

# The influence of RGO gain on Co 0.5 Mn 0.3 Cu 0.2 Fe 2 O 4 nano-composites' construction, morphology, and magnetic characteristics

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## Research Article

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# Abstract

One-pot simplified co-precipitation procedure was used to produce non-stoichiometric ferrite ( $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$ ) and diminish graphene oxide (rGO) nano-composites in this investigation. Powders such as XRD, FT-IR, and SEM were used to study the structural and surface analysis of the composites. An investigation of the magnetic characteristics of  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  nanocomposites (NCs) was also conducted using VSM. A vector network analysis tool was used to conduct microwave adsorption investigations on  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  NCs in the 2–18 GHz frequency spectrum. By adjusting the amount of rGO added to  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$ , the electromagnetic (EM) wave absorbing characteristics of  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  nanocomposites may be remarkably adaptable due to their customizable microstructure and synergistic effects. The addition of rGO can significantly improve EM wave absorption characteristics compared to the unadulterated  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  ferrites. The generated  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  NCs' minor reflection loss (RL) was 39.7 dB in the 11.9 GHz frequency spectrum. The effective absorption bandwidth is a maximum of 6.5 GHz (RL under 10 dB) with a thickness of two millimeters. The  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  NCs that haven't been changed since synthesis (as-synthesized) might be considered a novel alternative for the slight EM wave absorber due to their proper impedance matching and synergistic effect **among** dielectric and magnetic loss.

## 1. Introduction

Over the last few years, electromagnetic (EM) wave interference has become a significant environmental pollution issue that can endanger people's health and the regular operation of electrical devices due to the widespread use of wireless communication gadgets, local area networks, and other communication supplies [1–3]. Such issues have effects not just on the regular operation of specific devices but also on people's health. To eliminate the damage caused by electromagnetic (EM) interference, significant work has been carried into manufacturing materials that can successfully absorb EM waves and convert EM energy into heat or microwave energy, which is employed to reduce interference damage [4–7]. Electrical science devices, military armaments, and other supplies are commonly utilized materials of this type, furthermore commercially accessible. Yet, developing and synthesizing EM wave absorbers with slight and narrow thickness, wide absorption bandwidth, and considerable absorption over a broad frequency spectrum remains a challenge [8–11]. Also, numerous prior studies have shown that traditional single-phase EM wave absorption materials, including ferrites, conductive polymers, ceramics, and metal powders, cannot match the aforementioned functional criteria [12, 13].

The production and investigation of spinel nano ferrite ( $\text{M}^{2+}\text{Fe}_2\text{O}_4$ ,  $\text{M}^{2+} = \text{Fe, Co, Mn, Cu}$ ) are more attractive among the many magnetic materials because their hard/soft magnetic nature is influenced by the form of divalent cation ( $\text{M}^{2+}$ ) existing in nano ferrites [14–16].  $\text{CoFe}_2\text{O}_4$  stands out for its outstanding chemical stability, saturation magnetization ( $M_s$ ), considerable coercivity, significant magneto crystalline anisotropy (MCA), tremendous magneto strictive coefficient, and mechanical hardness, among others [17].

Due to their high-density magnetic monitoring, these features make  $\text{CoFe}_2\text{O}_4$  nanoparticles an appealing candidate for many magnetic necessities, like cameras, sensors, and electric motors [18]. Due to their medium saturation magnetization, outstanding chemical stability, low real part permittivity, and substantial magnetic loss, spinel ferrites ( $\text{M Fe}_2\text{O}_4$ ,  $\text{M} = \text{Fe, Co, Ni, Mn, Zn, etc.}$ ) are supposed as the proper candidates for microwave absorbing materials. Due to the single spinel ferrite microwave absorber's poor ability to attenuate, high density, quick aggregation, and tremendous covering thickness, getting into down-to-earth applications are genuinely challenging in any case. Then, the design of ferrite-based nanocomposites is of significant consideration for improving their microwave absorption efficiency [19]. In microwave absorption, carbon-based materials hold a wide variety of usages. Because of their remarkable physical and chemical characteristics, graphene, carbon nanotubes, graphite, and other carbon allotropic isomers are consequential in investigating microwave-absorbing materials [24–27]. Reduced graphene oxide (rGO) is a carbon-based substance employed in microwave absorbing materials as the ultralight one. To achieve lightweight absorbing materials, it's a practical way. The excellent carrier migration rate and dielectric constant of rGO distinguish it from other carbon-based materials. rGO is ultrathin, has a high optical transfer, and is stable at room temperature. Consequently, it has poor quality when used as a wave absorbing material, while it can lessen the skin effect impressively. Also, due to its specific surface area and fold form, rGO has a considerable resonance and interface polarization, advantageous for microwave absorbing and attenuating [28–32].

