Crystal structure engineering of GdFeO3/Mxene composites with excellent electromagnetic wave absorption: role of phase transition and high polarizability

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Research Article

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Abstract

The development of new materials that can absorb electromagnetic waves (EMW) is needed to address the problem of signal interference and crosstalk. In this study, a new composite material consisting of MXene and GdFeO$_3$ nanoparticles has been synthesized using crystal structure engineering to improve EMW attenuation performance between 2 and 18 GHz. The GFO nanoparticles, with a size of 30–40 nm, are evenly distributed on the surface of the MXene layers. The XRD and Raman spectra of the composite material show different phases of GdFeO$_3$, which have different crystal symmetries and coordination states. The XPS and EPR measurements indicate the coexistence of various valence states of Fe, which leads to oxygen vacancies in the lattice. The addition of MXene greatly increased the specific surface area and dielectric properties of the composite material. Due to the improved polarization and phase transition behavior, the P-E loop, DM constant, and attenuation constant were significantly enhanced. The combination of good ferroelectric GdFeO$_3$ and disordered crystal phase into the multilayered MXene matrix resulted in an enhancement of conductive and magnetic losses. Experimental results demonstrated that the Pbnm GdFeO$_3$/MXene composites exhibited excellent EMW absorption performance. At a thickness of 4 mm, the minimum reflection loss was −61.5 dB, and a maximum effective absorption bandwidth of 8.62 GHz was achieved at 10.8 GHz due to the good dielectric, magnetic, and multiple reflections contributing to superior EMW absorption performance with a broad band.

1. Introduction

Electromagnetic wave absorption (EWA) materials have garnered significant attention due to their potential applications in various fields such as radar stealth technology, communication, and electronic warfare [1]. However, current EMA materials often suffer from limitations such as narrow bandwidth, low absorption capacity, and poor thermal stability [2]. The mechanism of EM absorption is based mainly on the electromagnetic wave-matter interaction which consists of dielectric loss and magnetic loss [3, 4]. When the EMA passes through a material, the electric field of the wave causes the electrons in the material to oscillate, resulting in energy dissipation (dielectric loss), while the magnetic field of the wave causes the magnetic domains in the material to rotate, resulting in energy dissipation, particularly in the lower frequency ranges where magnetic fields play a more dominant role. (magnetic loss) [5–12]. Therefore, the dielectric constant and magnetic property are crucial for EMA.

To overcome these limitations, nano-composites are appealing to realize the broadband absorption, particularly through combining dielectric loss-type nanomaterials, such as MXene [13–16] with magnetic loss-type nanomaterials such as Fe$_3$O$_4$ [17]. Much promising impedance match and broadband absorbing capacity have been achieved due to the enhanced interfacial polarization loss capability and the synergistic effects coupling dielectric and magnetic losses [18–20].

MXene is a two-dimensional transition metal carbide or nitride with promising electrical conductivity, high specific surface area, and tunable surface chemistry. The layer structure and the combination of different
elements in MXene enables the formation of a conductive network, leading to improved conductivity and electromagnetic wave absorption performance [14]. The high surface area is one of the most attractive properties of MXene which provides a large contact area between the material and EM waves, resulting in efficient absorption [20–23]. MXene has a high conductivity and low dielectric constant, which means it can absorb a wide range of EM waves, from radio waves to microwave and even terahertz waves.

Currently, the electromagnetic wave absorption performance of MXene materials has been systematically studied. Based on its two-dimensional morphology and surface functional groups, MXene/ nano-magnetic composites were synthesized to enhance its absorption performance. For example, MXene/Ni-CNTs [8], MXene-rGO/CoNi [22], MXene/Fe$_3$O$_4$ [23], MXene/CoFe$_2$O$_4$ [24], MXene/FeCo composites [25]. These composites exhibited a decreased minimum reflection loss (RL) of -49.38 dB at 14.2 GHz [23], -38.9 dB at 4.82 GHz [24], -47 dB at 12.8 GHz [25] with an increased EMA performance, respectively. The results indicated that the incorporation of magnetic component with MXene is an efficient method to obtaining good EMA property.

Despite the promising results achieved in previous studies, there are still some limitations that need to be addressed. For example, the high electrical conductivity of MXene induces the impedance mismatch between composites and air, yielding much reflection and transmission, and reducing the absorption of electromagnetic wave. [26]. The magnetic susceptibility loss, the conductivity and the stability of the composites also need to be optimized to achieve efficient EW absorption.

To address these issues, GdFeO$_3$ (GFO) was proposed to composites with MXene for the EMA application in this study. By virtue of the co-existence of multi-valences and multi-coordination, transition metal GdFeO$_3$ is a complex magnetic oxide material that exhibits various types of magnetic ordering and different crystal phases at different temperatures [27]. Firstly, GdFeO$_3$ has good magnetic properties, such as high magnetic permeability, this makes GFO a promising material for EMA. The strong magnetic properties of GdFeO$_3$ are attributed to the presence of Gd and Fe ions, which have unpaired electrons that produce magnetic moments. The magnetism of GdFeO$_3$ can be further enhanced by the Gd $\rightarrow$ Fe magnetic moments exchange interaction [28–30].

Very recently, literatures reported that GFO and GFO- based composites showed highly efficient EMA performance due to high dielectric / magnetic loss capacity originated from the metallic feature and the d orbital splitting of Fe, respectively [27–30], such as GdFeO$_3$/graphene aerogels, GdFeO$_3$/Fe$_3$O$_4$@C, and GdFeO$_3$/Fe$_3$O$_4$@MWCNTs etc.

Secondly, GdFeO$_3$ commonly undergoes a structural phase transition from an orthorhombic phase ($Pbnm$ space group) to a rhombohedral phase ($R3c$ space group) upon increasing temperature or pressure [31]. The orthorhombic phase is characterized by a reduction in the symmetry of the crystal structure compared to the rhombohedral phase. The transition is driven by the competition between the exchange interaction and the crystal field interaction in GdFeO$_3$, which is sensitive to the crystal symmetry [27]. In GFO, the A-site polyhedron GdO$_{12}$ expands more rapidly versus temperature than the B-site and FeO$_6$ sites
[32], thus stabilizing the $R3c$ phase at higher temperature. The distortion of crystal structure and the tilting of the FeO$_6$ octahedra break the cubic symmetry and increase the polarizability of structure. Importantly, the phase transition in GdFeO$_3$ is always associated with a change in the electric, polarizability and magnetic ordering [28, 29]. For example, the P-E curves of GdFeO$_3$ exhibit significant changes across the structural phase transition [30]. In the rhombohedral phase, the P-E curve is linear and exhibits weak ferroelectric behavior. However, in the orthorhombic phase, the P-E curve shows a more pronounced hysteresis loop with a higher polarization value, indicating stronger ferroelectric behavior. These studies demonstrate the potential of the phase transition in GdFeO$_3$ to enhance its EMA properties, highlighting the importance of understanding the underlying physics and chemistry of this material for potential applications in electromagnetic absorption and related technologies. However, the phase transition mechanism is not fully understood, especially related to the EMA property of GFO-composites.

