second thought, this behavior may be due to the good porous structure of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite and the small crystallite size of the crystallite Ni_{0.5}Co_{0.5}Fe₂O₄ anchored on the graphene sheets. Besides, the existence of functional groups established by the XPS and FT-IR analysis and possible defects in the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite can act as polarization for improved dielectric loss mechanism [39]. The abundant mixed interfaces that exist within the widely distributed Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticles and graphene sheets, as well as the interface between G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite and paraffin matrix, can yield active sites for multi-interface polarization and promote multiple polarization loss mechanisms [40,41].

Likewise, the contribution of magnetic loss ($\tan\delta_\mu$), resulting

from different processes such as natural, domain wall and eddy current resonance, and hysteresis loss [42,43] is worth investigating. It is reported that domain wall resonance has an effect at much low low-frequency (1–100 MHz), and hysteresis loss has an insignificant effect in weak magnetization cases and thus can be excluded. Hence, eddy current loss and ferromagnetic resonance could be accountable for the magnetic losses contributed to the EM wave absorption. Investigation of the magnetic loss impact of eddy current is carried out by evaluating the dependence of $C_0 = \mu''(\mu')^{-2}f^{-1}$ variation with changes in frequency. In the case where C_0 is constant with increasing frequency, this confirms the impact of eddy current on the magnetic loss [44]. As revealed by Fig. 11b, the C_0 values for all the samples tend to be constant throughout X-band frequency, and widely vary in the Ku-band range. This indicate that eddy current contributes to magnetic loss at X-band frequency, while the magnetic loss contribution to EM wave absorption at Ku-band frequency can be ascribed to ferromagnetic resonance. Also, the several resonance peaks seen in the $\tan\delta_\mu$ curve (Fig. 11f) also suggest the contribution of ferromagnetic resonance to the magnetic loss. It is acknowledged that the capability of an absorptive material to dissipate EM wave is measurable by calculating its attenuation constant (α), which is a combination of the contribution from both loss mechanisms.

$$\alpha = \frac{\sqrt{2}\pi f}{C} \times \sqrt{(\mu''\epsilon'' - \mu'\epsilon') + \sqrt{(\mu''\epsilon'' - \mu'\epsilon')^2 + (\mu'\epsilon'' + \mu''\epsilon')^2}} \quad (6)$$

in which f is the propagating frequency, and C is the velocity of microwave in space. Presented in Fig. 11c, The GNC@15 has the best attenuation amongst the samples. Nevertheless, both GNC@5 and GNC@5 also have reasonably high EM wave attenuation capacity compared to other graphene-based absorbers earlier reported [45–47]. Therefore, we can infer that the efficient absorption of EM wave occurs due to moderate attenuation combined with a good match of impedance.

Based on the above findings, the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite benefits from the contribution of a few loss mechanisms which resulted in the satisfying EM wave absorption efficiency. Fig. 12a shows the schematic illustration of the propagating EM wave and dissipation processes in the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite. First, good impedance matching was achieved from the combination of magnetic loss of Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticles and dielectric loss contribution from graphene flaxes. This desired match of impedance would favor little or no reflection at the material surface and the composites dissipates the trapped waves by its attenuation power. The uniformly dispersed Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticles improve the impedance matching of the graphene sheet, which reduces the reflection of EM wave. Moreover, the preparation of the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite with little agglomeration of Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticles provide abundant interface for propagation and dissipation of EM wave. The small nanoparticles dimension of the Ni_{0.5}Co_{0.5}Fe₂O₄ would increase dipole polarization and enhance dielectric loss for optimum EM wave dissipation. The large surface area resulting from the graphene can also yield abundant active sites for polarization and enhancing the EM wave absorption. The graphene layers with high carbon content also provide a high dielectric loss mechanism and improved attenuation of the trapped EM wave via dissipating the EM wave loss into heat. While the residual groups and structural defects on the graphene sheets can lead to the formation of dipoles, which would contribute to the conversion of EM energy into heat energies through the polarization relaxation loss. The exceptional porous network and empty spaces porous also pave extra paths for the spreading of EM wave