According to the supplementary effects on dielectric and magnetic characteristics, graphene/ferrite nanocomposites have gained a lot of interest in the latest years, especially given the broadband and lightweight expectations of absorbers. A  $\text{BaFe}_{12}\text{O}_{19}@\text{Fe}_3\text{O}_4$  core-shell composite with a minor reflection loss (RL) of -33.6 dB at 11.6 GHz and 2.5 millimeters thick was synthesized by Lin et al. [19]. The lowest RL value of 29.7 decibel was reported by Bach et al. for a  $\text{La}_{1.5}\text{Sr}_{0.5}\text{NiO}_4/\text{NiFe}_2\text{O}_4$  nanocomposites [20]. The min RL reached 58.4 decibels at 13.68 GHz with 2.1 millimeters thick for  $\text{Fe}_3\text{O}_4/\text{NiFe}_2\text{O}_4/\text{Ni}$  heterostructure porous rods, synthesized by Li et al. in a controllable way [21].

According to the supplementary effects on dielectric and magnetic characteristics, graphene/ferrite nanocomposites have gained a lot of interest in the latest years, especially given the broadband and lightweight expectations of absorbers. Meanwhile, numerous graphene/ferrite NCs have been generated using diverse techniques such as hierarchical  $\text{CoFe}_2\text{O}_4/\text{rGO}$  NCs with a porous structure which were synthesized employing an in-situ solvothermal technique and demonstrated a remarkable RL of -57.7 decibels with 2.8 millimeters thick [22].  $\text{Fe}_3\text{O}_4/\text{rGO}$  hybrids holding the minimum RL of 22.7 decibels and a 3.13 GHz effective absorption bandwidth (90 percent absorption) were stated by Wu et al. [23]. By focusing on this perspective, nonstoichiometric  $\text{Co}_{0.8}\text{Fe}_{2.2}\text{O}_4$  ferrite is chosen for this research to incorporate with rGO to produce  $\text{Co}_{0.8}\text{Fe}_{2.2}\text{O}_4/\text{rGO}$  NCs. The transmission line theory is used to assess the EM wave absorption characteristics of the recently prepared specimens. The microwave absorbing function of the composite materials is appropriately adjusted by modifying the rGO amount. In addition, the related EM wave absorbing mechanism of  $\text{Co}_{0.8}\text{Fe}_{2.2}\text{O}_4/\text{rGO}$  NCs is investigated.

## 2. Experimental

### 2.1. substances

LOBA in India formulated the natural graphite powder available in the market. Merck in Germany supplied  $\text{H}_2\text{O}_2$  (30 weight percent),  $\text{H}_2\text{SO}_4$  (99 weight percent),  $\text{KMnO}_4$ ,  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ,  $\text{Cu}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  and  $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ , polyethylene glycol (PEG), as well as sodium acetate.

### 2.2. Graphene oxide's synthesis

The entire procedure for producing graphene oxide (GO) employing the modified Hummer's (MHM) technique brings as follows [33]: 4 grams of natural graphite were diffused thoroughly in 360 milliliters of  $\text{H}_2\text{SO}_4$ , 40 milliliters of  $\text{H}_3\text{PO}_4$ , and 18 grams of  $\text{KMnO}_4$  were gently added. The combination was then dried at 60 degrees Celsius and shook for 12 hours. After chilling at room temperature, 200 milliliters of deionized water and 30%  $\text{H}_2\text{O}_2$  solution were combined and shook till the solution acquired a brilliant yellow hue. The combination mentioned above was then centrifuged and thoroughly rinsed with deionized water, 30% HCl, and  $\text{C}_2\text{H}_5\text{OH}$  (ethanol) until the pH was approximately 6.

### 2.3. $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$ NPs' preparation

In DI water, stoichiometric proportions of cobalt nitrate ( $\text{Co}(\text{NO}_3)_2$ ), ferric nitrate ( $\text{Fe}(\text{NO}_3)_3$ ), and also copper nitrate ( $\text{Cu}(\text{NO}_3)_2$ ) are combined with ammonium hydroxide ( $\text{NH}_4\text{OH}$ ) and citric acid ( $\text{C}_6\text{H}_8\text{O}_7$ ). To achieve the transparent product, citric acid is utilized. Diverse doping percentages of copper nitrate employing the co-precipitation technique were controlled by the ammonium hydroxide synthesis. The precipitated components are filtered and then rinsed five times with distilled water. The ultimate precipitate material is dried for 8 hours at 120 degrees Celsius. The dried material is pulverized into a powder and set to sinter for 3 hours at 700 degrees Celsius. Ultimately, employing this method, the Cu doped Ba ferrite combination ( $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$ ) is acquired.

