Intercalation of Nanoscale Multiferroic Spacers between the Two-Dimensional Interlayers of MXene

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ABSTRACT: We report a facile synthesis of MXene/bismuth ferrite (Ti$_3$C$_2$T$_x$ @ BiFeO$_3$) nanocomposites using hydrazine hydrate induced with the hydrothermal method. Preparation of the composite with hydrazine hydrate prevents the formation of Bi$_{25}$FeO$_{39}$ inside the matrix. The 25 wt % loading of BiFeO$_3$ acts as spacers for the multilayer restacking of MXene flakes by retaining the (002) plane. This promotes large dc conductivity with interfacial and defect dipole polarization mechanisms inside the composite material. The thinner M-H loop also indicates the minimal magnetic loss inside the composites. The facile synthesis strategy provides outstanding properties in Ti$_3$C$_2$T$_x$ @ BiFeO$_3$ composites as two-dimensional multiferroic materials.

INTRODUCTION

The large-scale production of electric vehicles and wearable electronic devices is primarily integrated with the advanced technologies related to the efficient storage of electrical energies. The higher cyclability and energy density make the electrochemical capacitors (ECs) reliable for such applications. The charge storage mechanism and electrical properties of the electrode materials intrinsically determine the overall electrochemical properties of ECs. Some materials with superior mechanical and electrical properties and the surface area largely control the EC’s intrinsic electrode properties. MXenes, one of the evolving 2D classes of materials offers all these electrochemical properties for the better performance of the EC. The synthesis of MXenes is performed through selective etching techniques from the MAX phase (M$_{n+1}$AX$_n$). Here, M is a transitional metal ion such as Ti, A is a group XIII element such as Al, X is a carbonitride/carbide/nitride, and $n = 1, 2$, or 3. After the selective etching of the A layer, the MXene compositions are generally presented as M$_{n+1}$X$_n$T$_x$, where T$_x$ is the surface functional group such as F, O, OH, etc.

Very soon after discovering MXene, it grows to nearly 30 different compositions of transitional metal ions’ carbonitrides/carbides/nitrides. They mainly find applications in various fields of catalysis, water purification, sensors, optoelectronics, EMI shielding, and energy storage. Among all these fields, the MXene family is mainly popular for energy storage applications. Other than the properties such as superior mechanical and electrical properties and surface area, it also has functional groups and high conductivities. So far, the use of MXene as an energy storage material for ECs reported higher capacitance.

Generally, the conventional double-layer capacitors build electrical charge at the interface of the electrolytes and electrode materials. This process hindered the limit by which we could enhance the energy density of the capacitors. But the etching of the MAX powder leads to the formation of multilayers in MXene. Hence, integrating room-temperature multiferroics such as bismuth ferrite (BFO) as a spacer between the 2D MXene interlayers leads to a comprehensive capacitive charge storing mechanism. In this mechanism, the presence of the dielectric bismuth ferrite material enhances the interaction of ions between the MXene layers, known as capacitive intercalations. But so far, reported integration of BFO into Ti$_3$C$_2$T$_x$ MXene used solvothermal techniques with heavy agglomerations. Hence, the effort to optimize the synthesis technique for Ti$_3$C$_2$T$_x$ @ BiFeO$_3$ using the facile hydrothermal method is still lagging.

Herein, we synthesized a Ti$_3$C$_2$T$_x$ @ BiFeO$_3$ composite with the hydrothermal technique. We chose hydrazine hydrate as a reducing agent because it helps intercalate the BiFeO$_3$ phase between the 2D layers of Ti$_3$C$_2$T$_x$. We further discuss the
Impact of replacing hydrazine hydrate with ammonia on the fabrication of the ferrite phase between the interlayers.

**EXPERIMENTS**

**Materials.** Ti₃AlC₂ (titanium aluminum carbide) was purchased from Nanochemazone. Bi(NO₃)₃·5H₂O (bismuth nitrate pentahydrate), Fe(NO₃)₃·9H₂O (iron nitrate nanohydrate), KOH (potassium hydroxide), NH₃·H₂O (hydrazine hydrate), NH₄·H₂O (ammonia), HNO₃ (nitric acid), HF (hydrofluoric acid), and C₂H₅OH (ethanol) were supplied by Alpha Chemika. No further purification methods were used after that.

