ABSTRACT: It is still very challenging to effectively design nanocomposite microstructures with significantly improved electromagnetic interference shielding effectiveness (EMI SE). Herein, we developed a facile method for fabrication of molybdenum disulfide/graphene nanoplatelets (MoS$_2$/GNPs) nanocomposites, in which GNPs are utilized as highly effective electrical transport materials, while MoS$_2$ resolves the agglomeration problem of GNPs. GNPs also serve as an efficient cluster of electrical transport systems and dampen the incoming electromagnetic wave. Two types of samples are synthesized and compared in context of EMI SE values: physically mixed composite and layered samples. The sandwiched MoS$_2$ between GNP layers showed an EMI SE of $\sim$24 dB, which was an almost 14% improvement relative to MoS$_2$/GNPs nanocomposites exhibiting an EMI SE value of $\sim$21 dB, both containing 0.5 wt % GNPs. This work provides a new strategy for the design of multifunctional nanocomposites using the simple low-cost vacuum filtration method for EMI shielding for future applications.

INTRODUCTION

Electronics are everywhere, from autonomous vehicles to space technology in recent times. As a result, a new type of pollution known as electromagnetic (EM) pollution has emerged.$^{1,2}$ Electromagnetic interference (EMI) in electrical devices is a common problem that, if not properly addressed, can cause system damage. Medical gadgets suffer from EMI, which can cause injury to the user and prevent the device from achieving the desired outcomes.$^3$ EMI can momentarily disable a piece of technology and, if left unprotected for a long period of time, can cause irreparable damage. As a result, it is critical to ensure that all electronic devices are appropriately protected from interference. To resolve this issue of EMI, new materials and designs that can effectively absorb these EM waves must be developed.

Metal shielding is the most typical approach for EMI protection. Metals have the capability of absorbing and reflecting waves, removing all emissions, and providing electronic shielding. However, experiments are being conducted to see whether lightweight materials like carbon nanotubes, graphite microfibers, and metal oxides such as ZnO, CuS, MnO, Fe$_2$O$_3$, NiO/SiC, and Fe$_3$O$_4$@TiO$_2$ could be used to replace the metals. Molybdenum disulfide is a transition metal chalcogenide, with a layered structure comparable to that of graphite, that has gained a lot of consideration because of its semiconducting properties.$^{10}$

For EMI shielding, graphene-based nanocomposites have been widely studied. Stretched graphene nanosheets (GNSs) generating obstacle walls in melamine sponge were investigated by Guo et al.$^{11}$ for excellent EMI shielding. By using a fluid-assisted technique, the GO nanosheets covered with melamine sponge pores were utilized, followed by freeze-drying and reduction procedures. The composite foam had a density of 0.011 g/cm$^3$, an EMI SE of 37.2 dB, and a specific EMI SE of 845.5 dB·cm$^2$/g with a graphene loading of 0.1 vol %. Song et al.$^{12}$ investigated the EM shielding properties of flexible multilayer graphene sheets with high conductivity and found the effective EM shield equal to 27 dB. Guo et al.$^{13}$ reported the biomass-derived carbon and iron oxide with good EM wave absorption characteristics.

Composites of MoS$_2$/GNPs have found potential applications within the region of energy, capacity, and transformation. The dielectric properties and electromagnetic wave assimilation execution of the MoS$_2$/GNPs composite have never been detailed. In this aspect, the composites of MoS$_2$/GNPs
were arranged and their predominant electromagnetic wave retention properties were proved.\textsuperscript{14} Moreover, the shape of nanomaterials also affects the ultimate electrical properties such as electrical conductivity and electromagnetic interference shielding efficacy and dielectric properties.\textsuperscript{13,16}

The strategy for effective EMI shielding is to increase the real part of the complex permittivity, $\varepsilon''$, which is associated with the absorption of EM waves. The increase in $\varepsilon''$ comes from multiple polarization mechanisms such as electronic, ionic, dipolar, and space charge polarizations.\textsuperscript{17–20} Since ionic and electronic contributions dominate at low frequency regimes, therefore, key to the enhancement of $\varepsilon''$ at high EM wave frequencies will be to enhance interface and dipolar polarizations. Various interface design strategies have been adopted to increase space charge polarization, which are usually complex.\textsuperscript{17–19} In this study, we have adopted a simple strategy where void-free compact-layered structures of 2D GNPs and MoS$_2$ nanosheets were fabricated by the simple and cost-effective vacuum filtration method. Two design strategies were adopted to fabricate these layered structures. One strategy involved a single layer of physically mixed composite materials of GNPs and MoS$_2$ with varying loadings, and the other one considered the bi- and trilayer structures of these materials where each layer was composed of a single type of 2D material. The sandwich construction, i.e., trilayer structures, showed enhanced EMI shielding effectiveness due to more space charge polarization losses along with other types of losses because of multiple interfaces. These layered constructions could be used in many fields, such as flexible electronics, automobile industry, and space technologies.

**EXPERIMENTAL SECTION**

**Materials.** Bulk MoS$_2$ and graphite powder were purchased from Jiangsu XFNANO Materials Tech Co., Ltd., China. N-Methyl-2-pyrrolidone (NMP) and nylon filter papers with a pore size of 0.45 $\mu$m were obtained from Sigma-Aldrich.