### 2.4. $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$ nanocomposites" manufacturing

The as-synthesized  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  NPs were diffused in GO colloid solution employing a common approach. The ultra-sonication bath was then used to stir the diffused  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  solution for 4 hours. After that, the specimen was dried at  $80^\circ\text{C}$  for 12 hours to generate  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  composites. Ultimately, by heating them to  $400^\circ\text{C}$  for 3 hours, the Reduction process was applied to dried GO/  $\text{Co}_{0.5}\text{Zr}_{0.5}\text{Fe}_2\text{O}_4$  fabrics to obtain  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  nanocomposites.

### 2.5. measurements and attributes

X-ray diffraction (X-ray Powder Diffraction, Bruker D 8 -Advance) with Cu K- $\alpha$  radiation ( $\lambda = 0.154$  millimeters) was used to investigate as-synthesized  $\text{Co}_{0.8}\text{Fe}_{2.2}\text{O}_4/\text{rGO}$  composites' crystallinity and

crystal structure. Nicolet NEXUS 670 Infrared Spectrometer analyzed FT-IR spectra between 400 to 4000  $\text{cm}^{-1}$ . A scanning electron microscope (Flex SEM-1000) at 10 kilovolts was used to examine the microscopic morphology. An Agilent E8362B vector network analysis tool was used to test the EM variables (complex permittivity and complicated permeability) in the 2–18 GHz spectrum.

### 3. Discussion And Results

The XRD schemas of GO,  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$ , and  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  composites are shown in Fig. 1. The diffraction apexes from X-ray Powder Diffraction schemas of  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  and  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  at 2-theta = 18.5, 30.2, 35.6, 43.0, 53.5, 57.1, and 62.8 are consistent with the (111), (220), (311), (400), (422), (511), and (440) crystal surfaces of  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  (JCPDS numbers from 22 to 1086). This discovery demonstrates the cubic spinel form of  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  in the composite. The sharp apexes suggest that the manufactured  $\text{Co}_{0.8}\text{Fe}_{2.2}\text{O}_4$  NPs have excellent crystallinity [34, 35]. The intensity of the diffraction apexes for the  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  specimens appears to diminish compared to the  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  samples considerably, implying that the GO gain restrains the crystallinity of  $\text{CuFe}_2\text{O}_4$  particles partially.

Figure 3 depicts the Fourier transform infrared (FTIR) ranges of unadulterated  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  and composite containing graphene specimens. The prominent apex at  $1580\text{ cm}^{-1}$  may be ascribed to the stretching vibrancies of the not oxidized carbon skeleton.[36] Correspondingly, the O–H stretching and metamorphosis vibrancies are attributed to the bands at  $3380\text{ cm}^{-1}$  and  $1349\text{ cm}^{-1}$ . The bands at  $1209\text{ cm}^{-1}$  and  $1083\text{ cm}^{-1}$  are caused by epoxy category C–O and C–OH stretching vibrancies [37, 38]. The absorption apex at roughly  $584\text{ cm}^{-1}$ , which is ascribed to the M–O band in  $\text{CoFe}_2\text{O}_4$ , has been reported FTIR range of unadulterated  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$ , and the composite containing graphene specimens revealed effective GO reduction and pureness of the ternary metal oxide.

Scanning electron microscope captures of unadulterated  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  and graphene composite are shown in Fig. 3. The diameter of the agglomerated porous  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  nanostructures is 55 nanometers, as illustrated in Fig. 4a. Figure 4b. shows the SEM captures of  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  composites containing 40% graphene.

