

## Research Article

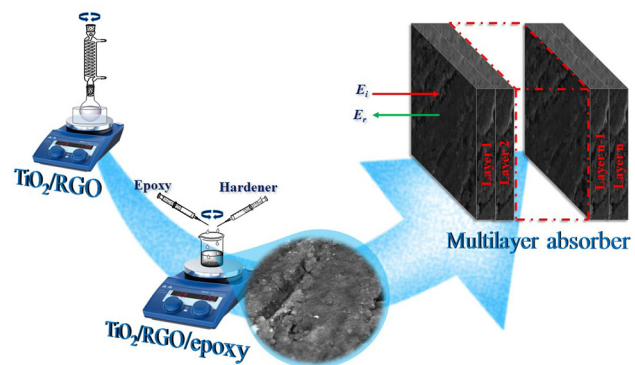
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# Design and optimization of a $\text{TiO}_2$ /RGO-supported epoxy multilayer microwave absorber by the modified local best particle swarm optimization algorithm

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**Abstract:** Microwave absorbers have many applications in medical, industrial, and military devices. Polymeric composites including carbon-based filler can be used as light-weight absorbers with high electromagnetic (EM) wave absorption performance. Hence, multilayer microwave absorbers were designed using titanium dioxide ( $\text{TiO}_2$ )/reduced graphene oxide (RGO)/epoxy nanocomposites with different weight percentages manufactured using refluxing and annealing methods. The characterization of nanocomposite indicated thin layers of  $\text{TiO}_2$ /RGO as divided sheets in epoxy. The EM properties of the nanocomposites were examined using the Nicolson-Ross-Weir (NRW) detection method. The S-parameters were measured using PNA-N522A Microwave Network Analyzer. The multilayer absorber software was designed based on the modified local best particle swarm optimization algorithm by MATLAB software, in which the material and thickness of layers were optimized with two cost functions in X-band frequencies. The first cost function seeks to reach the best absorption bandwidth, and the second cost function seeks to reach the maximum average return loss (RL) of the frequency range of 8.2–12.4 GHz. A maximum bandwidth with an RL of less than −12.81 dB was obtained with a thickness of 2.4 mm. A maximum average RL of −22.1 dB was obtained with a thickness of 2.6 mm. The maximum absorption peak was observed with a thickness of 2.5 mm with −62.82 dB at a frequency of 10.86 GHz.



Graphical abstract

**Keywords:** graphene oxide,  $\text{TiO}_2$  nanoparticles, multilayer microwave absorber, particle swarm optimization algorithm

## 1 Introduction

Developing electronic devices such as wireless telecommunication, radar systems, and local networks has recently led to electromagnetic (EM) wave radiation at different frequencies. Although electronic devices can simplify human life, the EM wave radiation of these devices is a serious environmental problem because it can damage the surrounding electronic devices and harm human health. Hence, efficient approaches must be investigated to control EM pollution effectively [1,2]. In electronic devices, the interference effect of neighboring systems and unwanted internal reflections may create external noises that cause problems in their operation. Therefore, EM absorbers can control those problems by the undesired waves and interferences. Nowadays, EM absorbers have many applications in improving EM compatibility and EM interference problems. By removing unwanted waves, they reduce environmental pollution and improve the performance of electronic devices [3,4].

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For this purpose, nanostructures have been introduced as EM wave absorbers. To achieve highly efficient EM absorbers, two significant steps can be followed [5–10]: (i) designing compounds with particular EM coefficients and (ii) creating the maximum impedance matching, which is obtained by calculating the optimal thickness and arrangement of the layers. This multilayer absorber has better performance than a single-layer absorber. The higher the impedance matching, the less the reflection from the outer layer, and the more power of the radiation wave entering the absorbing medium, and by passing through it, the possibility of EM wave absorption increases.

Conductive polymer-based composites, which consist of polymers and conductive fillers, have been widely used for EM removal because of their considerable properties of high flexibility, lightweight nature, compatibility, good chemical stability, and low-cost processing [11,12]. Epoxy resin is widely used in electronic devices because of its lightweight, thermal stability, mechanical features, simple processing, and corrosion resistance. It can be used as an affordable and available matrix to fabricate EM-absorbing materials [11–14]. Furthermore, polymeric composites with carbon-based fillers, such as carbon nanotubes, carbon black, and graphene, have exhibited excellent EM wave attenuation performance due to their unique electronic and mechanical features [15]. Carbon-based absorbing materials possess high complex permittivity values, outstanding thermal stability, and lightweight [16]. To attain affordable and light absorbers, graphene-based compounds are well known as promising candidates [17]. Graphene, which can be produced through facile chemical processing of graphite, is a two-dimensional lattice of single-layer carbon atoms. It is particularly attractive as an excellent EM wave-absorbing material, especially in its oxide forms, due to its high specific surface area and dielectric constant [18–20]. Graphene oxide (GO) is a graphene nanosheet decorated with oxygen functional groups, such as epoxy, hydroxyl, and carboxyl groups. GO sheets include hexagonal ring-based carbon network having both  $sp^2$  hybridized and  $sp^3$  hybridized carbons [21]. Reduced graphene oxide (RGO), fabricated by removing the oxygen groups of GO, has been introduced to improve EM absorption. This improvement in EM absorption is ascribed to the defects and functional groups [22–24]. Defects and functional groups can enhance impedance-matching characteristics and rapid energy transitions from contiguous states to the Fermi level. However, graphene materials are non-magnetic, and their EM absorption ability is due to their dielectric loss. In addition, the permittivity and permeability of graphene materials do not balance together, leading to bad impedance matching. The composition of graphene materials with other nanostructures can improve their absorption capability [13].

Titanium dioxide ( $TiO_2$ ) is a standard semiconductor metal oxide with wide applications due to its outstanding advantages such as nontoxicity, affordability, chemical stability, good dielectric properties, and high refractive index. It has a band gap of 3–3.2 eV and is available in three polymorphic crystalline forms: anatase, rutile, and brookite. Its low dielectric loss and high dielectric constant make it applicable as a nanofiller in a polymeric matrix for EM absorption [25–30].

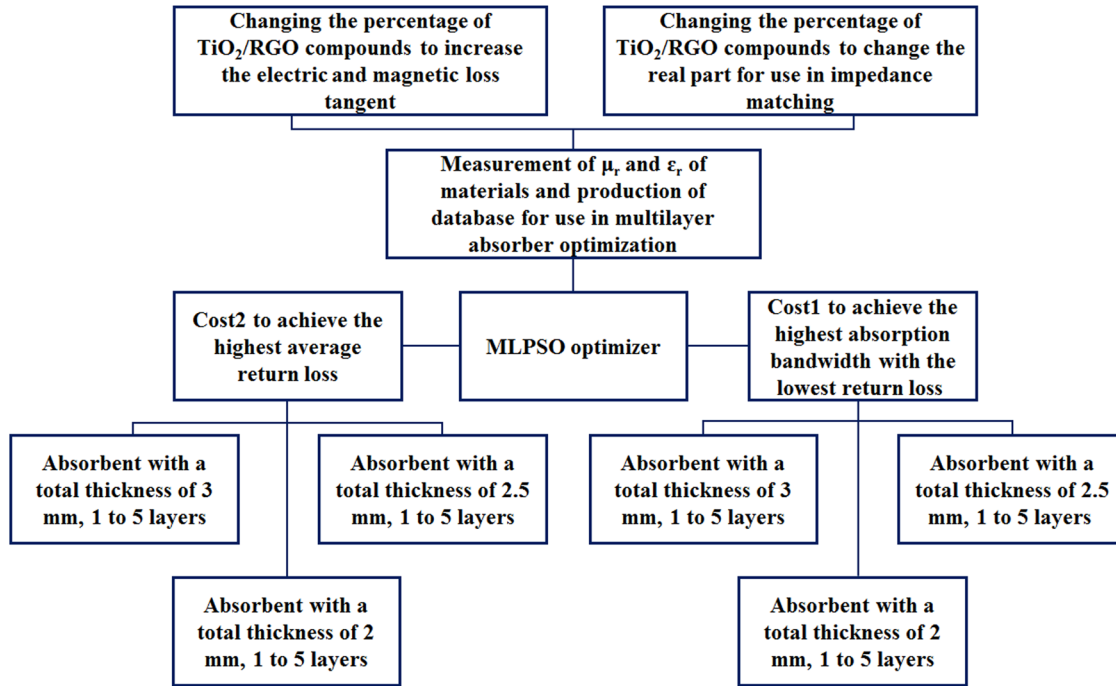
The particle swarm optimization (PSO) algorithm is a random optimization technique inspired by the social behavior of bird herding and fish training [31,32]. This population-based self-adaptive algorithm has been used for many system problems. Since the order and thickness of the layers are significant in multilayer absorbers and the change in each one causes a change in absorber's performance, its optimization is critical. Multi-objective optimizers are used in designing and manufacturing multilayer absorbers, the most common of which are the genetic algorithm and PSO algorithm [7,33–35]. In this work, the modified local best particle swarm optimization (MLPSO) is used, a modified version of PSO, because it has high speed and accuracy and is easy to implement.