**Synthesis of 2D Materials.** First, 2D materials of MoS$_2$ and graphene were prepared from bulk materials by exfoliation in NMP. For the synthesis of 2D nanosheets of MoS$_2$, 0.2 g of MoS$_2$ was mixed in 100 mL of NMP and dispersed by magnetic stirring for 45 min. After that, the dispersion was probe-sonicated for 64 h using 40% amplitude at 0.3 cycle. This 100 mL dispersion was equally distributed in six centrifuge tubes, which were centrifuged at 1500 rpm for 45 min to exfoliate MoS$_2$. The supernatants were transferred to another set of six tubes, which were further centrifuged at 1000 rpm for 45 min. Subsequently, the exfoliated dispersions were poured into the vacuum filtration assembly. After filtration through 0.45 $\mu$m nylon filter paper, the filtered cake was removed from the filtration assembly and dried overnight in a vacuum oven at the temperature of 60 °C. Once dried, MoS$_2$ was scratched from the nylon filter paper and was used for layered structure fabrication and further testing.

A similar procedure was used to prepare graphene nanoplatelets by exfoliation of bulk graphene powder in NMP.

**Layered Structure Fabrication by Vacuum Filtration.** Vacuum filtration was used to fabricate layered structures of synthesized 2D materials. Two design strategies were adopted: the first strategy involved a single-layer structure containing physically mixed nanocomposites of 2D materials, and the second one dealt with the bi- and trilayer structures where, instead of nanocomposites, each layer was composed of a single 2D material. The overall loading of 2D materials in all layered structures, single-, bi-, and trilayer ones, was kept constant, i.e., 6 mg. The required amount of 2D materials, for each layer, was dispersed in 12 mL of 1:1 ethanol–water solution in an ultrasonic bath for 30 min and then filtered through the nylon paper by vacuum suction. The loading and sequence of layers are given in Table 1. The single-layer structures were fabricated by physically mixing 0, 50, 75, and 100% MoS$_2$ nanosheets with graphene platelets in ethanol–water solution and then filtering each dispersion through the nylon filter paper by suction. The filtered cakes were dried in an oven at 60 °C for 1 h. The process schematic is shown in Figure 1.

| Table 1. Sample Designation for Single-, Bi-, and Trilayer Structures |
|-------------------|-----------------|-----------------|
| single layer      | composition and sequence of layers in a structure | sample/structure designation |
| 6 mg of graphene  | 6G              |
| 3 mg of MoS$_2$ and 3 mg of graphene | 3MS-3G |
| 4 mg of MoS$_2$ and 2 mg of graphene | 4MS-2G |
| 6 mg of MoS$_2$   | 6MS             |
| bilayer           |                 |
| 3 mg of MoS$_2$/3 mg of graphene | 3MS/3G |
| trilayer          |                 |
| 2 mg of MoS$_2$/2 mg of graphene/2 mg of MoS$_2$ | 2MS/2G/2MS |
| 2 mg of graphene/2 mg of MoS$_2$/2 mg of graphene | 2G/2MS/2G |

In bilayer structures, the first layer was fabricated by dispersing 3 mg of MoS$_2$ in ethanol–water solution followed by vacuum suction through nylon filter paper. The filtered cake was dried in an oven at 60 °C for 1 h. Subsequently, this dried cake was again loaded into the vacuum filtration assembly and a dispersion containing 3 mg of graphene platelets was filtered through it to fabricate the second layer. The filtered cake was again dried in an oven at 60 °C. A similar procedure was adopted for the fabrication of trilayer structures where 25% of each 2D material was sandwiched between the two layers of the other material (Table 1).

**Characterizations.** The surface morphologies of synthesized samples were analyzed using scanning electron microscopy (SEM) (JEOL-JSM-6490LA). The crystal structure of the prepared powder materials was probed using X-ray
diffraction (XRD, STOE Siemens D5005) with CuKα radiation (\( \lambda = 1.54 \text{ Å} \)) operating at a voltage of 40 kV and a working current of 40 mA. The samples were subjected to a step scan of 0.04° and a scan rate of 1.00 s.

EMI shielding is primarily determined by the electrical conductivity and magnetic permeability of materials, as well as the frequency of radiation has also the major role. The effectiveness of the shielding material is the ratio of incident power (\( P_I \)) to transmitted power (\( P_T \)) through the shield, as given by eq 1:

\[
\text{EMI SE (dB)} = 10 \log \frac{P_I}{P_T}
\]  

(1)

EMI testing of prepared samples was carried out at a PNA Model E8364B. The data was obtained as scattering parameters, which were a combination of reflection responses (eq 2) and transmission (eq 3). These scattering parameters are called S-parameters, which are \( S_{11}, S_{12}, S_{21}, \) and \( S_{22} \):

\[
\text{reflectance (R)} = \frac{P_R}{P_I} = S_{11}^2 = S_{22}^2
\]  

(2)

\[
\text{transmittance (T)} = \frac{P_C}{P_I} = S_{12}^2 = S_{21}^2
\]  

(3)