In this study, for the first time a new MXene/GFO composites was synthesized, the EMA performance were investigated in terms of the phase transition, polarizability and magnetic ordering of the composites.

2. Experiments

2.1 Synthesis of GFO/MXene composites

GdFeO$_3$ nanoparticles are synthesized via a sol-gel method. 15mmol gadolinium nitrate hexahydrate, 15mmol iron (III) nitrate nonahydrate, dissolved in equimolar acetic acid and ethylene glycol (40 mL) and magnetically stirred for 90 minutes at 40°C. The mixture is stirred at 80°C for 4 hrs until a homogeneous gel was formed, then the gel was dried at 120°C for 8 hrs. Then heated the resultant product at 600°C and 900°C (with 6 hours’ subsequent ball-milling) for 4 hrs to form the GdFeO$_3$ nanoparticles in different crystal phases. The corresponding samples are labeled as GFO6 and GFO9, respectively.

The MXene was synthesized using a two-step etching process. 10 g Ti$_3$AlC$_2$ powder was first immersed in a mixture of hydrofluoric acid and hydrochloric acid (200 mL, 39%) at 40°C through magnetic stirring for up to 36 hrs to remove the aluminum layers, followed by a washing step with deionized water. The resulting Ti$_3$C$_2$T$_x$ was then immersed in an ammonium fluoride solution to remove the surface terminations and obtain Ti$_3$C$_2$.

GFO/MXene composites was synthesized through mixing the GFO nanoparticles and Ti$_3$C$_2$ (GFO: Ti$_3$C$_2$ = 1:1 mol:mol) in acetic acid and ethylene glycol in a ratio of 1:1 (0.01M) and stirring/sonicated for 10 min at room temperature. The prepared solution was stirred and sonicated for 2 hrs at 80°C. Then rinsed the final product for several times with deionized water and was put in a drying oven at 60°C for 3 hrs. Figure 1 shows the synthesis scheme of GFO/MXene composites.

2.2 Characterization
X-ray diffractometer (XRD, Bruker D8) was used to confirm the crystal structure and phase purities of samples with a Cu$_{K\alpha}$ radiation source and angle range 10-70°C. Using XRD data, Rietveld refinements were conducted to calculate the lattice parameters. Raman spectra was recorded to analyze the vibrational modes of samples using 633 nm laser. Surface and morphology of composites were investigated using Transmittance electron microscopy (TEM, FEI, Tecnai G20) with Energy Dispersive X-Ray Analysis (EDX) for compositional analysis. The Binding energies of elements in the composites were checked using X-ray photoelectron spectroscopy (XPS, AXIS Supra). The dielectric parameters were measured using HITESTERLCR meter. The polarization versus electric field hysteresis loop was traced. The microwave absorption was test through mixing samples with paraffin wax (Sample: Paraffin = 7:3) in a coaxial ring (inner/outer diameter of 3/7 mm). The electromagnetic parameters of the material were measured by a vector network analyzer (VAN; Agilent, N5234A) with working frequency of 2 ~ 18 GHz (E5071C, Agilent). EMW absorption capability was evaluated with reflection loss (RL), which was calculated based on the transmission-line theory and metal back-panel model [33].

3. Results and discussion

3.1 Phase transition of GFO in composites

Figure 2 shows the XRD patterns of GFO/MXene and the Rietveld refinement. From Fig. 2a, the GFO synthesized at 600°C exhibited characteristic obvious orthorhombic perovskite crystal structure belonging to space group $Pbnm$ [34]. Particular, the diffraction peaks at $2\theta = 23.2°, 26.09°, 32.2°, 33.2°, 33.8°, 34.38°$ matched well with the (002), (111), (020), (112), (200) and (021) planes of orthorhombic structure ($Pbnm$ space group) [32]. The distinct diffraction pattern indicates the good crystallinity which is also in good agreement with the standard JCPDS 78–0451, and other previously published literatures [34–37]. No other satellite peaks were found, suggesting the purity of GFO.

The GFO9 treated at 900°C with ball-milling for 4 hours shows a different pattern in Fig. 2a, indicating the treatment induced the phase transition of GFO. Diffraction peaks at $2\theta = 24.5, 33.02, 35.1, 41.2, 50.03, 54.88°$ were indexed to be the (102), (104), (110), (113), (204) and (116) planes of rhombohedral cell ($R3c$) according to the PDF card 01-074-1900 [38]. On the other hand, the pattern is rather broader than GFO6 due to the relative smaller particle size. Based on the Debye-Scherrer formula, the size of GFO6 and GFO8 were estimated to be 26 and 22 nm, respectively.

Figure 2b shows the XRD patterns of GFO/MXene composites along with that of T$_3$AlC$_2$ and etched Mxene. Ti$_3$AlC$_2$ shows the strongest characteristic diffraction peak at $2\theta = 9.7, 19.1, 39°$ which was indexed to the (002), (004) and (008) crystal plane according to JCPDS 52–0875 [37]. After the etching process, 3 main peaks at $2\theta = 8.65, 18.2$ and 61.4° were observed corresponding to the (002), (004) and (100) planes of MXene, respectively. The (008) plane of T$_3$AlC$_2$ disappeared while the peak at $2\theta = 9.7°$ of (002) plane significantly shifted to 8.65°. This peak shift toward a lower angle shows an
increase in the interplanar distance, confirming the broken of Ti–Al metal bond, and the removing of the Al atom can increase the plane spacing \([39]\). At the same time, the great decreases in the peak intensity of MXene indicates the loss of crystalline nature of the MAX powder after aluminum etching \([34]\). Both the XRD patterns of GFO6/MXene and GFO9/MXene in Fig. 1b exhibit the mix of \(R3c/Pnma\) phases of GFO with the MXene. In order to verify the crystal phase of GFO in the composites, Rietveld refinement of XRD patterns of GFO9/MXene and GFO6/MXene are shown in Fig. 1c and 1d, respectively, the lattice parameters obtained are listed in Table 1. From Fig. 1c and 1d, the calculation curves matched well with the experimental data with very small differences.