**Synthesis of MXene Ti₃C₂Tₓ.** Initially, 4 g of MAX phase was slowly mixed with 200 mL of HF and kept in a polypyrrole container in a fume hood, avoiding any exothermic heating. After that, the mixture was kept at a temperature of 35°C for 72 h with gentle stirring with a polypyrrole rod at an interval of 12 h. The black powders were rinsed several times with demineralized water and finally washed with C₂H₅OH until they reached pH 6 using a centrifuge machine.

**Synthesis of the Composites.** In situ preparation of the Ti₃C₂Tₓ@BiFeO₃ composite was carried out by the hydrothermal technique. In general, 1.0244 g of Bi(NO₃)₃·5H₂O and 0.8533 g of Fe(NO₃)₃·9H₂O were dispersed into 20 mL of demineralized water with 4 M KOH until the pH reached 11. Both the mixtures were transferred into a Teflon container slowly with continuous stirring. Finally, the hydrothermal reaction was carried out for 8 h at 220°C. Thereafter, the brown powders were cleaned with demineralized water and vacuum-desiccated for 12 h at a temperature of 60°C to obtain the Ti₃C₂Tₓ@BiFeO₃ composite. The same procedure was also followed for the Ti₃C₂Tₓ@Bi₂5FeO₃₉ composite; only N₂H₄·H₂O was replaced by NH₃·H₂O in this case. For both cases, stoichiometric formulations were carried out to reach 25 wt % of ferrite phase loading within the MXene structure.

**Composite Characterizations.** X-ray diffraction (XRD) was performed with a Bruker D2 model diffractometer (CuKα, 1.5418 Å). The FTIR spectra were obtained from a Bruker Alpha Cube FTIR spectrophotometer (PerkinElmer 2000) up to 4000 cm⁻¹. The morphology of the composite was determined with a Hitachi SU1510 scanning electron microscope. In situ preparation of the MAX phase between the interlayers.

**RESULTS AND DISCUSSION**

**XRD and Reaction Mechanism.** The powder XRD profile of pure BiFeO₃, the composite Ti₃C₂Tₓ@BiFeO₃, the composite Ti₃AlC₂@Bi₂5FeO₃₉, MXene Ti₃C₂Tₓ, and pure Ti₃AlC₂ is shown in Figure 2. The HF etching completely removes the (104) plane reflection of Ti₃AlC₂ due to aluminum layers, as it completely disappears from the XRD profiles of Ti₃C₂Tₓ. It also resulted in a broadening and leftward shift from 9.5° to 8.9° of the (002) plane peak due to etching. The d spacing calculation with Bragg’s law indicates the available spacing of 0.93 nm between the 2D layers of MXene. The lattice constant a = 0.56 nm of BiFeO₃ corresponds to JCPDS 020-0169 with a rhombohedral R3c space group of symmetries. The retention of the (002) peak for Ti₃C₂Tₓ@BiFeO₃ indicates the successful interfacial interactions between the two materials. The higher intense peaks in the composite belong to BiFeO₃ in addition to Ti₃C₂Tₓ peaks. The characteristics peaks related to the BiFeO₃ phase appeared at (012), (104), (202), (024), (122), (208), (217), and (312). We may propose that hydrazine hydrate helps transfer the metal nitrates into metal hydroxide and oxide ions, which are then combined to form the BiFeO₃ phase:

\[
\text{Fe}^{3+} + 6\text{Bi}^{3+} + 16\text{OH}^- \rightarrow \text{Fe(OH)}_4^+ + 6\text{BiO}_6^{2+} + 6\text{H}_2\text{O} \tag{1}
\]

\[
6\text{Fe(OH)}_4^+ + 6\text{BiO}_6^{2+} \rightarrow 6\text{BiFeO}_3 + 12\text{H}_2\text{O} \tag{2}
\]
On the contrary, the use of another reducing agent like ammonia may initiate a different mechanism for the integration of the metal hydroxide and oxide ions as:

\[
\begin{align*}
6\text{Fe}^{2+} + 25\text{Bi}^{6+} + \text{Ti}^{2+} + 146\text{OH}^- & \xrightarrow{\text{NH}_3\cdot\text{H}_2\text{O}} 6\text{Bi}_{25}\text{FeO}_{39} + \text{Ti}_2\text{C}_2(\text{OH})_2 + 84\text{H}_2\text{O} \\
\end{align*}
\]  

(3)