The sum of the contributions from shielding effect reflection (\( \text{SE}_R \)), shielding effect absorption (\( \text{SE}_A \)), and various internal reflections makes up the total EMI SE (\( \text{SE}_T \)) (SEM). Because the re-reflected waves are dissipated as heat in the shielding material at higher EMI SE values and with a multilayer EMI shield, contribution from numerous internal reflections is integrated in the absorption. The total EMI SE (\( \text{SE}_T \)) is the sum of the contributions from reflection (\( \text{SE}_R \)), absorption (\( \text{SE}_A \)), and multiple internal reflections (\( \text{SE}_M \)) (eq 4):

\[
\text{SE}_T = \text{SE}_R + \text{SE}_A
\]  

(4)

where both \( \text{SE}_R \) (eq 5) and \( \text{SE}_A \) (eq 6) are as follows,

\[
\text{SE}_R = 10 \log \frac{1}{1 - S_{11}^2}
\]  

(5)

\[
\text{SE}_A = 10 \log \frac{1 - S_{21}^2}{S_{21}^2}
\]  

(6)

When an impedance mismatch exists between the shielding material’s surface and the incoming radiation, microwave reflection occurs. However, absorption occurs when incoming electromagnetic wave energy is attenuated, and the internal inhomogeneity of the shielding material causes repeated reflections. It is worth noting that an \( \text{SE}_A \) of less than 10 dB is adequate to eliminate multiple reflection contributions. The total shielding effectiveness (\( \text{SE}_T \)) is the total shielding contribution owing to the above-mentioned components, and the specifics tying it to the various scattering characteristics have been described elsewhere.

The complex permittivity properties of the synthesized materials including dielectric constant and dielectric loss were measured with a vector network analyzer (VNA, N5242A PNA-X, Agilent) at room temperature in the frequency range of 1 to 8 GHz. For finding the real factor of resistivity, the following relation (eq 7) was used

\[
\varepsilon' = \frac{C d}{A \varepsilon_0}
\]  

(7)

Here, \( \varepsilon' \) is the dielectric constant, \( C \) is the capacitance, \( d \) is the distance between the plates, and \( \varepsilon_0 \) is the permittivity of free space.

For finding the imaginary factor of resistivity, the following equation (eq 8) was used, where \( \tan \delta \) is the dissipation factor

\[
\varepsilon'' = \varepsilon' \tan \delta
\]  

(8)

### RESULTS AND DISCUSSION

**Morphological and Structural Analyses.** The morphologies of synthesized 2D materials of GNP's and MoS₂ are
shown in Figure 2. SEM images of synthesized GNPs revealed triangular and rectangular forms (Figure 2A,B) with layered sheets of sizes varying between 1 and 5 μm and thickness in the 2–10 nm range (Figure 2). A broad variety of layered agglomerates of varying shapes and numbers of layers were observed. There were no single- or few-layer sheets of graphene found. The flake-like structures with stacking of few layers were evident from the morphological analysis of synthesized MoS\(_2\) (Figure 2C,D). The MoS\(_2\) sheets were composed of clusters of various thicknesses. As reported in the literature, the mechanical strength between the layers of bulk MoS\(_2\) was weakened during probe sonication\(^{21}\) and nanosheets of sizes ranging from 1 to 5 μm were obtained.

The elemental maps of single-layer composite structures along with their corresponding SEM images are shown in Figure 3. The SEM images revealed the dominance of morphologies in composite structures similar to the ones observed for pristine graphite nanosheets (Figure 2A,B) when the concentration of GNPs was increased in the composite materials (Figure 3E,F). Meanwhile, the morphologies of composites having higher concentrations of MoS\(_2\) were similar to that of pristine MoS\(_2\). The sulfur and molybdenum contents decreased with increasing content of graphene in a physically mixed composite layer (Figure 3A−F). The dispersion of both phases was mostly uniform in the composite layer. The large interfacial contact between MoS\(_2\) and GNP 2D materials was evident for 3MS-3G composition (Figure 3C,D) compared to the other compositions (Figure 3A,B,E,F).

To find the thickness and lateral length of nanosheets, the 2D materials were analyzed by atomic force microscopy (AFM), and results are shown in Figure 4. The length of GNPs was around 0.2 μm, and the height was ~5.2 nm. Also, areal roughness parameters, \(R_a\) and \(R_z\), were 2.69 and 20.8 nm, respectively, for GNPs. MoS\(_2\) flakes were ~0.5 μm in length, and their thickness was ~10.2 nm. Similarly, for MoS\(_2\), \(R_a\) and \(R_z\) were 1.62 and 31.62 nm, respectively.

Figure 5 shows the XRD patterns of graphene nanoplatelets (GNPs). The characteristic peaks of graphene are evident in Figure 5A. A high intensity peak at a 2θ value of 26° was associated with the (002) plane of graphite. Also, two less intense peaks at 44° and 54° are evident in Figure 5A, which belonged to (100) and (004) planes of graphene, respectively. These characteristic peaks of GNPs match well with PDF card #00-02-0212 given in Figure 5B and conform to the reported literature.\(^{22}\)

Figure 6A shows the XRD pattern of MoS\(_2\). The diffraction peaks at 14.37°, 32.67°, 35.86°, 39.53°, 44.16°, 49.76°, 58.32°, and 60.12° belonged to (002), (100), (102), (103), (104), (105), (110), and (112) planes, respectively. The observed XRD peaks of MoS\(_2\) matched well with reference PDF card #01-077-1716 shown in Figure 6B and with a previous report.\(^{23}\)