Hence, in this work, the  $TiO_2$ /RGO was synthesized using refluxing and thermal reduction methods and then dispersed in epoxy resin as filler *via* the stirring method. The EM features of the as-prepared nanocomposites (the complex permittivity and permeability coefficients) were perused using the Nicolson-Ross-Weir (NRW) method of the ASTM D5568-14, in which the samples were placed in the rectangular waveguide setup. Their S-parameters in the microwave X-band frequency range (8.2–12.4 GHz) were measured by the PNA N5222A microwave network analyzer. The MLPSO algorithm was used to design a multilayer absorber, in which the material and thickness of layers were optimized with two cost functions in X-band frequencies. The optimal values of average return loss (RL) of –22.1 dB and absorption peak of –62.82 dB were acquired with the thickness of 2.6 and 2.5 mm, respectively. Figure 1 illustrates a flowchart of the design and optimization process of the multilayer absorber.

## 2 Experimental

### 2.1 Materials and instrumentation

Chemical materials (powdered flake graphite, sodium nitrate, sulfuric acid, and potassium permanganate) to synthesize GO



**Figure 1:** Flowchart showing the steps of design and optimization.

with high purity were purchased from the Merck company. TiO<sub>2</sub> nanoparticles (TiO<sub>2</sub> NPs), including biphasic anatase and rutile phases with a ratio of 80 to 20, were prepared from Degussa Co. (Germany). Epoxy resins with the commercial names resin 577 and hardener 567 were provided by the composite materials Co. (Turkish) supplier. X-ray diffraction (XRD, Italstructure, ADP200) patterns in 2 $\theta$  degree range of 10–80° were performed to identify the crystalline phases present in the samples. Fourier transform infrared spectroscopy (FT-IR, Thermo Nicolet) was performed to detect the functional groups of the sample. Scanning electron microscopy (SEM, TSCAN) images were used to characterize the surface features and evaluate the morphological changes. The EM properties of the materials were calculated via an N5222A PNA Microwave Network Analyzer. Complex permittivity and permeability coefficients were calculated to measure the amount of reflection and transmission of microwave waves from the materials.

## 2.2 TiO<sub>2</sub>/RGO/epoxy nanocomposite fabrication

GO nanosheets were synthesized via the oxidation of graphite through Hummer's method [36]. To fabricate the GO–TiO<sub>2</sub> nanocomposite, 0.03 g of GO and 0.03 g of TiO<sub>2</sub> NPs were separately dispersed in 100 ml of deionized water under

sonication for 45 min to acquire homogenous suspensions. TiO<sub>2</sub> suspension was then added to the GO suspension. The GO–TiO<sub>2</sub> suspension was refluxed for 10 h at 80°C. An annealing method was used to prepare the RGO–TiO<sub>2</sub> nanocomposite, in which the GO–TiO<sub>2</sub> suspension was dried at 180°C for 3 hours. Different weight percentages of 7RGO:3TiO<sub>2</sub>, 3RGO:7TiO<sub>2</sub>, and 5RGO:5TiO<sub>2</sub> were fabricated. These nanocomposites were added to epoxy resin with a specific weight ratio as shown in Table 1 to prepare TiO<sub>2</sub>-RGO-epoxy. The ingredients were mixed well for 20 min in 1.5 ml of ethanol using stirrer and poured into the mold. The samples were fabricated in a size of 10.1 × 22.8 × 3 and 10.1 × 22.8 × 2 mm<sup>3</sup>. Figure 2 illustrates the TiO<sub>2</sub>/RGO/epoxy nanocomposite preparation setup.

**Table 1:** Amount of material used for the preparation of TiO<sub>2</sub>/RGO/epoxy nanocomposite

| Sample name | TiO <sub>2</sub> (mg) | GO (mg) | RGO (mg) | Epoxy (mg) |
|-------------|-----------------------|---------|----------|------------|
| S1          | 750                   | —       | —        | 750        |
| S2          | —                     | 355     | —        | 750        |
| S3          | —                     | 750     | —        | 750        |
| S4          | 225                   | 525     | —        | 750        |
| S5          | 375                   | 375     | —        | 750        |
| S6          | 525                   | 225     | —        | 750        |
| S7          | 525                   | —       | 225      | 750        |
| S8          | 375                   | —       | 375      | 750        |
| S9          | 225                   | —       | 525      | 750        |

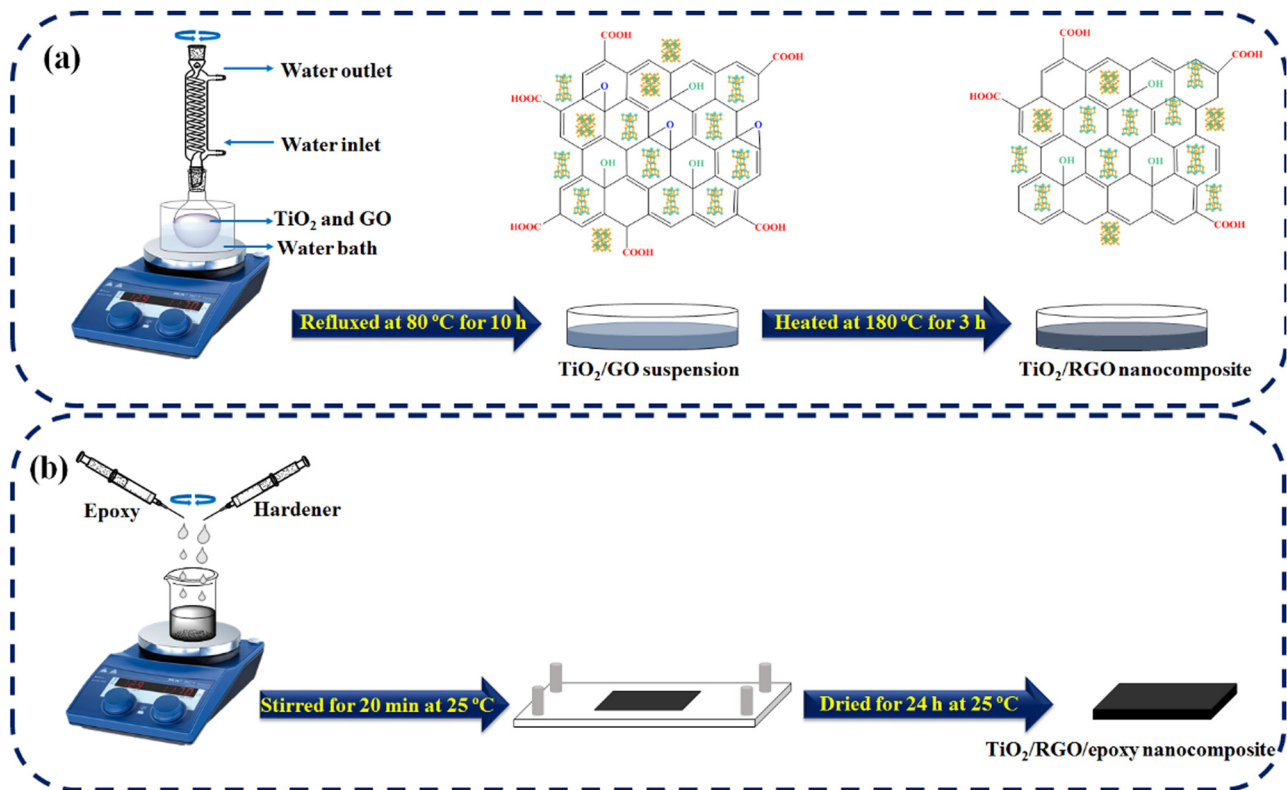


Figure 2: Schematic of the preparation of (a) TiO<sub>2</sub>/RGO and (b) TiO<sub>2</sub>/RGO/epoxy nanocomposites.