This might result in unsuccessfully induced orientations of the Bi$_{25}$FeO$_{39}$ phase within the available d space of 0.93 nm, as its lattice constant is \(a = 1.02\) nm for the cubic I23 space group (JCPDS 046-0416). Hence, the characteristic (002) plane reflection completely disappears in the composite structures due to new interfacial interactions in the Ti$_3$C$_2$Tx @ Bi$_{25}$FeO$_{39}$ composite. The gradual appearance of the Ti$_3$C$_2$Tx @ Bi$_{25}$FeO$_{39}$ composite. The absence of the MXene peaks at 8.9° coming from (002) for the Ti$_3$C$_2$Tx @ Bi$_{25}$FeO$_{39}$ composite indicates the aggregation of the Bi$_{25}$FeO$_{39}$ phase on the MXene flake.35

**SEM.** Figure 3 presents micrographs obtained for all the samples. As can be noticed, the HF etching resulted in multilayer nanosheets of Ti$_3$C$_2$Tx due to the removal of aluminum layers from the dense, compact structures of Ti$_3$AlC$_2$ shown in Figure 3b. The interlayer spacing in Ti$_3$C$_2$Tx resulted in the (002) peak in the XRD profiles of Ti$_3$C$_2$Tx @ Bi$_{25}$FeO$_{39}$ composites. The integration of BiFeO$_3$ retained the sheet structure of Ti$_3$C$_2$Tx, but it became thick and non-uniform as shown in Figure 3d. This may be the different orientation of the rhombohedral BiFeO$_3$ Bravais lattice in between the 2D MXene layers. But the use of ammonia while integrating the Bi$_{25}$FeO$_{39}$ phase in the Ti$_3$C$_2$Tx structure resulted in the aggregation of the particles all around the 2D structures as shown in Figure 3c.36

**FTIR Analysis.** The vibrational spectra mainly originate from the hydroxyl group obtained from the FTIR characterization shown in Figure 4 for the Ti$_3$C$_2$Tx @ Bi$_{25}$FeO$_{39}$ composite. The vibration \(\nu_O - H\) from out-of-plane stretching and bending of the hydroxyl group originated at 3470 and 1695 m$^{-1}$, respectively. The similar nature of the FTIR spectra between Ti$_3$C$_2$Tx and Ti$_3$C$_2$Tx @ BiFeO$_3$ indicates the absence of any oxidized phase in the composite structure.38

**Figure 3.** SEM micrographs for (a) Ti$_3$AlC$_2$, (b) Ti$_3$C$_2$Tx, (c) Ti$_3$C$_2$Tx @ Bi$_{25}$FeO$_{39}$ and (d) Ti$_3$C$_2$Tx @ BiFeO$_3$ composites.

**Figure 4.** FTIR spectra for Ti$_3$C$_2$Tx, Ti$_3$C$_2$Tx @ BiFeO$_3$, and Ti$_3$C$_2$Tx @ Bi$_{25}$FeO$_{39}$ obtained through the hydrothermal reaction (color online).
Bandgap Studies. Bandgap determination was performed using the reflectance spectra shown in Figure 5(a). The reflectance spectra of 

\[ \text{T}_1 \text{C}_2 \text{T}_x \] 

and 

\[ \text{T}_1 \text{C}_2 \text{T}_x @ \text{BiFeO}_3 \] 

samples shows a continuous pattern for 

\[ \text{T}_1 \text{C}_2 \text{T}_x \] 

samples, whereas the reflectance edge is visible for only 

\[ \text{T}_1 \text{C}_2 \text{T}_x @ \text{Bi}_2\text{FeO}_3 \] 

samples. MXene has superior conductivity to metals. Hence, the possible overlapping of the conduction and valence bands resulted in a zero bandgap for MXene, whereas the fabrication of BiFeO\(_3\) with a small quantity retained the original multilayer sheet-like structure of MXene in the 

\[ \text{T}_1 \text{C}_2 \text{T}_x @ \text{Bi}_2\text{FeO}_3 \] 

composite, which behaves similarly to the MXene powders. Not only that, the restacking in the 

\[ \text{T}_1 \text{C}_2 \text{T}_x @ \text{Bi}_2\text{FeO}_3 \] 

composite does not affect the overall surface reflectance of the multilayer flakes.

The strong aggregation of the 

\[ \text{Bi}_2\text{FeO}_3 \] 

particles strongly affects the interaction of the 

\[ \text{T}_1 \text{C}_2 \text{T}_x \] 

flakes with light, although the same 25 wt % anharmonic oscillations originating from the dipoles of the Fe, Bi, and O ions resulted in a separation between the two Morse curve states in 

\[ \text{T}_1 \text{C}_2 \text{T}_x @ \text{Bi}_2\text{FeO}_3 \] 

composites. The direct bandgap is determined by Tauc’s plot with the Kubelka–Munk equation 

\[ E_g = h\nu - \left[ F(R(\infty))h\nu\right]^2 \] 

shown in Figure 5(b). The narrow bandgap energy for the 

\[ \text{T}_1 \text{C}_2 \text{T}_x @ \text{Bi}_2\text{FeO}_3 \] 

composites was determined to be 2.2 eV.