Raman spectroscopy is a quick way to get a direct look at electron–phonon interactions, which means that it is very sensitive to electronic and crystallographic structures.\(^{24,25}\)

Figure 7A illustrates the Raman spectrum of GNPs where the main band for GNPs was observed at around 1600 cm\(^{-1}\). The Raman spectra of carbon compounds have three primary bands between 1200 and 2800 cm\(^{-1}\). The D and G bands at 1360 and 1600 cm\(^{-1}\), respectively, are caused by sp\(^2\) sites, while and
the T band at 1060 cm$^{-1}$ is due to sp$^3$ contributions.\textsuperscript{14} There was no D band observed for synthesized GNPs (Figure 7). Some literature reports suggest that the Raman spectrum would be redshifted when graphene is subjected to strain because of the stretch of the carbon–carbon bond.\textsuperscript{26,27}

In Figure 7B, first-order $E_{2g}$ at around 289 cm$^{-1}$ corresponding to MoS$_2$ was observed when the incident energy was considered nonresonant. MoS$_2$ showed aberrant resonance Raman characteristics as reported in ref 28. The resonant activation of exciton states mediated by acoustic phonon scattering causes the central peak.\textsuperscript{28} Furthermore, due to resonance with exciton or exciton–polariton state characteristics, several second-order Raman peaks were observed.

**Electromagnetic Shielding Performances of Multilayer Structure Films.** The effectiveness of EMI shielding with single-layer physically mixed composite structures is shown in Figure 8. The EM absorption capacities of both pristine 2D materials, i.e., MoS$_2$ and GNPs, were lower compared to those of their composites (Figure 8A). However, pristine GNPs illustrated greater EM reflection behavior compared to pristine MoS$_2$, which may be attributed to higher electrical conductivities of GNPs compared to those of MoS$_2$ nanosheets. The addition of MoS$_2$ into GNPs or vice versa led to enhanced EM wave absorption characteristics of the resultant composite (Figure 8A). The maximum EMI SE was established at 50% content of each phase, i.e., 3MS-3G, where it reached a value of $-20.8$ dB at a lower frequency of 2 GHz. A drop in EMI SE was observed for all compositions at frequencies higher than 5 GHz (Figure 8C).

As has previously been established,\textsuperscript{29} multiple polarizations such as interfacial, dipolar, ionic, and electronic polarizations\textsuperscript{9,30} and relaxation mechanisms can explain the absorption of EM waves impinging on the hybrid structures of various compositions. The electronic and atomic dipoles have a quick reaction to the alternating EM field in the GHz region and synchronize with the EM wave, resulting in no EM wave energy loss.\textsuperscript{30} The contributions from ionic and electronic polarizations usually occur at lower frequencies, i.e., UV or IR; therefore, interfacial and dipole polarization effects may dominate in increased EMI SE at higher frequency ranges.\textsuperscript{19}

It has been observed that the additions of GNPs in MoS$_2$ resulted in enhanced EMI SE (Figure 8), and the large interface contact due to these graphene additions may have provided the platform for the polarization. Also, with high electrical conductivity, graphene sheets with a high aspect ratio tend to raise the percolation network.\textsuperscript{31} The charge carriers get accumulated at the interface heterostructures, causing space charge polarization, and since these charges find themselves unable to respond in accordance with the incident EM field, they lead to the energy loss of EM radiation.\textsuperscript{32} Hence, the maximum EMI SE was obtained for the single-layer 3MS-3G composite.

The EMI SE of the bi- and trilayer structures was better compared to that of the single-layered composite structures (Figure 9). The bilayer structure, i.e., 3MS/3G, showed poor EM wave absorption and overall EM shielding characteristics

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**Figure 7.** Raman scan of 2D materials: (A) GNPs and (B) MoS$_2$.

**Figure 8.** EMI SE characteristics of various single-layer composite structures. (A) Absorption, (B) reflection, and (C) total EMI shielding effectiveness.

**Figure 9.** EMI shielding effectiveness of bi- and trilayer structures. (A) Absorption, (B) reflection, and (C) total EMI shielding effectiveness.
Table 2. EMI Shielding Effectiveness Data