From Table 1, the GFO in GFO6/MXene composite was ascertained to be orthorhombic perovskite with \(Pbnm\) space group, the lattice parameters are: \(a = 5.349\ \text{Å}, b = 5.611\ \text{Å}, c = 7.669\ \text{Å}\), and \(\alpha = \beta = \gamma = 90^\circ\) which are in good agreement with reported data \([40]\). The Rietveld refinement of XRD of two composites revealed that Fe\(^{3+}\) was coordinated with 6 O\(^{2-}\) in octahedral unit. However, the Fe-O angle is larger while Fe-O length of GFO9 is smaller than that of GFO6, ensuring a big distortion in octahedral FeO\(_6\) of \(Pbnm\) structure along the C axis. The Gd-O length is larger than that of Fe-O due to relative larger ionic radius of Gd, while both of them decreased for GFO9, indicating that the phase transition and tilting of FeO\(_6\) squeezed the Gd-O distance. The refinement results in Table 1 also indicate that the synthesis process of composites well remained the original crystal structure of GFO due to the relative lower temperature (80°C).
Table 1
Refined structural parameters of composites.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>GFO6/Mxene</th>
<th>GFO9/Mxene</th>
</tr>
</thead>
<tbody>
<tr>
<td>Crystal type</td>
<td>orthorhombic</td>
<td>rhombohedral</td>
</tr>
<tr>
<td>Space group</td>
<td>$Pbnm$</td>
<td>$R3c$</td>
</tr>
<tr>
<td>$a$(Å)</td>
<td>$5.729 \pm 0.003$</td>
<td>$5.4165 \pm 0.002$</td>
</tr>
<tr>
<td>$b$(Å)</td>
<td>$5.976 \pm 0.001$</td>
<td>$5.4165 \pm 0.002$</td>
</tr>
<tr>
<td>$c$(Å)</td>
<td>$7.669 \pm 0.003$</td>
<td>$13.345 \pm 0.005$</td>
</tr>
<tr>
<td>Angle</td>
<td>$\alpha = \beta = \gamma = 90^\circ$</td>
<td>$\alpha = \beta = 90^\circ, \gamma = 120^\circ$</td>
</tr>
<tr>
<td>$V$(Å)</td>
<td>229.76</td>
<td>223.64</td>
</tr>
<tr>
<td>Size (nm)</td>
<td>35.5</td>
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</tr>
<tr>
<td>Fe-O (Å)</td>
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<td>2.01</td>
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<tr>
<td>Fe-O-Fe ($\theta$)</td>
<td>109.5</td>
<td>134.2</td>
</tr>
<tr>
<td>Gd-O (Å)</td>
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<td>2.41</td>
</tr>
<tr>
<td>$R_p$ (%)</td>
<td>12.2</td>
<td>18.3</td>
</tr>
<tr>
<td>$R_{exp}$</td>
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<td>11.7</td>
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<tr>
<td>$\chi^2$</td>
<td>1.40</td>
<td>1.55</td>
</tr>
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</table>

Figure 3 shows the GFO with different crystal structures. From Fig. 3, the main differences between the $Pbnm$ and $R3c$ structures of GdFeO$_3$ are the unit cell shape and size, as well as the different position of the oxygen ions. As observed, the $Pbnm$ GFO crystal structure is organized in layers. The layers are composed of octahedral FeO$_6$ units that are connected by dodecahedral GdO$_8$ units. The FeO$_6$ units are tilted along the b-axis which produces the distorted structure. The oxygen ions form zig-zag chains along the b-axis and the FeO$_6$ octahedron are connected by sharing edges [33]. The bonds between the layers are weaker than the bonds within the layers, resulting in the layers being able to slide past one another. This sliding motion makes the $Pbnm$ phase more flexible and less prone to cracking than the $R3c$ phase. As comparison, the $R3c$ GFO has a rhombohedral unit cell which is organized in a three-dimensional lattice, with the Gd and Fe ions and oxygen atoms situated at the lattice points [32]. The oxygen atoms in octahedral FeO$_6$ form chains along the z directions and occupy tetrahedral voids in the lattice. They are slightly tilted along the z directions, forming a network of corner-sharing octahedral around the Gd and Fe cations. Fe and Gd ions occupy octahedral sites and are arranged in a distorted perovskite structure.

The Raman spectra are shown in Fig. 4a to evaluate the molecular vibrations in the composites and the phase transition of GFO. From Fig. 4a, the Raman spectra of composites are divided into peaks due to
GFO (yellow dashed lines) and Mxene (blue dashed lines) vibrations. The GFO generally exhibits 24 Raman-active vibrational modes ($7A_{1g}$, $7B_{1g}$, $5B_{2g}$ and $5B_{3g}$) [41]. In this study, GFO6 ($Pbnm$) shows four $A_g$ peaks at 105, 153, 245 and 507 cm$^{-1}$, respectively. In addition, two $B_{1g}$ at 245, 418 cm$^{-1}$ and two $B_{2g}$ peaks at 134 and 630 cm$^{-1}$. Among them, peaks below 200 cm$^{-1}$ are due to stretching vibrations of Gd-O bonds, while peaks between 200 and 350 cm$^{-1}$ are caused by the FeO$_6$ octahedral rotation [8], and the bending of O-Fe-O and symmetric stretching vibrational peaks of Fe-O in FeO$_6$ units locate at 350–500 cm$^{-1}$ and above 500 cm$^{-1}$, respectively. From the comparison of GFO6 and GFO9, the GFO6 shows much higher intensity in the range of 200 to 500 cm$^{-1}$, indicating a much stronger tilting and bending vibration of FeO$_6$ units. This is accordance with literatures [41]. On the contrary, the peak at 507 cm$^{-1}$ due to $A_g$ (3) vibration disappeared in $R3c$ crystal. And $R3c$ phase shows relative weaker FeO$_6$ rotation and bending modes. It is known that the Raman modes especially the $A_g$ (3) and $A_g$ (5) are sensitive to the changes of A site of perovskite [13]. Based on the Landau theory, significant wavenumber shifts stand for the large contribution from phase transition and highly asymmetry. And such shift is linearly proportional to the tilting/distortion degree of FeO$_6$. From Fig. 1a, the $A_g$ (5) of $R3c$ crystal phase was merged with $B_{2g}$ (5) while $A_g$ (3) disappeared. Therefore, these changes of Raman spectrum attest that octahedron tilting and structural distortion which agree with the XRD analysis. Such differences can be observed in the Raman spectra of GFO6/MXene and GFO9/MXene composites. The vibrational characters of $Pbnm$ and $R3c$ of GFO are well remained after being composited with Mxene.