The magnetic hysteresis loops of unadulterated  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  and  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  NCs at room temperature are shown in Fig. 4 in which all specimens exhibit ferromagnetic function. As indicated in Fig. 4, the saturation magnetizations (Ms) of the specimens  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  and  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  10 to 40 percent are reported as 80.1, 77.8, 73.6, 67.2 and 64.4 electromagnetic unit /gram, in the same order as mentioned. It is clear that Ms amount declines with GO amount growing due to the addition of nonmagnetic GO. Furthermore, as previously stated, the introduction of GO may effectively adjust the particle size, morphology, and microstructure of  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  NCs, which has a significant impact on the magnetic characteristics of the as

synthesized composites. In the case of Magneto-crystalline anisotropy, form anisotropy, stress anisotropy, and exchange anisotropy influence the coercivity  $H_c$  of magnetic NPs again. Furthermore, deformation of the crystal lattice, local chemical disruption, and exchange interaction interruption all significantly affect the coercivity of NPs. The equivalent coercivity  $H_c$  values for specimens 1–5 are 1681, 613, 517, 764, 423, and 551 Oe. This research presents that as-synthesized nanocomposites adjust  $H_c$  due to the addition of GO, mainly due to the deformation of the crystal lattice, surface atoms' deviation, and changeable anisotropy.[40]

Moreover, based on the natural resonance equation,  $2\pi fr = rHa$ , and  $Ha = 4|K1|/(3\mu_0 Ms)$ ; in the same order as here,  $f$ ,  $r$ ,  $Ha$  and  $|K1|$  equal the resonance frequency, the gyromagnetic ratio, the anisotropy energy, and anisotropy modulus. Consequently, magnetic NCs' natural resonance loss is mainly dictated by their magnetic characteristics. That seems to be the superior magnetic characteristics of as-synthesized NCs that help to enhance the EM wave absorption in the spectrum with low frequency.

Figure 5 depicts the frequency dependence of reflection loss at 2.0 mm thick, including all specimens. According to Fig. 5, the EM wave reflection loss is highly susceptible to the absorber's thickness. The RL apex shifts into a low frequency when the thickness increases. From an application perspective, a value of RL less than -10 decibels is regarded as the threshold value due to the proper 90% EM wave absorption. The EM wave absorption performance of  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  composites is clearly superior to that of the unadulterated  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  ferrite, as illustrated in Fig. 5. In the specimen  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$ , the minimum RL is -11.36 decibels at 12.5 GHz, whereas the effective absorbing bandwidth is 2.5 GHz. And the specimen  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  10% holds a min RL of 13.28 decibels at 14 GHz, equivalent to its effective absorbing bandwidth of 4.6 GHz. The specimen  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  20% has the best EM wave absorbing property, with a min RL of -20.2 decibels at 8.6 GHz and an absorbing bandwidth at 6.9 GHz. The  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  30% holds the maximum EM wave absorption property, with the min RL is -39.4 decibels at 11.5 GHz, and the absorption bandwidth is 6.8 GHz. Interestingly enough, The  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  40% demonstrates an extremely broad effective bandwidth, with the optimum absorbing bandwidth ranging from 8.7 to 18 GHz. At the same time, has 2.0 millimeters thick, and its min RL -25.2 is decibels.

Hereon, the as-synthesized  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  composite indicates great EM wave absorbing capabilities, with premier reflection loss and an extremely wide effective absorption bandwidth, and there is no doubt about that.

## 4. Conclusion

To sum up, a two-step methodology was used to favorably manufacture  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$  nanocomposites. XRD, FT-IR, TEM, XPS, and VSM investigations were used to evaluate the constructional, morphological, surface analytical, and magnetic characteristics of nanocomposites. The Mn and Cu doped cobalt ferrite monotonously surrounded the surface of the rGO sheets, according to the

Co<sub>0.5</sub>Mn<sub>0.3</sub>Cu<sub>0.2</sub>Fe<sub>2</sub>O<sub>4</sub>/rGO nanocomposites results. Moreover, measurements of microwave adsorption parameters indicate that the min RL of Co<sub>0.5</sub>Mn<sub>0.3</sub>Cu<sub>0.2</sub>Fe<sub>2</sub>O<sub>4</sub>/rGO 40% nanocomposites may attain – 25.2 dB at 12.7 GHz for a 2 mm thickness, and only 2 mm thickness in the effective absorption bandwidth (RL under 10 dB) spectrums from 8.7 to 18 GHz. As a result, Co<sub>0.5</sub>Mn<sub>0.3</sub>Cu<sub>0.2</sub>Fe<sub>2</sub>O<sub>4</sub> nanocomposites may be a wise option for microwave-absorbing materials.