### 2.3 MLPSO

The PSO is a population-based self-adaptive algorithm with inherent parallelism, introduced in 1995 by Kennedy and Eberhart [32]. It is a global random optimization technique inspired by the social behavior of bird flocking and fish schooling. Because PSO has high speed, good convergence, and easy implementation, this algorithm is widely used in different optimization problems. MLPSO is one of the versions of PSO that prevents premature convergence of PSO and increases its accuracy. This method divides society into several local groups that do not overlap. In PSO, the random solutions are particles with a velocity in the search space that fly through a hyper-dimensional search space. In addition, the velocity and position of each particle update based on its best position in the search space [33]. Each particle is considered a point of the D-dimensional search space, and the  $i$ th particle is introduced by  $x_i = (x_{i1}, x_{i2}, \dots, x_{iD})$ . The best previous position and velocity of the  $i$ th particle are demonstrated as  $p_i = (p_{i1}, p_{i2}, \dots, p_{iD})$  and  $v_i = (v_{i1}, v_{i2}, \dots, v_{iD})$ , respectively. The D-dimensional vector of  $p_g = (p_{g1}, p_{g2}, \dots, p_{gD})$  is shown to be the best particle in the population [37]. If the  $g$  index shows the best particle in the population, the velocity and position are acquired by the following equations [33,38]:

$$v_i(d+1) = \omega v_i(d) + c_1 r \text{ and } (p_i(d) - x_i(d)) + c_2 r \text{ and } (p_g(d) - x_i(d)), \quad (1)$$

$$x_i(d+1) = x_i(d) + v_i(d+1), \quad (2)$$

where  $v_i(d)$  and  $x_i(d)$  illustrate the velocity and position of the  $i$ th particle, respectively,  $\text{rand}$  is a uniformly distributed random number between 0 and 1, and  $\omega$  is the inertia weight. At the same time,  $c_1$  and  $c_2$  have denominated acceleration constants. The PSO algorithm is based on two versions of the global neighborhood (g-best) and local neighborhood (l-best). The aforementioned equations describe the gbest version of the PSO. For the lbest version,  $p_g(d)$  should be changed to  $p_l(d)$ . Although the PSO algorithm is facile and computationally effective, its efficiency is unsatisfactory in solving complex multimodal optimization problems. Hence, modifying the PSO algorithm can improve its efficiency [33].

Quick convergence in solving complex multimodal problems is the major imperfection of PSO-based algorithms. Local PSO spans a wider search space; however, it has slow convergence. An MLPSO solution has been presented to prevent local optimum traps in complex search environments, in which neighborhoods do not overlap. Therefore, trapping the particle of a neighborhood into

a locally optimized problem will not affect the other neighborhoods. Furthermore, one particle can only trap *via* each local optimum, increasing the chance of finding a global optimum by population [33].

## 2.4 Transmission line theory

Transmission line theory clarifies the results in terms of the transmitted/reflected waves, which was used to describe the electrical properties of nanocomposites. When a microwave enters into a nanocomposite consisting of absorber materials, the RL value can be acquired through the transmission line theory by the following equation [39–43]:

$$RL(\text{dB}) = 20 \log(\Gamma), \quad (3)$$

$$\Gamma = \frac{Z_{\text{in}} - Z_0}{Z_{\text{in}} + Z_0}, \quad (4)$$

$$Z_{\text{in}} = Z_0 \sqrt{\frac{\mu_r}{\epsilon_r}} \tanh \left[ j \left( \frac{2\pi df}{c} \right) \sqrt{\mu_r \epsilon_r} \right], \quad (5)$$

where  $\Gamma$  is the interface reflection coefficient, and  $Z_{\text{in}}$  and  $Z_0$  are the input impedance at the accessible space/nanocomposite interface and the intrinsic impedance of free space, respectively. In addition,  $\mu_r$ ,  $\epsilon_r$ , and  $d$  are the complex relative permeability, complex relative permittivity, and thickness of the sample.  $f$  and  $c$  also show the microwave frequency and velocity of light in a vacuum.  $j$  is the complex constant ( $j = \sqrt{-1}$ ). The microwave absorbent of a multilayer sample for normal incidence was calculated by the following equation [44,45]:

$$z_1 = \eta_1 \frac{z_2 + \eta_1 \tanh(k_1 d_1)}{\eta_1 + z_2 \tanh(k_1 d_1)}, \quad (6)$$

$$z_n = \eta_n \frac{z_{n+1} + \eta_n \tanh(k_n d_n)}{\eta_n + z_{n+1} \tanh(k_n d_n)}, \quad (7)$$

where  $Z_n$  is the input impedance of the  $n$ th layer, and  $\eta_n$  and  $k_n$  are the impedance and propagation constant of the  $n$ th layer, respectively.

$$\eta_n = \eta_0 \sqrt{\frac{\mu_{rn}}{\epsilon_{rn}}}, \quad (8)$$

$$k_n = j\omega(\mu_0 \epsilon_0 \mu_{rn} \epsilon_{rn})^{1/2} = \frac{j2\pi}{\lambda} \sqrt{\mu_{rn} \epsilon_{rn}}, \quad (9)$$

where  $\omega$  and  $\lambda$  are the radian frequency and wavelength, respectively, and  $\mu_{rn}$  and  $\epsilon_{rn}$  are the complex relative permeability and complex relative permittivity of the  $n$ th layer, respectively. Accordingly, the multilayer absorber possesses the RL as follows:

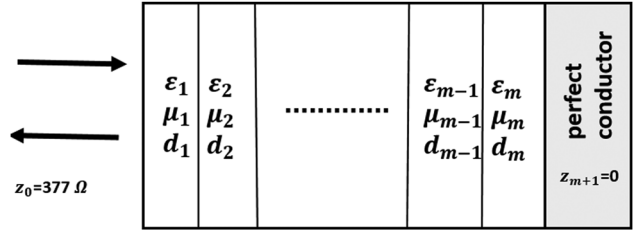


Figure 3: Multilayer absorber configuration.

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

Figure 3 shows a brief explanation of the multilayer absorber configuration.