The spectrum of MXene is shown in Fig. 1a. The very weak peaks at 167 cm$^{-1}$ is assigned to Al–Ti vibrations, indicating the successful removing of Al. The strong peak at 202, 367, 612 and 708 cm$^{-1}$ are due to the Ti–C bond vibrations. The whole Raman profile of MXene can be divided into 3 regions: The flake region below 202 cm$^{-1}$ is owe to the vibration of C with 2 Ti layers and surface groups ($A_{1g}$); The $T_x$ region between 250–500 cm$^{-1}$ ($E_{1g}$) represents the vibrations of the Ti atoms, and the carbon region 530–750 cm$^{-1}$ ($A_{1g}$) corresponds to both in- and out-of-plane vibrations of carbon atoms [42]. The deconvolution of MXene and composites were performed to see the position of Raman vibrational modes. Due to the band overlap of limited intensity detection, not all Raman modes were identified. It can be seen that in composites based on $Pbnm$ GFO, the MXene vibrational modes are stronger, while in the composites of $R3c$ GFO, they are relative lower, particular the vibration in the C region. This result suggests that $Pbnm$ GFO based composite exhibits strong interaction between two components. $A_{1g}$ (Ti, C, Tx: O) and $A_{1g}$ (C) vibrational modes of $R3c$ GFO/MXene at 202 and 750 cm$^{-1}$ are weakened due to the decrease of C atoms at higher temperature.

### 3.2 Morphology and chemical states of GFO/MXene composites

Figure 5 shows SEM and TEM images of samples with different magnifications. Figure 5a, 5b and 5c show closely packed layered structures of MXene, GFO6/ MXene and GFO9/MXene, respectively. From the enlarged cross-sectional images, these layers have demonstrated excellent preferential alignment...
along the surface, and the lamellar structures of MXene are flat, smooth with sharp edges. In addition, they are relatively complete which is helpful to the growth of GFO on their surfaces. From Fig. 5b1 and 5c1, the GFO nanoparticles are well dispersed (without agglomeration) and attached on the surface of MXene sheets, showing a hybrid formation. From Fig. 5, both low and high magnification images of the sample show a homogeneous morphology.

TEM images in Fig. 5a2 shows that layer feature with the characteristic spacing between the lattice fringes of \( d = 0.956 \) nm in the corresponding the (002) plane of MXene [39]. Similarly, the surface topography in Fig. 5b2 shows that a large number of GFO is loaded on MXene layer, where the measured spacing between lattice fringes of 0.345 nm which is indexed to the (111) crystal plane of GFO (\( Pbnm \)) [43]. The distribution of GFO (\( Pbnm \)) is plotted in the inset of Fig. 5b1. From the plot, the average size of GFO (\( Pbnm \)) is 30 nm which is slightly smaller than that of GFO (\( R3c \)) (38 nm) in the inset of Fig. 5c1. The lattice fringes in Fig. 5c2 is 0.297 nm which is corresponding to the (110) plane of GFO (\( R3c \)), confirming the rhombohedral crystal structure of GFO [34].

Figure 6 shows the EDX and BET analysis on the composites. EDX analysis on GFO6/MXene composite is shown in Fig. 6a and 6b. In the blank area, 4 elements of Ti, C, O and F are displayed while the analysis on nanoparticle area only shows O, Gd and Fe elements. According to the element atomic ratios, the main composition in blank is \( \text{Ti}_3\text{C}_2 \) while the nanoparticles are \( \text{GdFeO}_3 \). The tiny amount of O and F in the blank area was introduced during synthesis process because Ti atoms are easily oxidized and react with a large number of O atoms from the external environment.

The \( \text{N}_2 \) adsorption measurements of GFO/MXene and pristine MXene are shown in Fig. 5c-5e. All samples show type IV isotherms with obvious hysteresis loops, indicating their mesoporous voids between MXene nanosheets. The pristine Mxene gave BET surface areas of 18.484 m\(^2\)/g and little pore size, while MXene-GFO9 (Fig. 5c) and MXene-GFO6 (Fig. 5d) exhibit much increased specific surface area of 31.336 and 32.562 m\(^2\)/g, with pore size of 9.5 and 12.3 nm, respectively. These values reveal the domination of mesoporous with a pore size range of 2–14 nm of IV type isotherms. The higher surface area is helpful for electromagnetic wave absorption.

The overall elemental analysis of GFO6-MXene composites is shown in Fig. 7a which displays the Gd4d, C1s, Ti2p, O1s, Ti2s, Fe2p and Gd3d in the range of 100–1300 eV. The overall survey spectrum of GFO9-MXene did not show obvious difference in the binding energy with respect to the GFO6-MXene. However, the intensity of O1s of GFO9/MXene around 530 eV increased while, the C1s and F1s decreased on the contrary because the Ti atoms are easily reacted with the O and suppressed the functional groups with F and C. Figure 7b shows the core-energy level of Ti2p of GFO6-MXene. Two distinct peaks at 456.9 and 462.2 eV are classified as the Ti2p3/2 and Ti2p1/2, respectively [11]. From the deconvolution, several sub-peaks were observed, indicating the coexistence of Ti-C(2p3/2), Ti-O(2p3/2) and Ti-F(2p3/2) bonds at 454.83, 456.07, 457.15 eV and the Ti-C(2p1/2), Ti -O(2p1/2) and Ti-F(2p1/2) at 459.88, 461.84, and 463.24 eV respectively. In addition, one peak at 458.2 eV may be due to the existence valence of Ti\(^{4+}\) in
TiO$_2$ which was not detected in the pristine MXene. The fitted content of TiO$_2$ is 8.22% and 8.34% in GFO6/MXene and GFO9/MXene, respectively.

Figure 7- XPS spectra of overall (a), Ti2p (b), C1s (c), Gd4d(d), Fe2p(e) and O1s (f) of GFO/MXene.