| sr. no. | author | material | year | fabrication/synthesis method | dB ranges | reference |
|---------|--------|----------|------|--------------------------------|-----------|-----------|
| 1       | Ding et al. | rGO-MoS$_2$-Fe$_2$O$_4$ | 2021 | hydrothermal technique | −48 dB (reflection) | 37 |
| 2       | Prasad et al. | Co@MoS$_2$/rGO | 2020 | autoclave | 29−46 dB | 38 |
| 3       | Zahid et al. | RGO/TPU | 2020 | solution casting technique | 53 dB | 39 |
| 4       | Shakir et al. | PS/PANI blends | 2020 | solution casting method | 45 dB | 40 |
| 5       | Zhang et al. | S-rLGO/P(St-BA) latex | 2020 | blending and casting | 16 dB | 33 |
| 6       | Khan et al. | graphene and MoS$_2$ | 2020 | blending and filtration | 18 dB | 34 |
| 7       | Prasad et al. | MoS$_2$-rGO and MoS$_2$-rGO/Fe$_2$O$_4$ nanostructure | 2018 | hydrothermal method | 3.81 dB (MoS$_2$-rGO) and 8.27 dB (MoS$_2$-rGO/Fe$_2$O$_4$) | 35 |
| 8       | Guo et al. | GO nanosheets covered with melamine sponge | 2019 | fluid-assisted method | 37 dB | 11 |
| 9       | Shakir et al. | PVC/PANI/TRGO | 2019 | hydrothermal technique and solution casting methods | 56 dB | 41 |
| 10      | Zhang et al. | graphene/PMMA nanocomposite | 2011 | foaming with subcritical CO$_2$ | 13−19 dB | 42 |
| 11      | Guo et al. | rGO@BT/poly(vinylidene fluoride) (PVDF) | 2017 | physical mixing and drop casting | 22 dB | 36 |
| 12      | Guo et al. | rGO@MoS$_2$/PVDF | 2016 | blending and hot molding | 28 dB | 43 |
| 13      | Liang et al. | 15% functionalized graphene/epoxy resin | 2009 | in situ functionalization | 21 dB | 44 |
| 14      | Sajid et al. | single-layer composite 3MS-3G | | vacuum filtration | 21 dB | this study |
| 15      | Sajid et al. | trilayer structure 2G/2MS/2G | | vacuum filtration | 24 dB | this study |

Compared to the trilayer ones (Figure 9). In trilayer structures, the EMI SE was the maximum, i.e., −24.4 dB, when the MoS$_2$ layer was sandwiched between GNP layers (2G/2MS/2G; Figure 9) and was relatively lower (−23.7 dB) when the GNP layer was sandwiched between MoS$_2$ layers (2MS/2G/2MS; Figure 9C). Also, the EMI SE of 2G/2MS/2G was constant for the entire frequency region (Figure 9).

The better EM wave shielding properties of bi- or trilayer samples were due to multiple interfaces that might lead to space charge polarization and, hence, dissipation of wave energy as explained earlier. The highest EMI SE of 2G/2MS/2G can be attributed to the fact that the more conductive facing layers of the sandwich structures will allow more EM waves to enter the layered structures and, hence, higher absorption of ME waves due to conductive losses. Also, the difference in electrical conductivities between GNPs and MoS$_2$ will result in higher charge accumulation at these heterostructure interfaces and, hence, further energy losses due to space charge polarization. Therefore, sandwiching the insulator layer of MoS$_2$ in the conducting layer of GNPs was a better design strategy for EMI shielding, probably due to the less reflection and, hence, higher absorption and attenuation of EM waves.

Table 2 provides a comparison of the shielding effectiveness of materials and strategies used in this study and that of the already reported ones. The results of this study are better than those of many of the recently reported literature. For example, Zhang et al. reported sulfonated reduced less defect graphene oxide (S-rLGO) combined with P(St-BA) latex through blending and casting processes and reported an overall shielding effectiveness of 16 dB. Khan et al. found an EMI effectiveness of 18 dB with graphene- and MoS$_2$-based materials. The EMI shielding of the MoS$_2$-rGO/Fe$_2$O$_4$ nanostructure was assessed in the 8−12 GHz range by Prasad et al. and was 8.27 dB. Also, the MoS$_2$-rGO composite showed an inadequate shielding capacity (SE$_T = 3.81$ dB) across the entire spectrum of EM studied. According to the study, interfacial polarization played a vital role in dielectric losses along with other loss mechanisms. Guo et al. studied the rGO@barium titanate (BT)/poly(vinylidene fluoride) (PVDF) composite and concluded that this composite can exhibit an EMI SE$_R$ (reflection) of 23 dB with a filler loading of 25 wt %.

**Dielectric Properties.** The dielectric data of complex permittivity of the fabricated single-layer composite structures of MoS$_2$ and GNPs is given in Figure 10. This data showed the typical frequency-dependent behavior and can be explained based on the Debye theory (eq 9),

$$\varepsilon = \varepsilon' - j\varepsilon'' = \varepsilon_0 \exp(-\delta)$$

where the dielectric constant ($\varepsilon'$) is the ability of a material to store electric charge in the electric field. The imaginary part ($\varepsilon''$), usually referred to as the loss factor, is a material’s ability to absorb or dissipate energy through conversion of electric energy into heat.15

Figure 10A shows the data for the real part of the complex permittivity, $\varepsilon'$, for single-layer composite structures. At low frequencies, all the materials showed high values of dielectric constant and the values of dielectric constant dropped with increasing frequencies. This behavior can be explained based on Koop’s theory, according to which dielectric structures are the inhomogeneous Maxwell Wagner type.15 The materials are composed of conducting grains separated by nonconducting boundaries. The hopping of electrons at the interface because of grain boundaries in the composite interface facilitates the higher permittivities. GNPs had a dielectric constant of 20,
additions of MoS₂ in GNP layers resulted in an increase in dielectric constant, and it reached a value of around 120 for the 3MS-3G composite structure (Figure 10A). Primarily, the addition of GNP layers into MoS₂ was beneficial for the creation of multiple interfaces, facilitating the interfacial polarization of the composites and higher dielectric permittivity. The larger surface area of MoS₂/GNPs structures resulted in greater dipole polarization, which increased the dielectric constant. The charge on the surface of the MoS₂/GNPs heterostructure can drive the migration and redistribution of atoms or dipoles within the material.