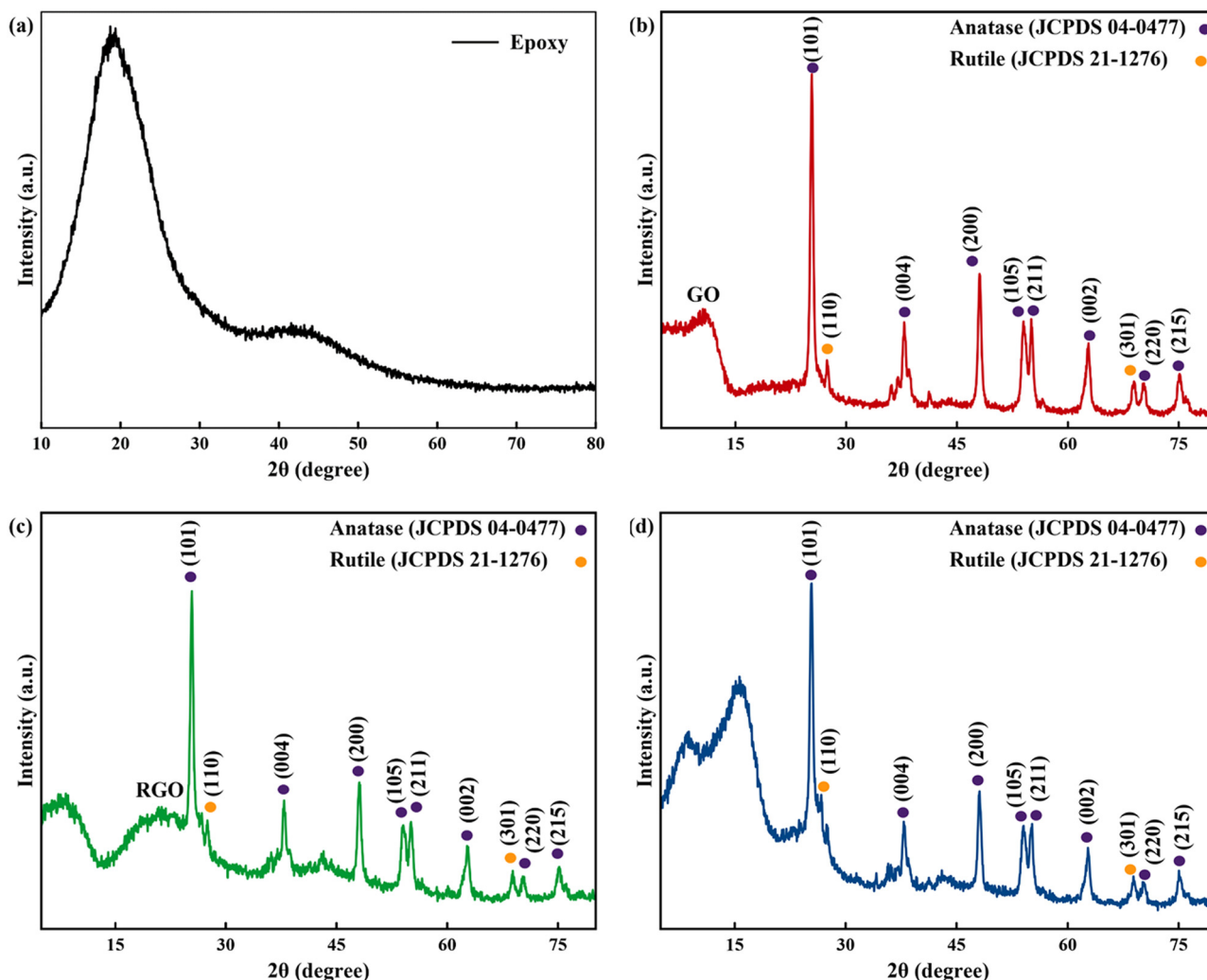
## 3 Results and discussion

### 3.1 Materials' characterization

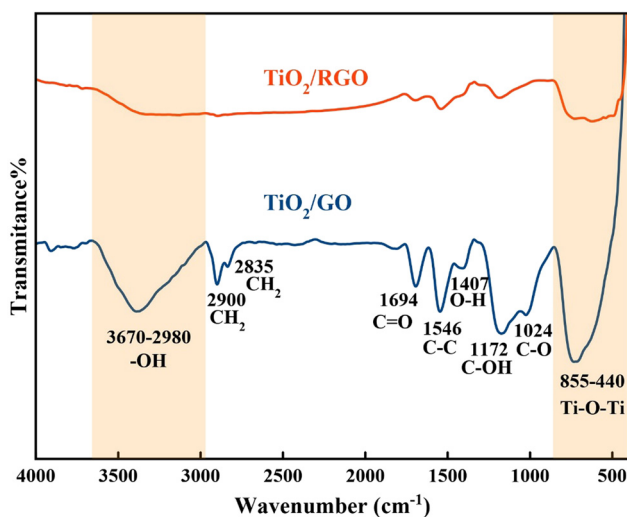
XRD analysis was deployed to study the crystalline structure and phase composition. As shown in Figure 4, the samples' XRD spectra confirm the nanocomposite's successful formation.

The XRD spectrum of the pure epoxy presents a wide diffraction peak (Figure 4(a)), indicating the amorphous phase of the epoxy resin. Figure 4(b) show sharp peaks at 25.4°, 37.9°, 48.2°, 54°, 55°, 62.8°, 70.4°, and 75.3°, assigned to the anatase phase (81%) of TiO<sub>2</sub>. The diffraction peaks at 27.5° and 68.9° can be assigned to the rutile phase (19%), indicating that the main phase of TiO<sub>2</sub> NPs is anatase [46]. The crystalline size of the TiO<sub>2</sub> NPs is found to be 24.7 nm, calculated by Scherer's formula and full width at half maximum of the (101) peak [47]. The peak at 10.9° (Figure 4(b)) is associated with the formation of the GO owing to graphite oxidation. There is a broad characteristic peak at 16–24.1°, which is attributed to the single layer of the RGO in the spectrum corresponding to the TiO<sub>2</sub>/RGO (30:70) (Figure 4(c)) [48–50]. Therefore, it can be concluded that the GO powder was incompletely converted to RGO. Moreover, the peaks of TiO<sub>2</sub> NPs show lower intensity compared to the spectrum of TiO<sub>2</sub>/GO (Figure 4(b)) due to the immobilization of the RGO surface. Figure 3(d) presents the XRD pattern of TiO<sub>2</sub>/RGO/epoxy (30:70:50).

Figure 5 shows the FT-IR spectra of TiO<sub>2</sub>/GO and TiO<sub>2</sub>/RGO nanocomposites. A wideband appeared in the 855–440 cm<sup>-1</sup> range, indicating stretching vibrations of Ti–O in Ti–O–Ti bands [51–53]. The different peaks appearing at 1,546 and 1,694 cm<sup>-1</sup>



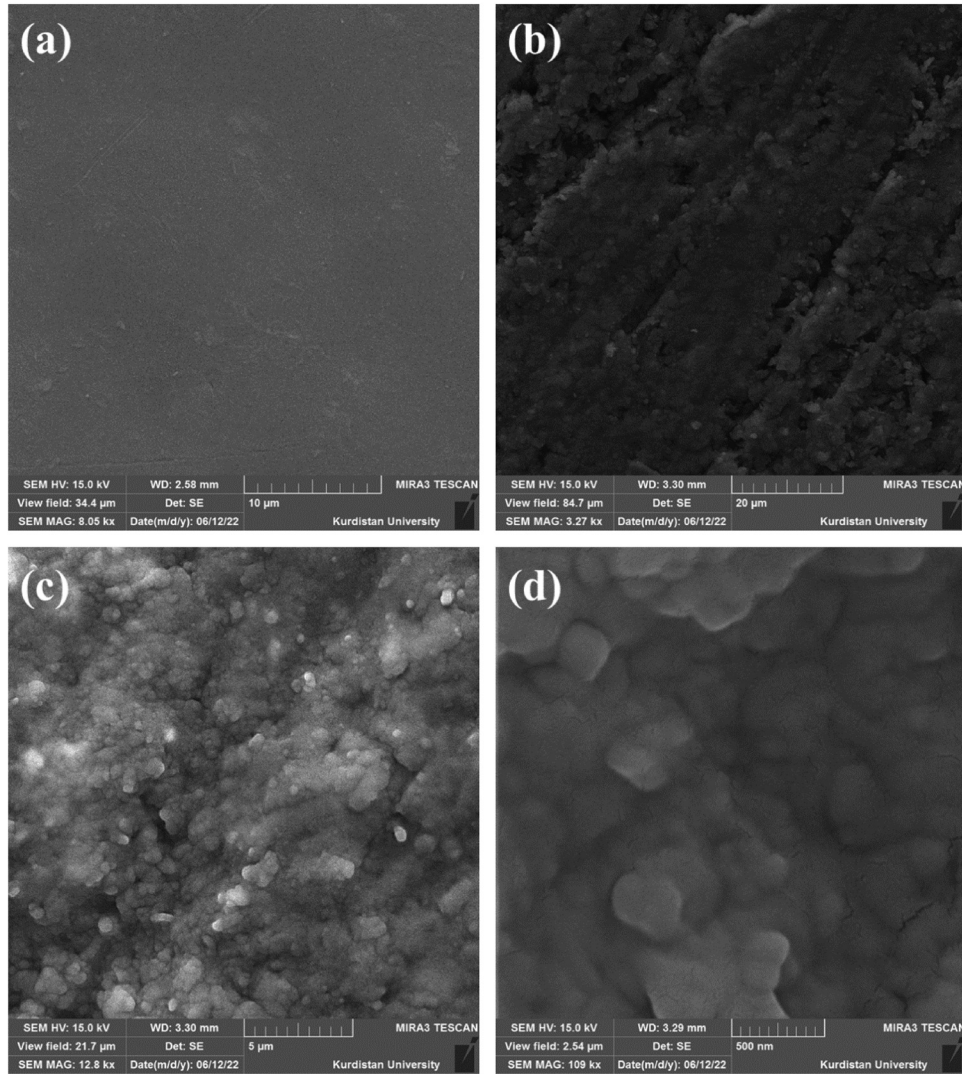
**Figure 4:** XRD pattern of (a) epoxy, (b)  $\text{TiO}_2/\text{GO}$  (30:70), (c)  $\text{TiO}_2/\text{RGO}$  (30:70), and (d)  $\text{TiO}_2/\text{RGO}/\text{epoxy}$  (30:70:50).