Figure 7c shows the core level spectrum of C1s which was deconvoluted into 5 sub-peaks at 284.46, 286.05, 287.37, 289.16 and 290.76 eV corresponding to the C − Ti, C − C, O = C − OH, C − O, and C − F bonds, respectively. It can be seen that the C-Ti peak shows the strongest intensity, indicating it dominates the carbon species [44]. The C − C may origin from external polluted carbon while C-O and C = O are due to the adsorption of oxygen in the air on the surface of samples [39]. The C-F bonds indicated the hydrophilic functional groups (− F, -OH) on the surface of MXene, which is helpful for modify the dielectric constant for electromagnetic wave absorption. The C-O and C = O bonds indicate the successful combination of MXene with GFO.

Figure 7d and 7e show the Gd4d and Fe2p spectra of GFO/MXene composite. Due to spin–orbit interactions between the lower J (= L - S) and higher J (= L + S) energy components, Gd and Fe exhibit Gd4d$_{3/2}$, Gd4d$_{5/2}$ and Fe2p$_{1/2}$, Fe2p$_{3/2}$ peaks in the core level spectra. In Fig. 7d, due to the electrostatic interactions between the 4d hole and 4f$^7$ valence electrons, two final ionic states (9D and 7D) were formed and the Gd4d peaks were separated into asymmetric spin-orbit doublets Gd4d$_{5/2}$ and Gd4d$_{3/2}$ at 143.11 and 146.25 eV with a spin-orbit energy separation of 3.14 eV [45]. The line shape and the peak positions of the Gd4d in Fig. 7d are in good agreement with literatures [40], confirming the 3+ oxidation state of Gd. The deconvoluted sub-peaks demonstrate the spin–orbit coupling from the interaction of 4d and 4f valence electrons in this composite [45].

The XPS spectrum of Fe2p in Fig. 7e displays two peaks centered at 712.1 and 724.9 eV for Fe 2p$_{3/2}$ and Fe 2p$_{1/2}$, respectively with a spin orbit energy separation of 12.8 eV. In general, the Fe$^{3+}$ shows a typical characteristic of Fe2p$_{3/2}$ – Fe2p$_{1/2}$ separation energy of 12.9 eV [32]. Therefore, the value of 12.8 eV suggests there is probably other valence of Fe (i.e. Fe$^{2+}$). Another possible reason is the existence of spin–orbit coupling between the 2p core hole and the unpaired 3d electrons of the Fe. From Fig. 7e, one distinct satellite around 719 eV near Fe2p$_{3/2}$ is originated from the dynamic charge transfer between Fe$^{3+}$ → Fe$^{2+}$ which is common in other transition metal compounds [22]. Another relative weak satellite around 707 eV near Fe2p$_{1/2}$ peak reveals the existence of the Fe$^{2+}$ state even though only the Fe$^{3+}$ is present in the original GFO. The deconvolution on the Fe2p shows two sub-peaks in both Fe2p$_{3/2}$ and Fe2p$_{1/2}$ peaks, and they are ascribed to the Fe$^{2+}$2p$_{3/2}$, Fe$^{2+}$2p$_{1/2}$, Fe$^{3+}$2p$_{3/2}$, Fe$^{3+}$2p$_{1/2}$ components, respectively. These results indicate that the compositing with MXene reduced partial Fe$^{3+}$ into Fe$^{2+}$. According to the integrated area of deconvoluted peaks, the contents of Fe$^{3+}$ and Fe$^{2+}$ are calculated to be 73.5% and 26.5% in GFO6/MXene composite, which is very close to that of GFO9/MXene (73.6% and 26.4%).
It is known that surface oxygen vacancies play. The presence of Fe$^{2+}$-induced charge imbalance would result in surface oxygen vacancies which play significant role in the EMA process. The oxygen vacancies can be detected from the core level energy of O1s (Fig. 7f). From Fig. 7f, the O1s spectrum of GFO6/MXene shows two peaks at 533.1 and 531.2 eV. The deconvolution of this spectrum exhibits 4 sub-peaks. The sub-peaks at highest bonding energy 534 eV is due to the loosely bonded oxygen on the surface which corresponds to the dangling bond i.e. oxygen vacancy [34]. This is because all lanthanide oxide materials present hygroscopicity nature including the GFO. Such it is easy to absorb oxygen on its surface such as H$_2$O or carbonates. The low binding energy peaks at 532.8, 530.9 eV are corresponding to the lattice oxygen (O$^{2-}$) in structure of composites due to the two different crystallographic positions of O ion in orthorhombic GFO (Fe-O, Gd-O). The last sub-peak at lowest binding energy 529.2 eV is ascribed to the Ti-O bonds which was confirmed by the XPS analysis. These different types of O on the surface of composites can provide much active sites for absorbing the electromagnetic wave.

The oxygen vacancies were confirmed by the EPR analysis in Fig. 8. One significant strong asymmetric single EPR feature was detected at 160 mT for all samples with gyromagnetic (g) factor g = 4.3. This is the characteristic signal of Fe$^{3+}$ ions, indicating that the + 3 valence state is the dominant charge of Fe in samples. The signal at g = 4.3 shows the highest intensity for GFO9/MXene composites, also the intensity of GFO6/MXene at this value is much higher than that pristine GFO. This discover implies that the combination with MXene enhanced the spin-orbit coupling of Fe ions, which is very plausible for subsequent EMA performance. The EPR spectra in Fig. 8 also present another relative strong asymmetric signal at g = 2 with a similar tendency of intensity to that g = 4.3. The signal at g = 2 is corresponding to oxygen vacancies defect in the structure of composite [41] which was not detected in the pristine GFO. This result suggests that the defects were associated with the MXene, agreeing well with the XPS analysis. From the spectra, this signal becomes broader and more asymmetric at 360 mT which confirms the existence of oxygen vacancies on the surface of Ti$^{3+}$ layers [46]. Another wide and less intense signal around 500 mT (g = 1.613) is associated with Fe$^{2+}$ ions due to the reduction of MXene which is absent for pristine GFO. The intensity of this signal agrees with the Fe2p core level energy result. From Fig. 8, EPR intensity of this signal was very close to each other for $Pbnm$ and $R3c$ GFO phases. In general, the change in EPR intensity depends on the various factors, such as defect valence state, low to high spin configuration, and spin-lattice distortion [33]. In case of this study, the relative higher intensities of EPR signal of $Pbnm$ GFO at g = 4.3 and g = 2 is due to the oxygen vacancies (V$_o$) and spin-lattice defect formation through the charge compensation process. The slight deviation of the g-factor (with respect to that standard g values of unpaired electrons of Fe) verified the spin-orbit coupling. In addition, the broader line-width feature of $Pbnm$ GFO/MXene indicates stronger magnetic dipolar-dipolar interaction within the sample.