The increasing concentrations of MoS₂ in GNP layers led to an increase in complex permittivity, ε″, and maximum values of the complex permittivity, ε″, at all frequencies were observed for 3MS-3G structures (Figure 10B). Since the contributions to the complex permittivity, ε″, due to electronic and ionic polarizations mostly dominate in IR or visible regions of the EM spectrum, the observed enhancement of the complex permittivity, ε″, would come from dipolar and interface polarization mechanisms. The peaks in complex permittivity, ε″, versus frequency graphs (see arrows in Figure 10B) at high frequencies are usually indicative of contributions from the interface polarization effect. Therefore, heterostructure interfaces in the 3MS-3G single-layer composite structure resulted in higher complex permittivity, ε″, as was evident by the peaks at higher frequencies (Figure 10B).

The tangent loss behavior of single-layer composite structures is shown in Figure 10C. The dielectric loss was comparatively large in the lower frequency region due to the charge mobility in the composite materials, whereas the dielectric loss was decreased in the higher frequency region due to the dipolar and interfacial polarization losses. Furthermore, in the high frequency range, a steady increase can be seen, it is because of dynamic relaxation of segmental motions in the amorphous phase, and it is called the dielectric relaxation peak of dielectric loss. These dielectric loss results were consistent with the imaginary part and real part of dielectric constants (Table 3).

**Table 3. Dielectric Properties of Different Samples at 2 GHz**

| Sample  | ε′  | ε″  | tan δ
|--------|-----|-----|-------|
| 6MS    | 70.9| 600.10| 0.68 |
| 6G     | 18.5| 100 | 0.95 |
| 3MS-3G | 112.0| 1100 | 0.52 |
| 2MS-4G | 16.3| 150 | 0.10 |
| 4MS-2G | 79.7| 700 | 0.15 |

**CONCLUSIONS**

This study successfully exploited the simple low-cost vacuum filtration method for the fabrication of layered heterostructure interfaces of 2D materials of graphene and MoS₂ to mitigate the electromagnetic interference. Various layered structures were established by varying the concentrations of 2D materials in a single layer or by changing the order of each 2D material in bi- or trilayer structures. The heterostructures of bi- or trilayers exhibited better EMI SE with dB values reaching 24 compared to the single-layer composite structures, which could be due to the multiple interfaces present in these structures because of interface polarization effects. The sandwiching of MoS₂ in GNP layers was a better strategy compared to the physical mixing of these materials into a single composite layer with the same loading of each 2D material.