**Figure 5:** FT-IR spectra of  $\text{TiO}_2/\text{GO}$  (30:70) and  $\text{TiO}_2/\text{RGO}$  (30:70) nanocomposite.

correspond to the vibration mode of  $\text{C}=\text{C}$  and  $\text{C}=\text{O}$  of  $\text{COOH}$  groups, respectively. The vibration mode of  $\text{C-O}$  and  $\text{C-OH}$  also appears at  $1,024$  and  $1,172\text{ cm}^{-1}$ , respectively. In addition, two peaks at  $2,900$  and  $2,835\text{ cm}^{-1}$  can be indexed as a  $\text{C-H}$  vibration [53,54]. A broad band at  $3,670\text{--}2,980\text{ cm}^{-1}$  and a weak peak at  $1,407\text{ cm}^{-1}$  are imputed to stretching vibrations of  $\text{OH}$  [55–57]. The FT-IR spectrum of  $\text{TiO}_2/\text{RGO}$  nanocomposite presents weak peaks owing to the decomposition of  $\text{GO}$  bonds through the synthesis of  $\text{RGO}$ .

SEM analysis of cured epoxy composites was performed to determine the exfoliation and dispersion of  $\text{TiO}_2$  NPs,  $\text{RGO}$ , and  $\text{TiO}_2/\text{RGO}$  nanocomposite in the matrix. Figure 6 shows the morphology of pure epoxy resin and  $\text{TiO}_2/\text{RGO}/\text{epoxy}$  (30:70:50) in different magnifications. A smooth surface of pure epoxy can be clearly seen in Figure 6(a). Figure 6(b)–(d) reveals that  $\text{TiO}_2/\text{RGO}$  forms thin layers as divided sheets with great dispersion in the



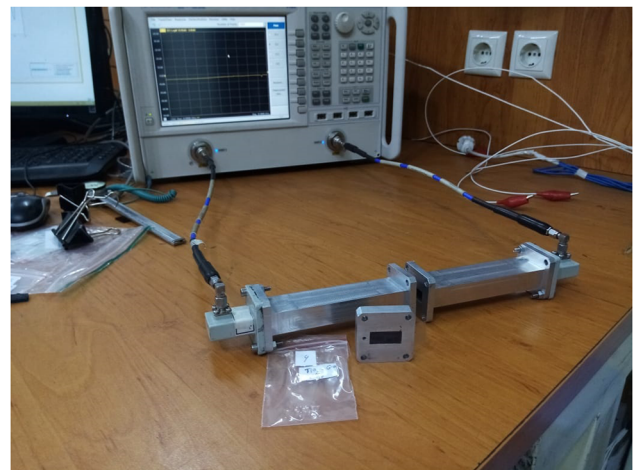
**Figure 6:** SEM images of (a) pure epoxy and (b)–(d) TiO<sub>2</sub>/RGO/epoxy (30:70:50) in different scales.

epoxy matrix. Therefore, this nanofiller in the epoxy resin at 50 wt% can improve epoxy performance.

Furthermore, it is found that the roughness of cured composite epoxy is higher than that of cured pure epoxy and that the cured composite epoxy exhibits more irregular and coarse surface features.

### 3.2 EM properties of the TiO<sub>2</sub>/RGO/epoxy

The RL has strongly pertained to the  $\mu_r$  and  $\epsilon_r$  of the samples, which influence the impedance matching and attenuation characteristics. The microwave absorption features of the samples determine the impedance matching and attenuation characteristics. The impedance matching indicates if the microwave can transmit through the samples, and the



**Figure 7:** Setup for measuring EM properties of samples by NRW method with microwave network analyzer.

attenuation characteristic is a factor for the evaluation of the ability of the samples toward the conversion of the microwave energy to heat dissipation [39]. Figure 7 shows the NRW method. This setup includes a pair of 3.5 mm coax-to-waveguide adapters, two coaxial cables with SMA connectors, two waveguide standard sections, an N5222A PNA microwave network analyzer, and a waveguide shim. The aperture dimensions of waveguides follow WR-90 rectangular waveguide size. The manufactured samples were placed in the rectangular aperture of the WR-90 waveguide and connected to an N5222A PNA Microwave Network Analyzer. Their S-parameters were measured in the 8.2 to 12.4 GHz frequency range. Therefore, their permittivity and permeability coefficients were calculated using the NRW formula [58] as follows:

$$\mu_r^* = \mu' - j\mu'' = \frac{2\pi}{A\sqrt{k_0^2 - k_c^2}} \left( \frac{1 + \Gamma}{1 - \Gamma} \right), \quad (11)$$

$$\varepsilon_r^* = \varepsilon' - j\varepsilon'' = \frac{1}{\mu_r^* k_0^2} \left( \frac{4\pi^2}{A^2} + k_c^2 \right), \quad (12)$$

$$\Gamma = X \pm \sqrt{X^2 - 1}, \quad (13)$$

$$X = \frac{S_{11}^2 - S_{21}^2 + 1}{2S_{11}}, \quad (14)$$

$$T = \frac{S_{11} + S_{21} - \Gamma}{1 - (S_{11} + S_{21})\Gamma}, \quad (15)$$

$$\frac{1}{A^2} = - \left[ \frac{1}{2\pi} \ln(T) \right]^2, \quad (16)$$

where  $S_{ij}$  is the scattering parameter of the 2-port microwave network;  $\mu_r^*$  is the relative complex permeability of specimen,  $\varepsilon_r^*$  is the relative complex permittivity of specimen,  $k_0 = \frac{2\pi}{\lambda_0}$  is the wavenumber in free space,  $k_c = \frac{2\pi}{a}$  is the waveguide cutoff wavenumber (rad/m);  $a = 22.86$  is the width of rectangular waveguide (m),  $\lambda_0 = \frac{c_0}{f}$  is the wavelength in free space (m);  $c_0$  is the speed of light in free space; and  $f$  is the measurement frequency (Hz).