The asymmetric parameter magneto-crystalline anisotropy (P) can be calculated from the EPR spectra through $P = h_U/h_L$, referring to ratio of the maximum height of upper/lower peak above/below the baseline [47]. After calculation, the value of $Pbnm$ of GFO (2.26) at g = 4.3 is higher than other two (1.24 for $R3c$ GFO and 1.73 for GFO9/MXene), indicating a stronger magnetic interaction. This is mainly
because the Fe\(^{3+}\) has significant higher magneto-crystalline anisotropy (1.4cm\(^{-1}\)/ion) than that of Gd\(^{3+}\), while the bond length of Fe-O is smaller than that of Gd-O (Table 1), particularly in R3c GFO. Therefore, the short length and high magneto-crystalline anisotropy resulted in strong magnetization which is good to EMA applications.

### 3.3 Dielectric, magnetic and polarization study

The materials interact with the electromagnetic wave mainly through polarization (dielectric constant \(\varepsilon'\)) and magnetization (magnetic permeability \(\mu'\)) responses in materials [1]. The \(\varepsilon'\) represents the polarization intensity under the action of EMW and shows the ability to consume the EM energy in the form of electrical energy. Therefore, the \(\varepsilon'\) and dielectric loss are important factors for EMA samples. Figure 9a shows the \(\varepsilon'\) and dielectric loss (inset) of GFO/MXene composites. It is known that the pure phase MXene (Ti\(_3\)C\(_2\)) shows dielectric constant within 14.06 \(\sim\) 24.21 [3]. This is because MXene do not possess intrinsic polarization due to their metal-like nature, which makes them unable to support a significant internal electric field. Therefore, the \(\varepsilon'\) is comparatively low. However, ferroelectric GFO has asymmetrical arrangement of ions within the crystal lattice, and GFO possesses a spontaneous polarization which aligns in a particular direction under external electromagnetic field, leading to an increase in dipole moment and hence the dielectric constant. From Fig. 9a, the \(\varepsilon'\) of GFO6 (1000) and GFO9 (400) are much higher than that of MXene and the values are close to that in literatures (300 \(-\) 1200) [23]. In addition, we notice that the \(\varepsilon'\) of \(Pbnm\) GFO (GFO6) is higher than that of \(R3c\) (GFO9) due to the lower crystal symmetry of the \(Pbnm\) phase. The lower symmetry allows for more complex polar structures that can store more electrical charge, leading to a higher dielectric constant. On the contrary, \(R3c\) phase has fewer complex polar structures due to its relative higher symmetry, resulting in a lower dielectric constant. On the other hand, the orthorhombic structure of GFO has a larger unit cell, which allows for more atomic dipoles to align with the electric field, leading to a higher dielectric constant compared to the rhombohedral structures [33].

From Fig. 9a, the \(\varepsilon'\) of all samples decreases rapidly from lower to higher frequency region due to the rapid polarization occurring in samples. However, a clear high frequency-independent behavior of \(\varepsilon'\) was observed for all samples. This is because the electric dipoles do not comply with the field at high frequency, and then lag behind the field [6]. Similar to the \(\varepsilon'\), dielectric losses in Fig. 9b show dispersion behavior of GFO6 and GFO9. This is because the space charge of GFO cannot sustain at higher frequency and the values of dielectric loss gets diminished with external field. On the other hand, at higher frequency, domain wall rotation of GFO predominate their motion, resulting in low dielectric losses [11]. However, the GFO based composites show much stronger dielectric loss, particularly at higher frequency range due to the contribution of MXene. Firstly, MXene flakes have a large surface area and highly conductive nature, which leads to high interfacial polarization and electrical conductivity losses in the composite material. Secondly, the dielectric properties of GFO/MXene are highly sensitive to their surface termination, with commonly used terminations such as \(\sim-O\), \(\sim-OH\), and \(\sim-F\) exhibiting relatively high losses at higher frequencies. Thirdly, the presence of residual water molecules or other polar impurities can also contribute to high dielectric loss at higher frequencies in MXene-based composites [34]. Finally,
the morphology and alignment of MXene flakes in the composite structure can also affect the dielectric loss. For example, the GFO6/MXene with less ordered structures shows higher losses at higher frequencies than GFO9/MXene. The much higher dielectric loss indicates the complementary advantages of GFO and MXene which is very welcome for EMA application.

When a material is exposed to an electromagnetic wave, it will experience an oscillating electric field that can cause polarization within the material, and such polarization process consumed and absorbed the energy of EM wave. In general, the polarization behavior is dependent on the crystal structure and associated symmetry degree, different crystal phases possess different crystal symmetries, leading to variations in their polarization-electric field response. Figure 9c shows the polarization versus electric field performance. From Fig. 9c, both GFO6 (Pbnm) and GFO6/MXene have a steep slope in the P-E curve at low electric field strengths. This means that even small electric fields can induce significant polarization, allowing for greater absorption of the electromagnetic waves. Additionally, a high maximum polarization at high electric field strengths was observed which is beneficial for absorbing strong electromagnetic waves. In this study, the Pbnm and R3c crystal structures of GFO have different symmetry properties, which gave rise to different polarization-electric field (P-E) behaviors. From Fig. 9c, the R3c (GFO9) and R3c based composites show relative lower polarization than that of Pbnm GFO due to the lower crystal symmetry of Pbnm structure. This result agrees well with literatures in which the orthorhombic phase of GdFeO₃ also exhibited stronger polarization-electric properties than rhombohedral and cubic phases [45]. The much lower symmetry of orthorhombic Pbnm results in a larger number of active modes for ferroelectricity and a greater degree of distortion in the crystal lattice, allowing for a stronger P-E coupling under an applied electric field. On the contrary, R3c crystal structure has a more symmetric lattice and fewer active modes for ferroelectricity. As a result, under an applied electric field, the distortion in the R3c structure is lower, leading to a weaker P-E coupling. In addition, the active modes in Pbnm phase has a preferred to growth orientation while that of R3c tends to be more random in orientation. Thus the Pbnm phase presents a highly polarizable feature. That is why both Pbnm GFO and Pbnm GFO based composite show much stronger P-E performances in Fig. 9c.