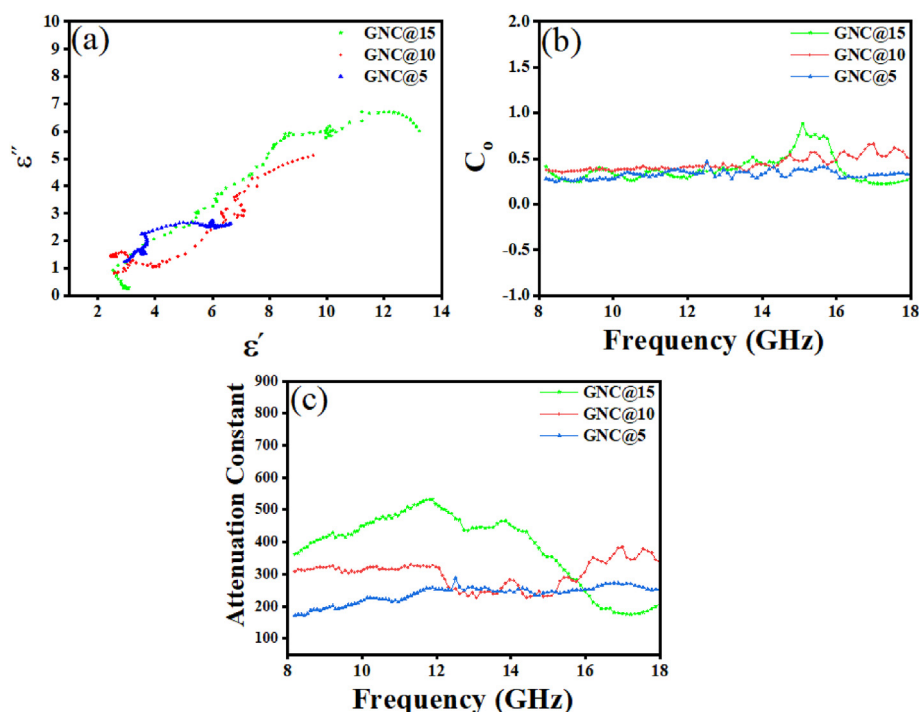


Fig. 11. Cole-Cole plot (a), Co values (b), and attenuation constant of all samples (c).

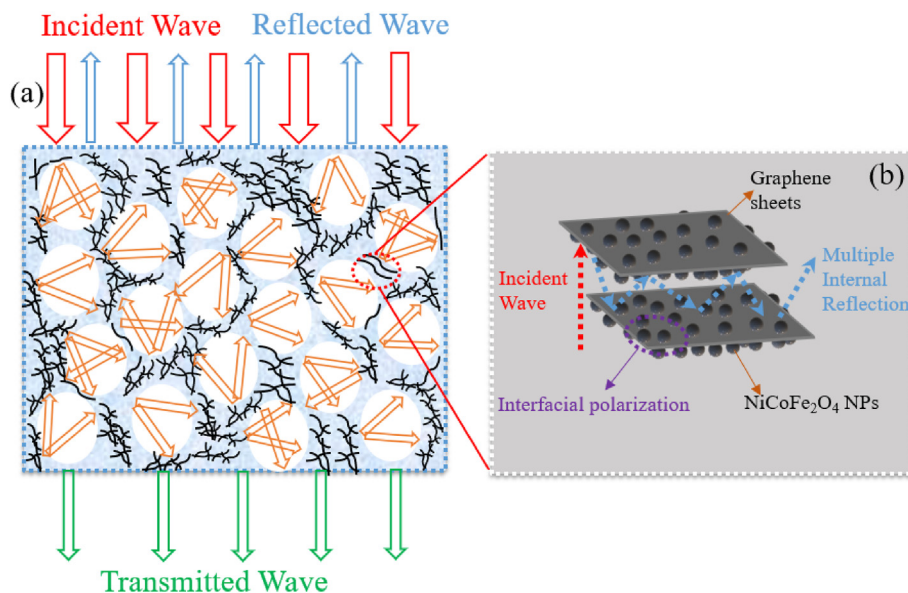


Fig. 12. Schematic illustration of propagation and attenuation of EM waves in the G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite (a), and multiple internal reflection illustration (b).

within the absorber resulting in dissipation of EM wave by multiple reflections and scattering mechanism Fig. 12b. As a heterogeneous absorber, interfacial polarization arising from the several interfaces between Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticles and graphene sheets is another possible loss mechanism, which contributes to the enhanced EM wave absorption.

4. Conclusion

In summary, a lightweight and porous G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite was prepared using a facile coprecipitation technique,

followed by a high-temperature calcination process. The physicochemical properties of the sample were investigated by FESEM, TEM, XRD, and XPS. Results show that G@Ni_{0.5}Co_{0.5}Fe₂O₄ composite has good morphology, composition, and structure that favor the absorption of EM wave. Our investigation of the EM wave absorption performance indicates that GNC@15 has excellent EM wave absorption properties. A minimum RL of −44.7 dB at 17.45 GHz, with 1.5 mm absorber thickness was achieved. This enhanced performance could be ascribed to interfacial polarization, the unique porous structure, void spaces between the magnetic Ni_{0.5}Co_{0.5}Fe₂O₄ nanoparticles and graphene, and good impedance

matching. Hence, we present that $G@Ni_{0.5}Co_{0.5}Fe_2O_4$ composite can function as EM wave absorbing materials at a minimum thickness and low density.

CRediT authorship contribution statement

Fatai Adisa Wahaab: Conceptualization, Writing - original draft, Methodology, Investigation. **Wasiu Yahya:** Writing - review & editing, Validation. **Lawal Lanre Adebayo:** Writing - original draft, Methodology. **Issa Kazeem:** Visualization, Validation. **Abdulganiyu Abdulraheem:** Writing - review & editing. **Bilal Alqasem:** Formal analysis, Validation. **Jemilat Yetunde Yusuf:** Writing - review & editing, Resources. **Abibat Asabi Adekoya:** Writing - review & editing, Software. **Chai Mui Nyuk:** Formal analysis, Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jallcom.2020.157259>.

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