Capacitive and EIS Properties. The room-temperature variation of complex permittivity (\(\epsilon’\) and \(\epsilon''\)) with frequency is presented in Figure 6. The frequency dispersion due to 25 wt % ferrite phase loading on MXene can be observed for both the composites. But the higher value of the complex permittivity (\(\epsilon’\) and \(\epsilon''\)) was observed at lower frequencies for the BiFeO\(_3\) phase fabrication on MXene. These directly indicate the better storage and dissipation capabilities of the electrical energies by the 

\[ \text{T}_1 \text{C}_2 \text{T}_x @ \text{Bi}_2\text{FeO}_3 \] 

composites. The different dielectric properties between the two composites indicate a different dielectric relaxation mechanism within the composites. This nature relates to the interfacial nature of the MXene sheets and the existence of the rhombohedral lattice structure of the BiFeO\(_3\) nanoparticles. The interfacial nature of the MXene sheets resulted in the accumulation of electrons and ions in alternative layers, resulting in interfacial polarization. On the other hand, the rhombohedral lattice resulted in orientational relaxation inside the 

\[ \text{T}_1 \text{C}_2 \text{T}_x @ \text{BiFeO}_3 \] 

composites due to the spontaneous dipole moment with numerous defects in the BiFeO\(_3\) crystals causing permanent defect dipoles. These defect dipoles aligned themselves to the applied electric field when the frequencies were low, resulting in the orientational polarization. At higher frequency, due to the misalignment...
owing to the effect of inertia, permittivity decreases. In general, the dielectric nature of the composites can be described by Debye and Maxwell–Wagner (M–W) mechanisms:\(^43\)

\[
\varepsilon''(\omega) = \frac{\varepsilon_0 - \varepsilon_\infty}{1 + \omega^2 \tau^2} \quad \text{(Debye)}
\]

\[
\varepsilon''(\omega) = \frac{\varepsilon_0 - \varepsilon_\infty}{1 + \omega^2 \tau^2} + \frac{\sigma}{\omega} (M - W)
\]

whereas the first part of eq 5 is the same as the Debye mechanism of eq 4. The fitting of the dielectric data for both the composites is presented in Figure 6b. It can be visibly noticed that the Ti₃C₂Tₓ@Bi₂₅FeO₃₉ composites satisfy the Debye relaxation mechanism inside the Ti₃C₂Tₓ@Bi₂₅FeO₃₉ composites.

The resistance of the composites was further calculated using the Nyquist plots shown in Figure 7. A single semicircular half-arc with different radii was observed for the composites. The higher half-arc radius for the Ti₃C₂Tₓ@Bi₂₅FeO₃₉ composite indicates higher interface resistance, whereas the smaller half-arc radius of the Ti₃C₂Tₓ@BiFeO₃ composite indicates the easy charge transfer process between the interlayers.\(^4^4\) Not only that, at higher frequencies, it offers the minimum real impedance and maximum imaginary impedance. The nature reverses with the decrease in frequency, indicating the capacitance behavior at room temperature.

This shift of the intra- and interlayer effects on the charge carriers can be understood with the equivalent circuit of the impedance data shown in the inset of Figure 7b, and the impedance of the circuit can be written as \(Z_{\text{CPE}} = R_l + R_{il} Z_{\text{CPE}}\), where \(R_l\) is the intralayer resistance, \(R_{il}\) is the interlayer resistance, and \(Z_{\text{CPE}}\) is the constant phase element. The obtained parameters with the fitting results are presented in Table 1. The interlayer resistance for the Ti₃C₂Tₓ@Bi₂₅FeO₃₉ composite was found to be \(2.506 \times 10^8\) Ω, whereas the interlayer resistance for the Ti₃C₂Tₓ@BiFeO₃ composite is the same as that for the 2D Ti₃C₂Tₓ as reported previously.\(^4^5\) Hence, the successful fabrication of BiFeO₃ in the 2D Ti₃C₂Tₓ resulted in a single relaxation mechanism, non-exponential variation, and decreased interlayer barrier on the mobility of charge carriers.