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## Figures

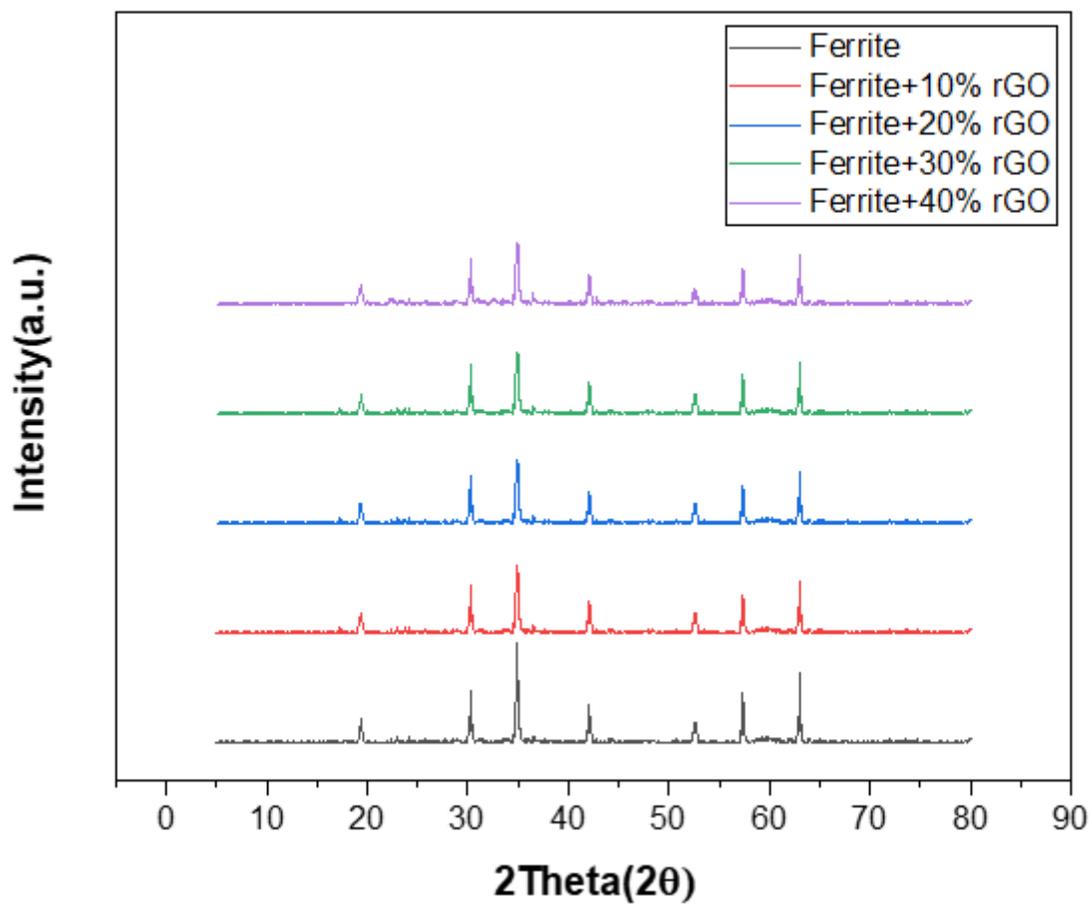


Figure 1

X-Ray Diffraction (XRD) schemas of  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  and  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$ .