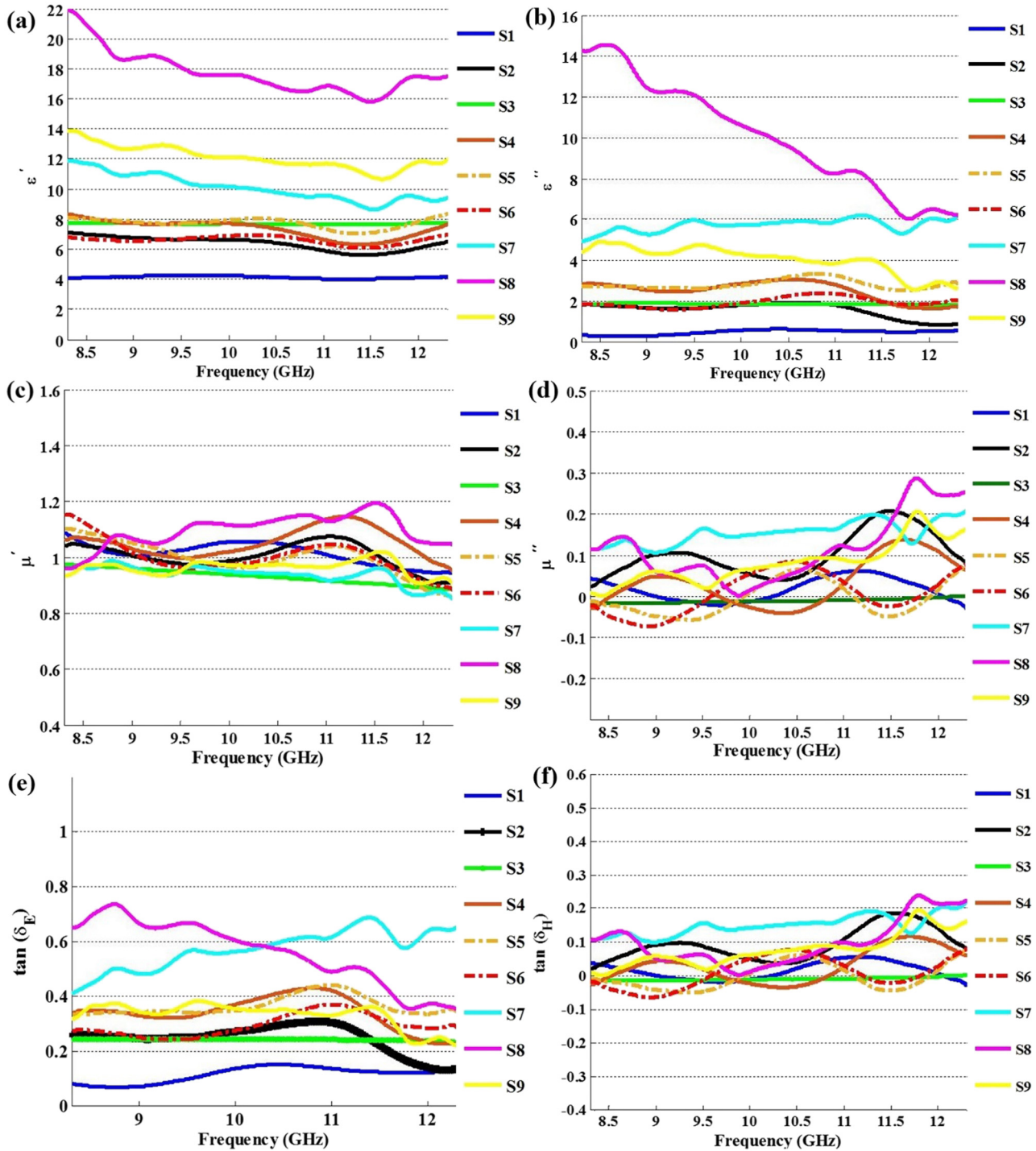
The real ( $\varepsilon'$  and  $\mu'$ ) and imaginary ( $\varepsilon''$  and  $\mu''$ ) parts of  $\varepsilon_r$  and  $\mu_r$  were calculated for the samples, as depicted in Figure 8. The real parts are related to energy storage. In contrast, the imaginary parts are attributed to energy loss for electric/magnetic fields. According to Figure 8(a) and (b), the TiO<sub>2</sub>/epoxy is a pure dielectric material and shows almost constant values over the whole frequency range. After the incorporation of the GO and TiO<sub>2</sub> composition into the epoxy matrix, both  $\varepsilon'$  and  $\varepsilon''$  are slightly improved in the frequency range of 8.2–12.4 GHz compared with S1 and S2. However, increasing the TiO<sub>2</sub> content leads to a reduction of the permittivity (S6 and S7). This means that a high TiO<sub>2</sub> percentage leads to a reduction of the dielectric

constants. The reduction of GO to RGO enhances both parts of permittivity, leading to the composition of TiO<sub>2</sub>, RGO, and epoxy being able to absorb more microwave energy. Adding the TiO<sub>2</sub> and RGO causes more capacitance and increases dielectric constants [59]. Multiple polarization consisting of interfacial and dipole polarization and some remaining oxygen functional groups of GO improve the permittivity [60]. The annealing process improves the graphitization and conductivity of RGO, increasing the real and imaginary parts of the permittivity [61]. Figure 8(c) and (d) illustrates the real and imaginary parts of the permeability in the frequency range of 8.2–12.4 GHz, respectively. The  $\mu'$  behavior of the samples showed almost constant values around 1 from 8.2 to 12.4 GHz. Among the samples, the maximal microwave magnetic loss value was achieved for S8 in the X-band (8.2–12.4 GHz) frequency. Dielectric loss ( $\tan \delta_E = \varepsilon''/\varepsilon'$ ) and magnetic loss ( $\tan \delta_H = \mu''/\mu'$ ) are depicted in Figure 8(e) and (f). The highest  $\tan \delta_E$  can be observed for S7 and S8 over the whole frequency range, suggesting that these specimens are more conducive to microwave absorption. Due to magnetic losses, all samples have low magnetic field attenuation. Therefore, the electric losses are the dominant properties of the proposed samples to design multilayer absorbers.

### 3.3 Transmission line method and impedance matching optimization

In the case of multilayer absorbers, the number of layers and the total thickness are determined as design constraints. However, the material arrangement and the layer thickness are the variables that create the statistical population. For example, if there are nine types of material to design a 3-layer absorber, the minimum thickness change step is considered equal to 0.1 mm, and the total thickness is determined to be 3 mm maximum, each point has six dimensions  $p_i = (n_1, n_2, n_3, t_1, t_2, \text{ and } t_3)$ . The three primary dimensions are related to the type of material in each layer, each of which has nine possibilities. Furthermore, the other three dimensions are related to the thickness of each layer, which can vary between 0.1 and 3 mm. The RL is calculated for each point using the transmission line theory method in the X-band range for each frequency with a step of 0.01 GHz.

Moreover, the cost function 1 and 2 described below will determine the score of each point for subsequent comparisons. The optimization starts with 50 local groups of 5 people, which are randomly created, and according to the obtained results, the p-best, l-best, and g-best points are identified and stored, and then, they start their movement