**Magnetic Properties.** The magnetic hysteresis loop for the Ti₃C₂Tₓ@Bi₂₅FeO₃₉ and Ti₃C₂Tₓ@BiFeO₃ composites is presented in Figure 8 at room temperature with a maximum of 16,000 Oe of the applied magnetic field. It is evident from the graph that the composite shows a weak magnetic behavior and the maximum of 16,000 Oe field is not sufficient to saturate the magnetization. The magnetization in this regard is expressed with the formula \(M = \frac{m M_s}{5585}\) in terms of Bohr magneton (\(\mu_B\)).\(^4^6\) The obtained values of the saturation magnetization (\(M_s\)) were 0.0036 and 0.0031 \(\mu_B\) for the

![Figure 7](https://example.com/fig7.png)

**Figure 7.** Nyquist plot of the (a) Ti₃C₂Tₓ@Bi₂₅FeO₃₉ and (b) Ti₃C₂Tₓ@BiFeO₃ composites with an equivalent circuit (inset) (color online).

![Figure 8](https://example.com/fig8.png)

**Figure 8.** Room-temperature magnetic hysteresis loop for the Ti₃C₂Tₓ@Bi₂₅FeO₃₉ and Ti₃C₂Tₓ@BiFeO₃ composites. The inset shows the retentivity and coercivity of the composites (color online).

### Table 1. Electrical Parameters Obtained from EIS Studies

| Parameter | Ti₃C₂Tₓ@Bi₂₅FeO₃₉ | Ti₃C₂Tₓ@BiFeO₃ |
|-----------|------------------|----------------|
| \(R_l\) (Ω) | 44               | 45             |
| \(R_{il}\) (Ω) | 2.506 \times 10^8 | 60             |
| \(T\) (Ω s⁻¹) \times 10^−10 | 6.654 | 1.130 |
| \(P\) | 0.70 | 0.99 |

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BiFeO$_3$ with MXene retained the original multilayer sheet-like spacer between the multilayers. The fabrication of 25 wt % with the 2D Ti$_3$C$_2$T$_x$ reducing agent while hybridizing the multiferroic structure force of attraction of the ions. The use of ammonia as a desired morphology and performances using the electrostatic appropriate reducing agent and balanced wt % may lead to the 2D materials.

The design and synthesis of the composite material with an appropriate reducing agent and balanced wt % may lead to the desired morphology and performances using the electrostatic force of attraction of the ions. The use of ammonia as a reducing agent while hybridizing the multiferroic structure with the 2D Ti$_3$C$_2$T$_x$ structure using the hydrothermal method may lead to severe agglomeration with the Ti$_3$C$_2$T$_x$ @ Bi$_{25}$FeO$_{39}$ phase. The electrostatic interactions and neutralization of the −ve hydroxyl ion vibration by the +ve metal ions were observed with the redshift of the FTIR spectra with a 2.2 eV narrow bandgap energy in Ti$_3$C$_2$T$_x$ @ Bi$_{25}$FeO$_{39}$ agglomerated samples.

At the same time, the use of N$_2$H$_4$·H$_2$O as a reducing agent while hybridizing the multiferroic structure with the 2D Ti$_3$C$_2$T$_x$ structure leads to a Ti$_3$C$_2$T$_x$ @ BiFeO$_3$ composite as a spacer between the multilayers. The fabrication of 25 wt % BiFeO$_3$ with MXene retained the original multilayer sheet-like structure and metallic behavior observed from SEM and UV–Vis analysis. The dielectric analysis indicates a dc conductivity of 1.30 × 10$^{-11}$ S/m magnitude with the presence of both interfacial and defect dipole polarization mechanisms. Moreover, the thin M-H loop means a minimal magnetic loss inside the composites. Hence, this straightforward, inexpensive, and quick method of preparation with superior properties may be applied to 2D multiferroic materials.

**CONCLUSIONS**

The engineering of the 2D Ti$_3$C$_2$T$_x$ structures with interconnected multilayers with nanometer spacing is crucial for capacitor applications. The better electrical conductivity plays a vital role in the transport of electrons within this interconnected network of 2D layers. Hence, the hybridization of the multiferroic BiFeO$_3$ with spontaneous polarization due to the rhombohedral structure with MXene can be a critical step in fabricating superior capacitor materials. The main challenge associated with it is to stop agglomeration and provide mechanical support for the restacking of the flakes by finding a balance between the multiferroic spacers inside the 2D materials.

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**Notes**

The authors declare no competing financial interest.

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