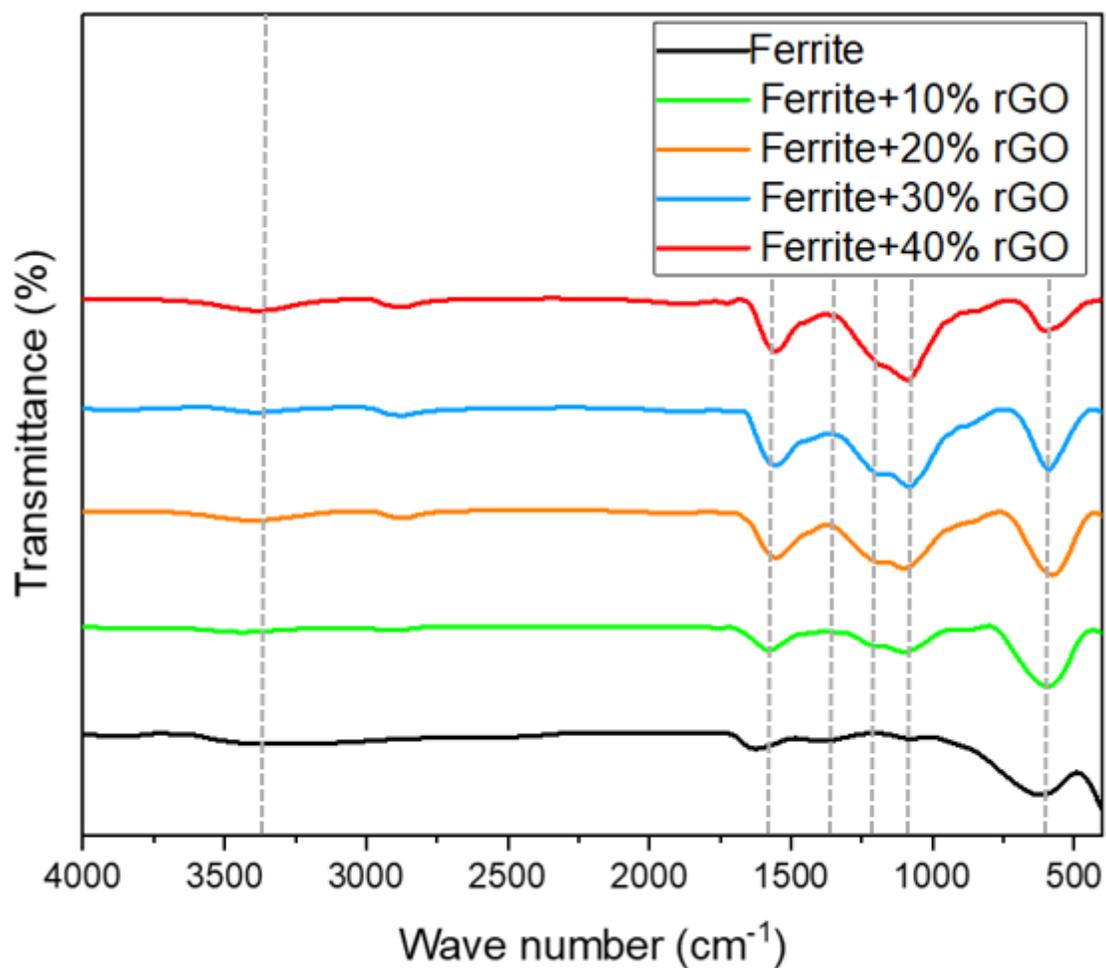
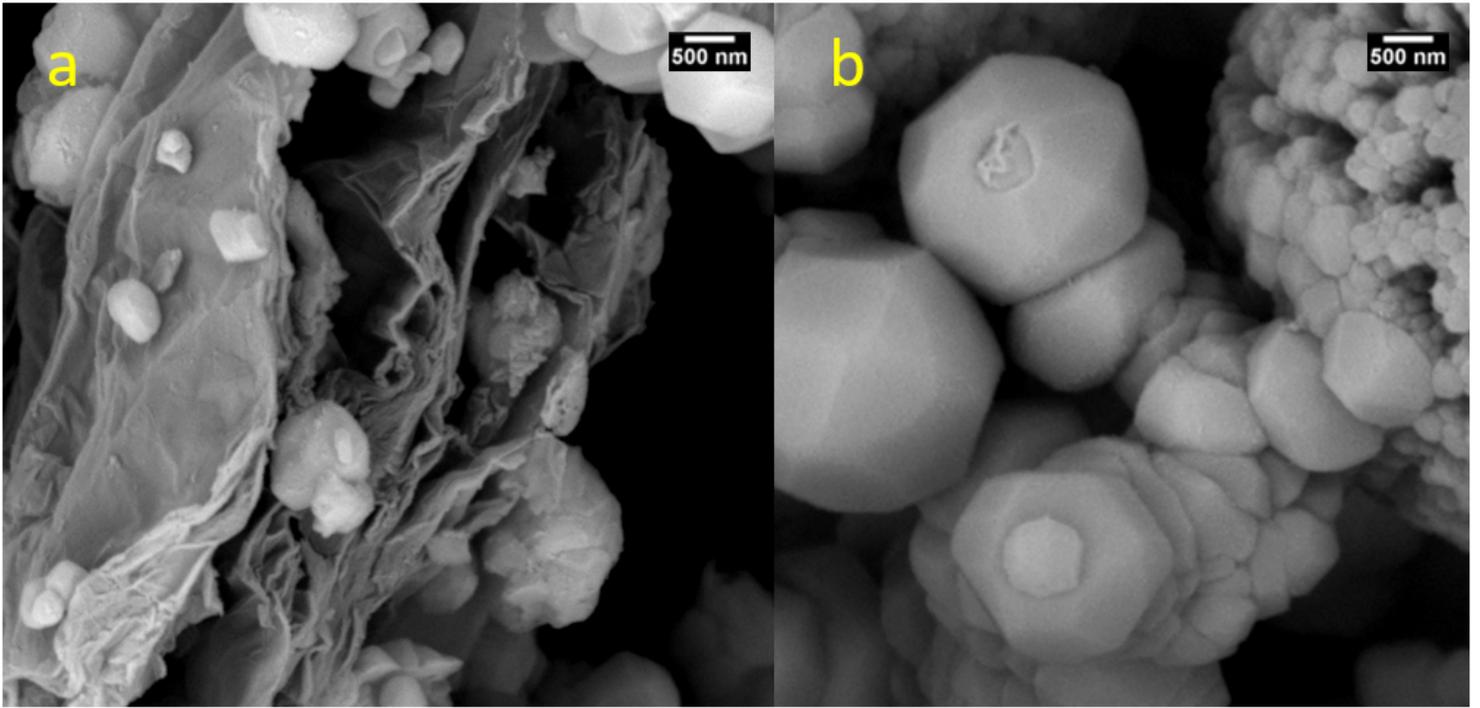