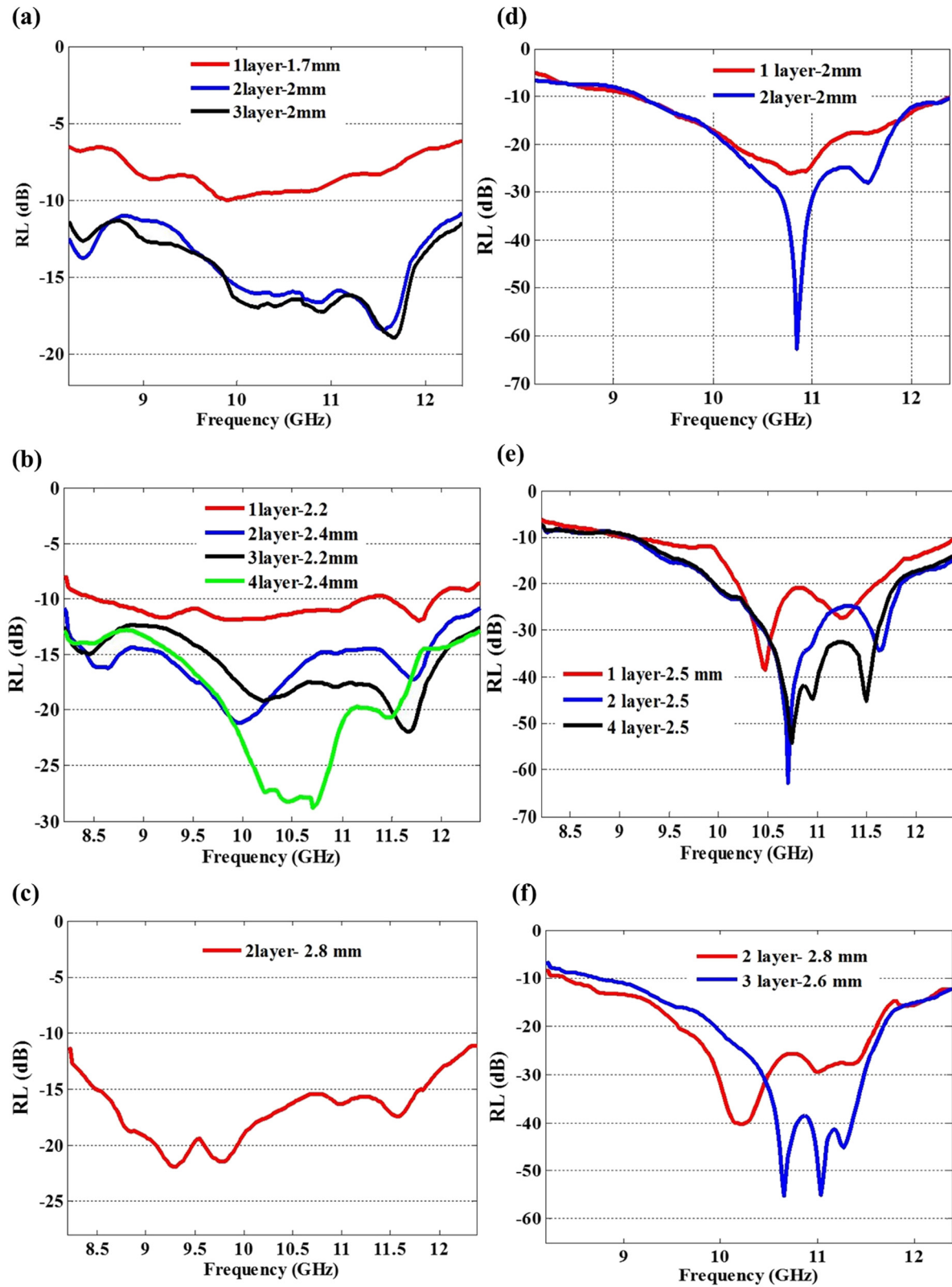


**Figure 8:** Frequency dependence of (a) and (b) complex permittivity, (c) and (d) complex permeability, (e) dielectric loss, and (f) magnetic loss of samples.

with the described speed and acceleration in the direction of l-best. In each step, p-best, l-best, and g-best are retrieved to identify the best. This operation is repeated until the points are concentrated in g-best, and after the desired convergence in enough iterations, the iteration loop can be ended. Moreover, finally, the best multilayer absorber

and its performance will be reported in g-best. Two cost functions were defined as shown in the following: Cost function 1 (to achieve the maximum absorption bandwidth with the lowest RL):

$$\text{cost}_1(n_1, t_1, n_2, t_2, \dots) = \max\{\min_i |I(f_i)|\}. \quad (17)$$



**Figure 9:** Sample optimization by (a)–(c) cost function 1 and (d)–(f) cost function 2. (a) Cost1-2 mm, (b) Cost1-2.5 mm, (c) Cost1-3 mm, (d) Cost2-2 mm, (e) Cost2-2.5 mm, and (f) Cost2-3 mm.

**Table 2:** Samples optimized by cost function 1 with limitation of total thickness 2 mm and the number of layers from 1 to 5

| N layers | Material in layers | Layer thickness (mm) | Min RL (dB) | Max RL (dB) | Mean RL (dB) | RL < -10 (dB) | RL < -20 (dB) |
|----------|--------------------|----------------------|-------------|-------------|--------------|---------------|---------------|
| 1 Layer  | S8                 | 1.7                  | -6.15       | -9.98       | -8.41        | —             | —             |
|          | Total thickness    | 1.7                  |             |             |              |               |               |
| 2 Layers | S8                 | 0.5                  | -10.82      | -18.41      | -14.23       | X band        | —             |
|          | S6                 | 1.5                  |             |             |              |               |               |
|          | Total thickness    | 2                    |             |             |              |               |               |
| 3 Layers | S8                 | 0.5                  | -11.33      | -18.89      | -14.67       | X band        | —             |
|          | S1                 | 0.2                  |             |             |              |               |               |
|          | S5                 | 1.3                  |             |             |              |               |               |
|          | Total thickness    | 2                    |             |             |              |               |               |

**Table 3:** Samples optimized by cost function 1 with limitation of total thickness 2.5 mm and the number of layers from 1 to 5

| N layers | Material in layers | Layer thickness (mm) | Min RL (dB) | Max RL (dB) | Mean RL (dB) | RL < -10 (dB) | RL < -20 (dB) |
|----------|--------------------|----------------------|-------------|-------------|--------------|---------------|---------------|
| 1 Layer  | S7                 | 2.2                  | -8.07       | -11.94      | -10.72       | 3.2 GHz       | —             |
|          | Total thickness    | 2.2                  |             |             |              |               |               |
| 2 Layers | S8                 | 0.8                  | -10.82      | -21.15      | -15.79       | X band        | 0.3 GHz       |
|          | S6                 | 1.6                  |             |             |              |               |               |
|          | Total thickness    | 2.4                  |             |             |              |               |               |
| 3 Layers | S8                 | 0.2                  | -12.35      | -21.97      | -16.1        | X band        | 0.2 GHz       |
|          | S7                 | 0.4                  |             |             |              |               |               |
|          | S6                 | 1.6                  |             |             |              |               |               |
|          | Total thickness    | 2.2                  |             |             |              |               |               |
| 4 Layers | S8                 | 0.3                  | -12.81      | -28.89      | -18.59       | X band        | 1.2 GHz       |
|          | S1                 | 0.9                  |             |             |              |               |               |
|          | S7                 | 0.6                  |             |             |              |               |               |
|          | S6                 | 0.6                  |             |             |              |               |               |
|          | Total thickness    | 2.4                  |             |             |              |               |               |

**Table 4:** Samples optimized by cost function 1 with limitation of total thickness 3 mm and the number of layers from 1 to 5

| N layer  | Material in layers | Layer thickness (mm) | Min RL (dB) | Max RL (dB) | Mean RL (dB) | RL < -10 (dB) | RL < -20 (dB) |
|----------|--------------------|----------------------|-------------|-------------|--------------|---------------|---------------|
| 2 Layers | S2                 | 1.1                  | -11.13      | -21.93      | -16.86       | X band        | 0.7 GHz       |
|          | S7                 | 1.7                  |             |             |              |               |               |
|          | Total thickness    | 2.8                  |             |             |              |               |               |

**Table 5:** Samples optimized by cost function 2 with limitation of total thickness 2 mm and the number of layers from 1 to 5

| N layers | Material in layers | Layer thickness (mm) | Min RL (dB) | Max RL (dB) | Mean RL (dB) | RL < -10 (dB) | RL < -20 (dB) |
|----------|--------------------|----------------------|-------------|-------------|--------------|---------------|---------------|
| 1 Layer  | S9                 | 2                    | -5.13       | -26.1       | -15.14       | 3.2 GHz       | 1 GHz         |
|          | Total thickness    | 2                    |             |             |              |               |               |
| 2 Layers | S9                 | 1.3                  | -6.56       | -62.65      | -17.90       | 3.2 GHz       | 1.6 GHz       |
|          | S1                 | 0.7                  |             |             |              |               |               |
|          | Total thickness    | 2                    |             |             |              |               |               |