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■ REFERENCES
(1) Anderson, L.; Govindaraj, P.; Ang, A.; Mirabedini, A.; Hameed, N. Modelling, Fabrication and Characterization of Graphene/Polymer Nanocomposites for Electromagnetic Interference Shielding Applications. Carbon Trends 2021, 4, No. 100047.
(2) Kulkarni, G. K.; JadHAV, S. A.; Patil, K. T.; patil, P. S.; Puri, V. R. α-MnO2 nanorods-polyaniline (PANI) nanocomposites synthesized by polymer coating and grafting approaches for screening EMI pollution. Ceram. Int. 2021, 47, 15044−15051.
(3) Mesquita, A.; Glenn, J.; Jenny, A. Differential activation of eMI by distinct forms of cellular stress. Autophagy 2021, 17, 1828−1840.
(4) Shen, B.; Zhai, W.; Tao, M.; Ling, J.; Zheng, W. Lightweight, multifunctional polyetherimide/graphene@Fe3O4 composite foams for shielding of electromagnetic pollution. ACS Appl. Mater. Interfaces 2013, 5, 11383−11391.
(5) Zubair, K.; Shakir, M. F.; Afzal, A.; Rehan, Z. A.; Nawab, Y. Effect of barium hexaferrites and thermally reduced graphene oxide on EMI shielding properties in polymer composites. J. Supercond. Novel Magn. 2021, 34, 201−210.
(6) Kruželj, Ľ.; Kvasničáková, A.; Hložeková, K.; Hudec, I. Progress in polymers and polymer composites used as efficient materials for EMI shielding. Nanoscale Adv. 2021, 3, 123−172.
(7) Hu, X.-S.; Shen, Y.; Xu, L.-H.; Wang, L.-M.; Lu, L.-s.; Zhang, Y. Preparation of flower-like CuS by solvothermal method for photocalytic, UV protection and EMI shielding applications. Appl. Surf. Sci. 2016, 385, 162−170.
(8) Agarwal, P. R.; Kumar, R.; Kumari, S.; Dhakate, S. R. Three-dimensional and highly ordered porous carbon−MnO2 composite foam for excellent electromagnetic interference shielding efficiency. RSC Adv. 2016, 6, 100713−100722.
(9) Choi, Y.-J.; Gong, S. C.; Johnson, D. C.; Golledge, S.; Yeom, G. Y.; Park, H. H. Characteristics of the electromagnetic interference shielding effectiveness of Al-doped ZnO thin films deposited by atomic layer deposition. Appl. Surf. Sci. 2013, 269, 92−97.
(10) Chevelanath, P.; Hassain, M. I.; Husna, J.; Alghoul, M.; Sopian, K.; Amin, N. Effects of transition metal dichalcogenide molybdenum disilicide layer formation in copper−tin−tin−sulfur solar cells from numerical analysis. Jpn. J. Appl. Phys. 2012, 51, 10NC32.
(11) Guo, T.; Chen, X.; Su, L.; Li, C.; Huang, X.; Tang, X.-Z. Stretched graphene nanosheets formed the “obstacle walls” in melamine sponge towards effective electromagnetic interference shielding applications. Mater. Des. 2019, 182, No. 108029.
(12) Song, W.-L.; Cao, M.-S.; Lu, M.-M.; Bi, S.; Wang, C.-Y.; Liu, J.; Yuan, J.; Fan, L.-Z. J. C. Flexible graphene/polymer composite films in sandwich structures for effective electromagnetic interference shielding. Carbon 2014, 66, 67−76.
(13) Guo, Z.; Ren, P.; Zhang, F.; Duan, H.; Chen, Z.; Jin, Y.; Ren, F.; Li, Z. Magnetic coupling N self-doped porous carbon derived from biomass with broad absorption bandwidth and high-efficiency microwave absorption. J. Colloid Interface Sci. 2022, 610, 1077−1087.
(14) Flik, H.; Avan, A. A.; Tokatl, Z. F. A Review on Colorimetric Sensing of Tumor Markers Based on Enzyme-Mimicking Nanomaterials. Curr. Med. Chem. 2021, 28, 6123−6145.
(15) Zhang, L.; Wang, L.; Wang, Q.; Ding, W. Dielectric, magnetic, and microwave absorbing properties of Ag-plated terapod-like ZnO whiskers. Mater. Sci. Eng., B 2020, 262, No. 114682.
(16) Ji, R.; Cao, C. Cobalt ferrite sphere-coated buckhorn-like barium titanate: Fabrication, characterization, its dielectric resonance, and microwave attenuation properties. J. Appl. Phys. 2014, 116, 144106.
(17) Liu, Y.; Liu, X.; E, X.; Wang, B.; jia, Z.; Chi, Q.; Wu, G. Synthesis of MnxOy@C hybrid composites for optimal electromagnetic wave absorption capacity and wideband absorption. J. Mater. Sci. Technol. 2022, 103, 157−164.
(18) Guo, R.; Shen, L.; Wang, H.; Lim, Z.; Lu, W.; Yang, P.; Ariando; Gruverman, A.; Venkatesan, T.; Feng, Y. P.; Chen, J. Tailoring Self-Polarization of BaTiO3 Thin Films by Interface Engineering and Flexoelectric Effect. Adv. Mater. Interfaces 2016, 3, 1600737.
(19) Li, H.; Guo, Y.; Wu, G.; Ji, G.; Zhao, Y.; Xu, Z. J. Interface Polarization Strategy to Solve Electromagnetic Wave Interference Issue. ACS Appl. Mater. Interfaces 2017, 9, 5660−5668.
(20) Ameer, S.; Gul, I. H. Influence of Reduced Graphene Oxide on Effective Absorption Bandwidth Shift of Hybrid Absorbers. PLoS One 2016, 11, No. e0153544.
(21) Rahman, M. T.; Vargas, M.; Ramana, C. V. Structural characteristics, electrical conduction and dielectric properties of gadolinium substituted cobalt ferrite. J. Alloys Compd. 2014, 617, 547−562.
(22) Gutic, S.; Dobrota, A. S.; Gavrilov, N.; Baljozovic, M.; Pasli, I. A.; Mentus, S. V. Surface charge storage properties of selected graphene samples in pH-neutral aqueous solutions of alkali metal chlorides-particularities and universalities. Int. J. Electrochem. Sci. 2016, 11, 8662−8682.
(23) Garadkar, K. M.; Patil, A. A.; Hankare, P. P.; Chaté, P. A.; Sathe, D. J.; Delekar, S. D. MoS2: Preparation and their characterization. J. Alloys Compd. 2009, 487, 786−789.
(24) Ferrari, A. C.; Robertson, J. Resonant Raman spectroscopy of disordered, amorphous, and diamondlike carbon. Phys. Rev. B 2001, 64, No. 075414.
(25) Choi, W.; Lahiri, I.; Seelaboyina, R.; Kang, Y. S. Synthesis of Graphene and Its Applications: A Review. Crit. Rev. Solid State Mater. Sci. 2010, 35, 52−71.
(26) Yoon, D.; Son, Y.-W.; Cheong, H. Strain-Dependent Splitting of the Double-Resonance Raman Scattering Band in Graphene. Phys. Rev. Lett. 2011, 106, No. 155502.
(27) Havener, R. W.; Zhuang, H.; Brown, L.; Hennig, R. G.; Park, J. Angle-Resolved Raman Imaging of Interlayer Rotations and Interactions in Twisted Bilayer Graphene. Nano Lett. 2012, 12, 3162−3167.
(28) Lee, J.-U.; Park, J.; Son, Y.-W.; Cheong, H. Anomalous excitonic resonance Raman effects in few-layered MoS 2. Nanoscale 2015, 7, 3229−3236.
(29) Liu, Y.; Liu, X.; E, X.; Wang, B.; jia, Z.; Chi, Q.; Wu, G. Technology. Synthesis of MnxOy@C hybrid composites for optimal electromagnetic wave absorption capacity and wideband absorption. J. Mater. Sci. Technol. 2022, 103, 157−164.
(30) Karlovets, D.; Potylitsyn, A. Universal description for different types of polarization radiation. arXiv preprint arXiv:0908.2336, 2009, DOI: 10.48550/arXiv.0908.2336.
(31) Worsley, M. A.; Kucheyev, S. O.; Mason, H. E.; Merrill, M. D.; Mayer, B. P.; Lewicki, J.; Valdez, C. A.; Suss, M. E.; Stadermann, M.; Pauzauskas, P. J.; Satcher, J. H.; Jr.; Bienner, J.; Baumann, T. F. Mechanically robust 3D graphene macroassembly with high surface area. Chem. Commun. 2012, 48, 8428−8430.
(32) Cheng, B.; Zhao, J.; Xiao, L.; Cai, Q.; Guo, R.; Xiao, Y.; Lei, S. PMMA interlayer-modulated memory effects by space charge polarization in resistive switching based on CuSCN-nanopyramids/ZnO-nanorods pn heterojunction. Sci. Rep. 2015, 5, 1−9.
(33) Zhang, W.; Wei, L.; Ma, J.; Bai, S.−L. Exfoliation and defect control of graphene oxide for waterborne electromagnetic interference shielding coatings. Composites, Part A 2020, 132, No. 105838.
(34) Khan, R.; Khan, Z. M.; Aqeel, H. B.; Javed, S.; Shatqat, A.; Qazi, I.; Basit, M. A.; Jan, R. 2D nanosheets and composites for EMI shielding analysis. Sci. Rep. 2020, 10, 21550.
(35) Prasad, J.; Singh, A. K.; Shah, J.; Kotnala, R. K.; Singh, K. Synthesis of MoS2-reduced graphene oxide/Fe3O4 nanocomposite for enhanced electromagnetic interference shielding effectiveness. *Mater. Res. Express* 2018, 5, No. 055028.