Figure 2

Fourier-transform infrared spectroscopy (FTIR) schemas of  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  and  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4/\text{rGO}$ .



**Figure 3**

Scanning electron microscopy images of  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4 + 40\% \text{rGO}$  (a) and unadulterated  $\text{Co}_{0.5}\text{Mn}_{0.3}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$  (b).

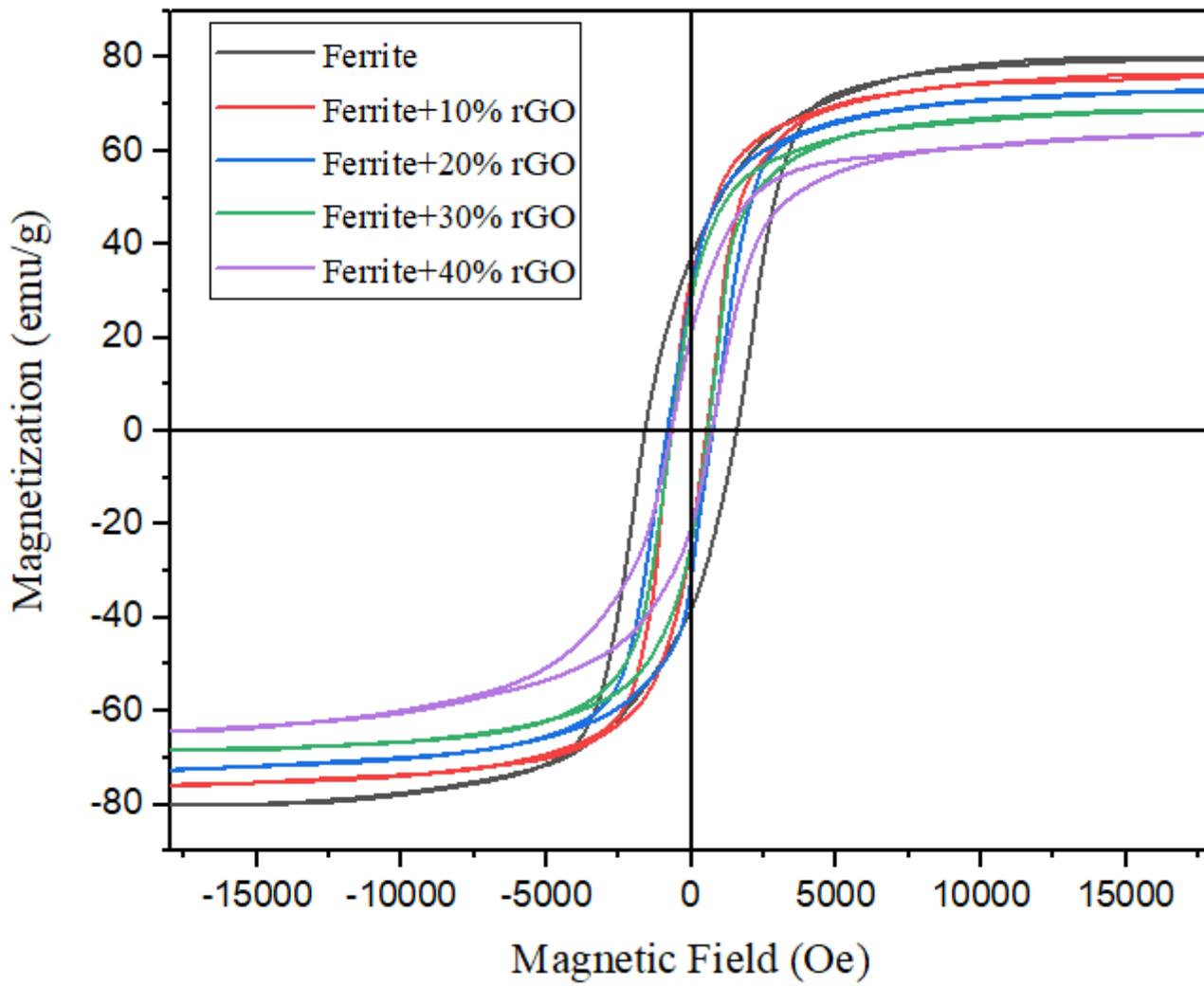


Figure 4

Specimen's hysteresis loops at room temperature.