**Table 6:** Samples optimized by cost function 2 with limitation of total thickness 2.5 mm and the number of layers from 1 to 5

| N layers | Material in layers | Layer thickness (mm) | Min RL (dB) | Max RL (dB) | Mean RL (dB) | RL < -10 (dB) | RL < -20 (dB) |
|----------|--------------------|----------------------|-------------|-------------|--------------|---------------|---------------|
| 1 Layer  | S5                 | 2.5                  | -6.19       | -38.55      | -16.22       | 3.5 GHz       | 1.3 GHz       |
|          | Total thickness    | 2.5                  |             |             |              |               |               |
| 2 Layers | S7                 | 0.7                  | -7.59       | -62.82      | -20.54       | 3.4 GHz       | 1.8 GHz       |
|          | S1                 | 1.8                  |             |             |              |               |               |
|          | Total thickness    | 2.5                  |             |             |              |               |               |
| 4 Layers | S7                 | 0.5                  | -7.44       | -54.23      | -21.66       | 3.2 GHz       | 1.6 GHz       |
|          | S4                 | 0.4                  |             |             |              |               |               |
|          | S1                 | 1.4                  |             |             |              |               |               |
|          | S7                 | 0.2                  |             |             |              |               |               |
|          | Total thickness    | 2.5                  |             |             |              |               |               |

At each step with this cost function, the lowest RL value for each member in the entire frequency range is recorded. The best absorption occurs when cost function 1 is maximized. The maximum value recorded in cost1 is stored in p-best, the maximum value in each group is stored in l-best, and the maximum value in all groups is stored in g-best. According to the results recorded in p-best and l-best, the members move toward the target points with the acceleration and speed described previously, and new points are made. The new points are re-evaluated according to the cost function. Therefore, p-best, l-best, and g-best are updated. This iteration continues

until the points converge toward g-best. Cost function 2 (to achieve the maximum average RL) was calculated as follows:

$$\text{cost2}(n_1, t_1, n_2, t_2, \dots) = \frac{\sum_i |F(f_i)|}{\sum_i 1}. \quad (18)$$

In cost function 2, the average RL at all frequencies is calculated. Any member whose cost2 is higher has a higher value. Like cost function 1, the maximum value recorded in cost2 is stored in p-best, the maximum value in each group is stored in l-best, and the maximum value in all groups is stored in g-best. The rest of the steps will be repeated like cost1.

**Table 7:** Samples optimized by cost function 2 with limitation of total thickness 3 mm and the number of layers from 1 to 5

| N layers | Material in layers | Layer thickness (mm) | Min RL (dB) | Max RL (dB) | Mean RL (dB) | RL < -10 (dB) | RL < -20 (dB) |
|----------|--------------------|----------------------|-------------|-------------|--------------|---------------|---------------|
| 2 Layers | S2                 | 1.5                  | -8.41       | -40.35      | -21.03       | 4 GHz         | 2 GHz         |
|          | S7                 | 1.3                  |             |             |              |               |               |
|          | Total thickness    | 2.8                  |             |             |              |               |               |
| 3 Layers | S2                 | 0.8                  | -6.80       | -55.18      | -22.1        | 3.5 GHz       | 1.7 GHz       |
|          | S7                 | 0.6                  |             |             |              |               |               |
|          | S3                 | 1.2                  |             |             |              |               |               |
|          | Total thickness    | 2.6                  |             |             |              |               |               |

**Table 8:** Comparison of the microwave-absorbing properties of different related composites or structures reported in the literature

| Materials                                                                            | Matrix | d (mm) | Min RL (dB) | Bandwidth (GHz) (<-10 dB) | Ref.      |
|--------------------------------------------------------------------------------------|--------|--------|-------------|---------------------------|-----------|
| TiO <sub>2</sub> /RGO                                                                | Epoxy  | 2      | -62.65      | 3.2                       | This work |
| TiO <sub>2</sub> /RGO                                                                | Epoxy  | 2.5    | -62.82      | 3.4                       | This work |
| GO-CNTs/epoxy foam                                                                   | Epoxy  | 3      | -41.5       | 7.1                       | [62]      |
| Carbon black and Ni <sub>0.6</sub> Zn <sub>0.4</sub> Fe <sub>2</sub> O <sub>4</sub>  | Epoxy  | 2      | -24         | 4.8                       | [63]      |
| MXene/Fe <sub>3</sub> O <sub>4</sub>                                                 | Wax    | 2.6    | -50.3       | 1.9                       | [64]      |
| ZnO/RGO                                                                              | —      | 3.2    | -67.13      | 7.44                      | [65]      |
| [CaTiO <sub>3</sub> /ZnFe <sub>2</sub> O <sub>4</sub> ]@C                            | Epoxy  | 2      | -22         | 7.2                       | [66]      |
| Fe <sub>3</sub> O <sub>4</sub> /carbon fiber and Fe <sub>3</sub> O <sub>4</sub> /RGO | Epoxy  | 2      | -52         | 4                         | [67]      |

The cost function, total thickness, and the number of layers are given to the software as design limitations in each optimization. The number of absorption layers is chosen between 1 and 5. Also, the maximum total thickness was determined to be 2, 2.5, and 3 mm. Furthermore, the minimum thickness changes were considered to be 0.1 mm. The four components of permittivity and permeability of the samples are provided to the software as primary data. The cost functions 1 and 2 are used in optimization, as shown in Figure 9. Also, the total absorber thickness is considered to be between 2 and 3 mm. Furthermore, the number of layers is assumed to be between 1 and 5. Used materials and their thickness in optimum multilayer absorbers are listed in Tables 2–7; the arrangement of the layers from top to bottom indicates the order of the layers from the air side to the metal side.

Based on the optimization results, in a single-layer absorber design produced by cost1, sample S7 with a thickness of 2.2 mm has a maximum bandwidth of more than –8.07 dB RL compared to other specimens. For cost2, the maximum average RL equals –16.22 dB for sample S5 with a thickness of 2.5 mm. In our study, increasing the thickness did not improve absorption properties for the single-layer absorber design. In some multilayer absorber design processes, enhancing the number of layers improves absorber's performance. It is shown in Table 3 that when the number of layers increases from 1 to 4, the performance of the designed absorber shows more improvement. Nevertheless, enhancing layer numbers does not improve absorption characteristics in most multilayer absorber design processes. In these cases, the presented optimization software calculates the thickness of one or more layers equal to zero values, as shown in Table 5. This table shows that when cost function 2 is used for optimizing multilayer absorber design with a maximum thickness of 2 mm limitation, a two-layer absorber with a thickness of 2 mm has the best performance, and introduced software automatically considers zero values for the thickness of other absorber layers. Also, the optimized multilayer absorbers, designed by developed software based on increasing the layer numbers and enhancing total thickness limitation, are stated in Tables 2–7. As indicated in Table 3, a 4-layer absorber with a thickness of 2.4 mm, which is designed based on cost function 1, has the best RL bandwidth of less than –12.81 dB compared to other introduced samples. Also, a 3-layer absorber with a thickness of 2.6 mm, which is designed based on cost function 2, has the best result of average RL of about –22.1 dB compared to other specimens, as shown in Table 7.

The material type and layer thickness of multilayer absorbers in optimization with the cost function 2 and the total thickness limitation of 2, 2.5, and 3 mm are summarized in Tables 5–7, respectively.

A comparison of the absorption performance of the synthesized nanocomposites with the reported materials is summarized in Table 8. According to reports, a superior ability to absorb microwave was observed compared with other reports, and the effective absorption was broadened to the whole X band.

## 4 Conclusion

In this research, TiO<sub>2</sub>/RGO/epoxy nanocomposites with a different weight ratio were fabricated using a simple method of annealing followed by magnetic stirring. The SEM images of the nanocomposite exhibited good exfoliation and dispersion of TiO<sub>2</sub>/RGO as nanofiller in the epoxy resin. Investigation of the EM properties shows that the annealing process improves the graphitization and conductivity of RGO, increasing the real and imaginary parts of permittivity. The measurement of the EM properties of the prototypes shows small magnetic losses; then, the electric loss is the dominant property for designing of multilayer absorbers.

The optimization algorithm shows that multilayer absorbers perform better than single-layer absorbers with a certain thickness. With cost function 1, the performance of a single layer with a thickness of 2.2 mm, the minimum loss of –8.07 dB reached –12.81 dB in four layers with a thickness of 2.4 mm. With cost function 2, the single-layer absorption with a thickness of 2.5 mm, the average RL of –16.22 dB reached –22.1 dB in three layers with a thickness of 2.6 mm. Therefore, increasing the total thickness and number of layers is effective up to a range.

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