The EMA performance of sample is also greatly affected by the magnetic permeability (µ’ or magnetic-dielectric MD) constant which describes the ability to store magnetic energy inside materials. Figure 9e-9h shows the MD constant of GFO and GFO based composites. Materials with a high µ’ can effectively attenuate the amplitude of electromagnetic waves as they pass through the materials. The µ’ is the ratio of the magnetic dipole moment to the magnetic field that causes it. When a magnetic field is applied to a material, the magnetic dipole moment of its atoms or molecules will align with the field. However, the degree of alignment and the resulting MD constant depend on the frequency. As can be seen from Fig. 9e-9h, at low frequencies, the magnetic moments of the atoms or molecules of all samples align with the magnetic field as ferromagnetic resonance, therefore, the MD constant is relatively high at this resonance frequency due to the strong alignment of the magnetic moments. However, at higher frequencies, the magnetic moments of the atoms or molecules no longer keep pace with the changing magnetic field and their alignment (ferromagnetic resonance) was disrupted, resulting in a significant decrease in the MD
constant. At very high frequencies, the magnetic moments no longer follow the magnetic field at all and become completely randomized. At this point, the MD constant approaches zero [22]. From Fig. 9e-9h, the MD constant of Pbnm GFO is slightly higher than that of R3c, but the compositing with MXene greatly enhanced the MD constant of samples. This is because GFO Pbnm has a high net magnetic moment orientation, causing more energy to be dissipated within GFO Pbnm during electromagnetic wave propagation, thereby contributing to an increase in attenuation constant in Fig. 9d. The attenuation constant $\alpha$ was obtained using Eq. (1):

$$\alpha = \frac{\omega}{c} \times \sqrt{\mu' \varepsilon'} \sqrt{\frac{1}{2} \left[ \tan \delta_c \tan \delta_m - 1 + \sqrt{(1 + \tan^2 \delta_c) (1 + \tan^2 \delta_m)} \right]}$$ (1)

On the other hand, the oxygen vacancies are fundamental important for attenuation constant because they create extra absorption sites during EMW passing through. From previous EPR and XPS O1s analysis, there are $V_0$ existed in Pbnm GFO. In addition, from the Fig. 9a, 9b and 9c, the higher dielectric constant, polarization and magnetic permeability of Pbnm GFO contributed to the higher attenuation constant of Pbnm GFO in Fig. 9d.

### 3.4 Reflection loss and EMA property

The EMA capability of material is closely associated with the minimum reflection loss ($R_{L_{min}}$), thickness of material, and the effective bandwidth [48]. The reflection loss can be obtained using Eqs. (2) and (3):

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

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

Where, the $f$, $d$, $c$, $Z_0$, and $Z_{in}$ refer to the frequency, thickness, light speed, free-space impedance, and the input impedance, respectively. Figure 10 shows the simulated reflection loss and 3D reflection loss plane and contour maps samples. The $R_{L_{min}}$, thickness, frequency, and effective bandwidth lists are compared, and presented in Table 1. The sharp peak observed in the reflection loss curve is known as a resonance peak or absorption peak, and it occurs at the frequency where the material strongly absorbs the incident electromagnetic wave. The location, shape and intensity of peak are dependent on dielectric constant, magnetic permeability, thickness, and structure. It can be seen from Fig. 10 that the maximum EMA of GFO6 and GFO9 appears at 7.2 and 9.5 GHz, with $R_{L_{min}}$ of -24.9 and -32.8 dB at thickness of 3, and 2.5 mm, respectively. The effective bandwidth (EWB) is 3.36 GHz at 2.5 mm and 3.11 GHz at 3 mm thickness. From Fig. 10a, the reflection peaks of GFO9 are broad while that of GFO6 are sharp and stronger. The effective bandwidth of EMA of GFO9 is also smaller than that of GFO6. From precious studies, Pbnm GFO possess larger $\varepsilon'$, polarization and $\mu'$. In general, a higher $\varepsilon'$ induces $R_{L_{min}}$ appear at lower frequencies due to the strong ability to store energy in an electric field. While a higher $\mu'$ causes the resonance peak to occur at higher frequencies [23]. The rhombohedral phase GFO in Fig. 10a exhibits a
magneto-electric resonance frequency around 9 GHz. The broad absorption peak occurs due to the random orientation of dipoles within the material, which leads to inefficient energy absorption at specific frequencies. On the contrary, the orthorhombic phase of GFO in Fig. 10b exhibits a sharp reflection loss peak at around 11 GHz due to the alignment of its electric and magnetic dipole moments along a particular axis. This alignment leads to the formation of an electric and magnetic polariton, which results in a strong and narrow resonance peak. From 3D reflection loss maps in Fig. 10a1 and Fig. 10b1, the GFO6 clearly exhibits a broader EMA bandwidth than GFO9, indicating strong absorption over a wide range of frequencies.

From Fig. 10c and 10d, after incorporation GFO with MXene, the composites show much enhanced EMA performance with $RL_{\text{min}}$ of -51.6 and -61.5 dB for GFO9/MXene and GFO6/MXene at 15.2 GHz (3 mm) and 10.08 GHz (4 mm), respectively. The EWB is 6.44 GHz at 3 mm and 8.62 GHz at 4 mm thickness. The greatly improvement on EMA property mainly due to the large surface area and highly conductivity of MXene. Based on the Debye theory \[43\], dielectric loss mainly originates the conduction loss and polarization loss \[44\]. The highly conductive MXene has layered structure which provides a conduction pathway for carrying and attenuating the EMW, at the same time, the electron polarization, dipole polarization, ion polarization on MXene surface contributed the high EMA of GFO6/MXene composite as well. From the maps in Fig. 10d1, the resonance peaks appear at both low and high frequencies due to the both higher ‘ and µ’ values of GFO6, resulting in a significant broadening of effective bandwidth of EMA.

The impedance matching contour plots are shown in Fig. 11. As addressed, the impedance matching between samples and air is critical for EMA performance. In the contour maps in Fig. 11, the colors represent different levels of impedance. The blue, red, and yellow colors in the plot typically represent regions of poor, fair, and good impedance matching, respectively. Thus, for a good impedance match, we would expect to see more yellow color in the plot. Besides the color, the shape of the impedance matching contour plot provide impedance matching characters. For example, a steeply sloping curve may indicate that the impedance matching is particularly sensitive to changes in frequency (not steady and constant matching), while a broad, shallow curve may suggest a constant and broad matching of impedance.