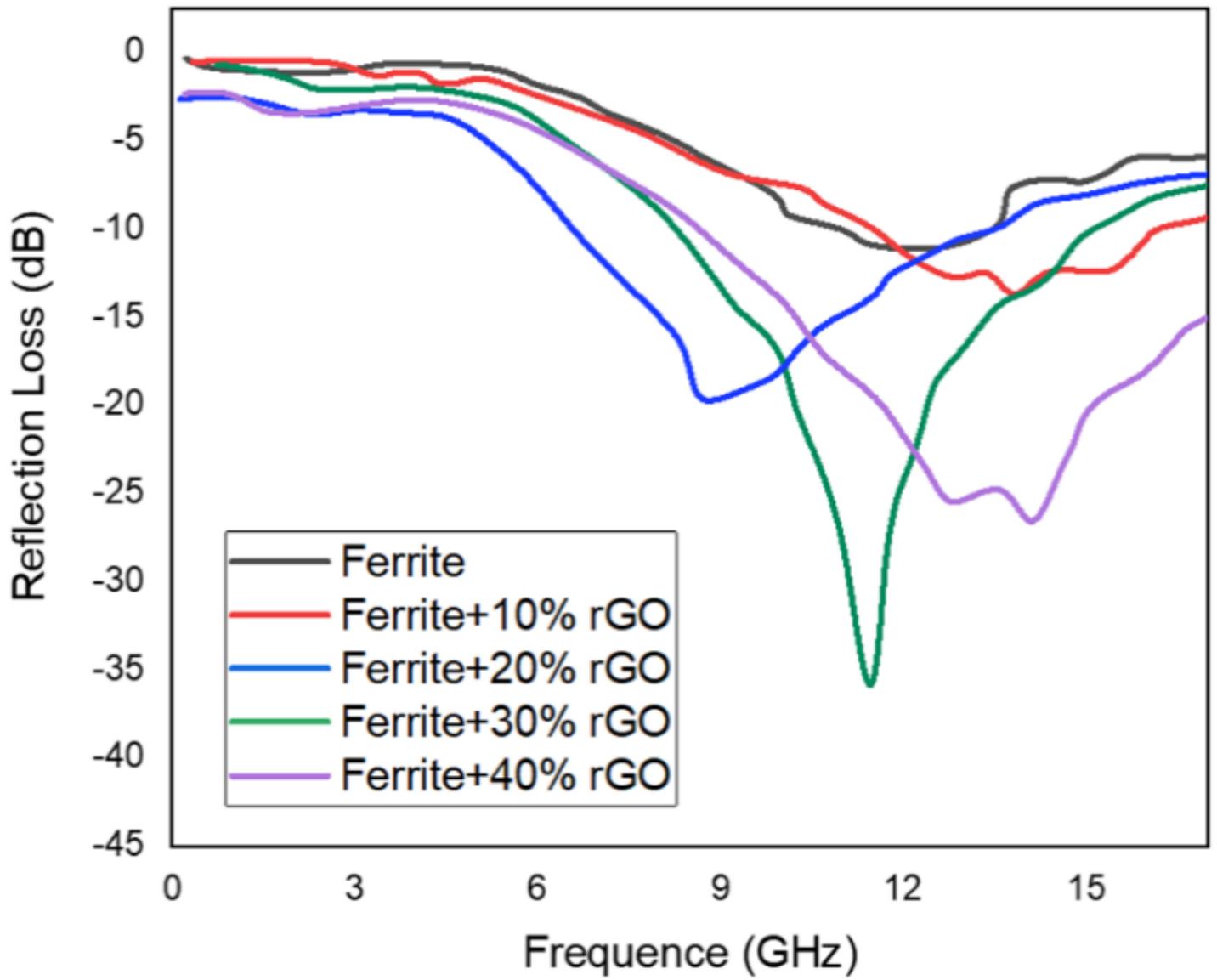


Figure 5

Specimen's reflection loss