(36) Guo, A.-P.; Zhang, X.-J.; Qu, J.-K.; Wang, S.-W.; Zhu, J.-Q.; Wang, G.-S.; Guo, L. Improved microwave absorption and electromagnetic interference shielding properties based on graphene−barium titanate and polyvinylidene fluoride with varying content. *Mater. Chem. Front.* 2017, 1, 2519−2526.

(37) Ding, X.; Fan, G.; Huang, Y.; Wang, J. Preparation and characterization of an effective microwave absorbent: RGO-MoS2-Fe3O4 nanocomposite. *J. Mater. Sci.: Mater. Electron.* 2021, 32, 9640−9649.

(38) Prasad, J.; Singh, A. K.; Tomar, M.; Gupta, V.; Singh, K. High-efficiency microwave absorption and electromagnetic interference shielding of Cobalt-doped MoS2 nanosheet anchored on the surface reduced graphene oxide nanosheet. *J. Mater. Sci.: Mater. Electron.* 2020, 31, 19895−19909.

(39) Zahid, M.; Nawab, Y.; Gulzar, N.; Rehan, Z. A.; Shakir, M. F.; Afzal, A.; Abdul Rashid, I.; Tariq, A. Fabrication of reduced graphene oxide (RGO) and nanocomposite with thermoplastic polyurethane (TPU) for EMI shielding application. *J. Mater. Sci.: Mater. Electron.* 2020, 31, 967−974.

(40) Shakir, M. F.; Abdul Rashid, I.; Tariq, A.; Nawab, Y.; Afzal, A.; Nabeel, M.; Naseem, A.; Hamid, U. EMI Shielding Characteristics of Electrically Conductive Polymer Blends of PS/PANI in Microwave and IR Region. *J. Electron. Mater.* 2020, 49, 1660−1665.

(41) Shakir, H. M. F.; Tariq, A.; Afzal, A.; Abdul Rashid, I. Mechanical, thermal and EMI shielding study of electrically conductive polymeric hybrid nano-composites. *J. Mater. Sci.: Mater. Electron.* 2019, 30, 17382−17392.

(42) Zhang, H.-B.; Yan, Q.; Zheng, W.-G.; He, Z.; Yu, Z.-Z. Tough Graphene−Polymer Microcellular Foams for Electromagnetic Interference Shielding. *ACS Appl. Mater. Interfaces* 2011, 3, 918−924.

(43) Guo, A.-P.; Zhang, X.-J.; Wang, S.-W.; Zhu, J.-Q.; Yang, L.; Wang, G.-S. Excellent Microwave Absorption and Electromagnetic Interference Shielding Based on Reduced Graphene Oxide@MoS2/ Poly(Vinylidene Fluoride) Composites. *ChemPlusChem* 2016, 81, 1305−1311.

(44) Liang, J.; Wang, Y.; Huang, Y.; Ma, Y.; Liu, Z.; Cai, J.; Zhang, C.; Gao, H.; Chen, Y. Electromagnetic interference shielding of graphene/epoxy composites. *Carbon* 2009, 47, 922−925.

(45) Radon, A.; Łukowiec, D.; Kremzer, M.; Mikula, J.; Wlodarczyk, P. Electrical Conduction Mechanism and Dielectric Properties of Spherical Shaped Fe3O4 Nanoparticles Synthesized by Co-Precipitation Method. *Materials (Basel)* 2018, 11, 735.