From Fig. 11a, the pristine GFO ($R3c$) shows a stripe from top to bottom with more red color, indicating that the impedance matching is not constant across the frequency range of interest. In addition, the narrow stripe reveals that the EMA bandwidth is narrow. However, the impedance matching of GFO6 in Fig. 11b is broader contour shape which exhibits more yellow area, however, the line still is steep which decreases sharply from top to bottom across the whole frequency, suggesting that the impedance matching is not stable and constant. This issue was significantly optimized in Fig. 11d and particularly in Fig. 11c, not only the line become flatter and broader, but also more and more yellow color present in the contour plot. From the indicator bar, the yellow color stands for the best impedance matching value of 1. Therefore, this result implies that the composites, particularly the GFO6/MXene reach the best impedance matching with air. The synergistic effect between Mxene and GFO improved the impedance matching of
the composite and make EMW easier to enter the material. In addition, the flat/shallow and broad curve also indicate a steady and broader-band impedance matching capabilities for EMA [48].

Table 2 lists the EMA values of work of this study and from literatures. From the comparison in Table 2, the values obtained in this study are superior. The mechanism of enhanced EMA property of GFO6/MXene composite is illustrated in Fig. 12. As addressed previously, GFO shows promising EMA properties due to its strong coupling between electric and magnetic degrees of freedom. The Pbnm and R3c phases of GFO exhibited different EMA properties due to differences in their dielectric constant, dipole polarization, magnetic loss, and crystal defects such as low symmetry lattice and highly distortion of FeO6 units. These factors provided more active sites and much stronger dipolar polarization, resulting in the higher EMA for Pbnm GFO. In addition, the combination of MXene with Pbnm GFO take advantages of the high surface area, layered structure and broad-band absorption of MXene, giving rise to greatly enhanced EMA behavior. Figure 12 shows various contributions to the EMA of Pbnm GFO/MXene composites.

From Fig. 12, the excellent EMA property has various contributions and 5 main sources are displayed.
The first contribution is the strong Gd$^{3+} \rightarrow$ Fe$^{3+}$ and Fe$^{3+} \rightarrow$ Fe$^{3+}$ dipole coupling between electric and magnetic degrees of freedom \([46−52]\). From Fig. 9e–9h, the GFO6/MXene owns high magnetic permeability constant $\mu'$ and exhibits stronger ferromagnetic ordering than the R3c phase. In addition, the lower crystal symmetry in the $Pbnm$ phase allows for greater magnetic anisotropy, which enhances the alignment of the magnetic moments along a preferred direction. The vibration of the dipoles within GFO/MXene under an applied electric field, leading to energy dissipation in the form of heat. All of these factors contributed to the much high attenuation loss of EMA in GFO6/MXene (Fig. 9d).

The second main contribution to EMA of GFO6/MXene is the high $\varepsilon'$ and dielectric loss as shown in Fig. 9a and 9b. When EMW passes through the sample, the movement, collision and oscillation of electrons inside sample exhausted a large number of EM energy in electric form. is owe to the large dielectric constant which yield high dielectric loss which transferred much EM energy. The third source is the interfacial polarization loss or oxygen defects. From previous FT-IR, XPS, EPR analysis, the existence of functional groups, oxygen vacancies, dislocations, grain boundaries and lattice defects on the surface of MXene can form interfacial polarization and enhance the polarization loss. These defects can act as scattering centers for electromagnetic waves, leading to a consumption of EM energy. The formation of hetero-structured interfaces between GFO and MXene layer is conducive to improving the interface polarization, thus increasing the polarization loss; On the other hand, when the EMW touch the interface, electrons on the surface was polarized near the surface and such polarization process consumed a lot of EM energy as well. This can be confirmed from the high P-E loop in Fig. 9c. Other sources such as the Eddy current loss, multi-reflection loss, thermal loss and good impedance matching all contributed to the high EMA performance of GFO6/MXene composite.

4. Conclusion

In this study, two different crystal phases of GdFeO$_3$ were composited with MXene to obtain novel composites with high EMA property and broad bandwidth. The synergic effect of multilayered interfaces with both dielectric and magnetic elements enhanced the wave absorption effect. The $Pbnm$ GFO/Mxene composite obtained at 600°C showed highly asymmetry and strong polarization, exhibiting more active sites for electron and magnetic dipolar coupling which jointly improved the impedance matching and MD constant. The minimum RL reached $-61.5$ dB (with a frequency of 10.08 GHz, and 4 mm thickness), and maximum EAB of 8.62 GHz. The R3c GFO/MXene obtained at 800°C also exhibited excellent EMA performance with RL$\text{min} - - 51.6$ dB (with a frequency of 15.2 GHz, and 3 mm thickness), and maximum EAB of 6.44 GHz. In addition, the composites of GFO/MXene is a novel composites and for the first time for EMA application to provide excellent EMA property and broad bandwidth which is promising for EMA applications such as the defense, environments, medical and energy fields.

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**Figures**

![Diagram of MXene fabrication process and its dielectric properties](image)

**Figure 1**
Synthesis scheme of GFO/MXene composites

Figure 2

XRD patterns of GFO (a), GFO/MXene composites(b) and Rietveld refinement (c and d).
Figure 3

Crystal structures of GFO in $Pbnm$ and $R3c$ phases
Figure 4

Raman spectra (a) and deconvolution of samples.
Figure 5

SEM and TEM images of Mxene (a, a1 and a2), GFO($Pbnm$)/Mxene (b, b1 and b2), and GFO ($R3c$)/Mxene (c, c1 and c2). Insets: GFO distribution and SAED fingers.
Figure 6

EDX analysis (a, b) and BET $N_2$ adsorption–desorption isotherms and pore sizes (insets) of composites and Mxene.
Figure 7

XPS spectra of overall (a), Ti2p (b), C1s (c), Gd4d(d), Fe2p(e) and O1s (f) of GFO/MXene.
Figure 8

X-band EPR spectra of samples.
Figure 9

Dielectric properties (a, b), P-E loop (c), DM constant (e-h) and attenuation loss (d) of samples.
Figure 10

Simulated (a-d) and corresponding 3D reflection loss plot (a1-d1) of samples
Figure 11

Impedance matching contour plots of GFO9/GFO6 (a, b) and GFO9/MXene, GFO6/MXene composites (c, d)
Figure 12

EMA diagram of GFO6/MXene composite