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The journey of PDMS based nanocomposites for EMI shielding applications: from bench to translational research

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Abstract

The advancement in the field of electronics has allowed the miniaturization of electronic devices. Also, lightweight devices have become an integral part of our lives, especially concerning wearable gadgets. In this regard, researchers are exploring polymer nanocomposites as a potential candidate due to their inherent advantages over traditional metal for shielding electromagnetic radiation. In this journey, many polymers, ranging from thermoplastic/thermoset to intrinsically conducting polymers, were explored. Although few reviews have been published in this field, a comprehensive study on PDMS-based shielding material did not receive much attention. However, its use in electronics has increased significantly in the recent past, especially in biomedical devices. In the last decade, researchers have explored PDMS use for making a composite, coating material, and foam-like structures. In this review article, the journey of PDMS-based shielding materials has been highlighted alongside the potential applications targeted and the underlying mechanism of shielding. This comprehensive review focuses on the crucial role of functional nanoparticles that render PDMS composites conducting and make them likely candidates for EMI shielding applications. The importance of cure-kinetics and processing of PDMS-based composites is stressed here as it decides the final application like flexible gaskets to block radio leakage to reinforced sheets for structural applications.
1. Introduction

In recent years, many researchers have focused on miniaturizing electronic devices for different applications as medical devices, electronics, and others\(^1\). These devices generate (electromagnetic) EM pollution in the ambient. Thus, they can affect the working of nearby devices. Therefore, we need to protect the devices from surrounding EM pollution. Although metals prove to be efficient EM shields, they usually reflect the incoming radiation to the ambient atmosphere and are also prone to corrosion. Thus, researchers moved towards polymer-based composite for designing lightweight, easy to integrate, and adapt shielding materials for EM waves.

In a quest to design lightweight, easy to process, integrate and adapt with the current process lines, researchers have explored thermoset, thermoplastic, and rubber for preparing various functional composites\(^2, 3\). They have also explored blends of two polymers as they provide the advantage offered by individual components\(^4\). Also, one can achieve better EMI shielding by controlling the dispersion state of nanoparticles in the blend\(^5\). Many reviews have already discussed the role of polymer nanocomposite in EMI shielding\(^6, 7\), although the use of PDMS in EMI shielding application has not received much attention. In figure 1 we represent the use of PDMS-based various types of shielding materials.
PDMS, since its discovery by FS Kipping in 1901 has been used in various applications such as microfluidic\(^8\), oil separation, hydrophobicity, sensors, electromechanical devices\(^9\)-\(^12\), actuators, sound absorber\(^13\) etc because of its versatile nature. In recent years, many researchers are trying to develop flexible electronic skin using PDMS\(^14\). This review highlights the journey of PDMS-based EMI shielding materials in the last decade. This article is divided into three sections based on the various forms of PDMS that are being used currently, such as (a) foam-like structure (b) coatings (c) composites as depicted in fig 1.

Under the foam category we have consider the composite where porous structure is used as a filler and PDMS is infiltrated such that the porosity is maintained. Secondly, PDMS is also used as coating material for making hydrophobic surface and hence, finds application in making hydrophobic EMI shielding material. And lastly, PDMS-based composite for EMI shielding application have been discussed where PDMS is used as a matrix with some filler. Also, all the PDMS-based EM shielding research works are tabulated in table 1.

2. **Mechanism of shielding in general**
Figure 2: Interaction of incoming EM radiation with the shield.

2.1 EM matter interaction

The incoming EM waves can undergo absorption, reflection, and transmission after interacting with the sample, as depicted in fig.2. The mode of interaction majorly depends upon the nature of the material. In general, magnetic, dielectric, or conducting material plays a vital role in EMI shielding. The incoming EM waves interact in different ways and result in the following losses.

2.1.1 EM interaction with conducting material.

Most conductors like metals have loosely bound electrons; thus, they cancel the electric field inside the metals. Therefore, when the incoming EM waves interact with the metal, they tend to move the electrons. These electrons move so that they generate a cancelling field; thus, metals tend to reflect the EM radiation\textsuperscript{15}.

Apart from metals, the loss in other conducting materials like MWCNT and other nanowire is majorly due to conduction loss. Therefore, the incoming EM radiation tends moves the electron inside these materials thus, resulting in loss due to conduction. For conduction loss the particles should form a closed percolating network for the conduction of charges under the impact of EM waves. To get insights into the impact of conductivity on EMI shielding generally AC
conductivity response with varying frequency is studied as per the Jonscher’s power law given below\textsuperscript{16}.

\begin{equation}
\sigma_{ac} = \sigma(0) + \sigma(\omega) = \sigma_o + A\omega^n
\end{equation}

According to the law, the ac conductivity in a complex system like composite can be considered as the combination of resistor and capacitors. At low frequency, only resistive pathways show conductivity through tunnelling. As the frequency increases the capacitive pathways become conductive in nature therefore, conductivity increases through hopping.

Thus, the law gives insights into the charge transport mechanism in a system. The value of exponent $n$ can vary from 0 to 1 and is dependent on temperature and frequency both. It represents the capacitive pathways inside the system. With increasing conductive pathways inside the system, the value of $n$ decreases.

Nanomaterials with elongated geometry (like chains, rods and tubular shapes) tends to form percolating network at low concentration and hence, are favourable for EMI shielding application.

\subsection*{2.1.2 EM interaction with a dielectric material}

When EM waves interact with the dielectric material, it can result in loss due to multiple factors. The losses can be due to the generation of dipoles, an account of the interaction between the oscillating electric field and dipoles within the material\textsuperscript{17}.

\subsection*{2.1.3 EM interaction with magnetic material}

Magnetic material shows significant loss due to the absorption of EM waves. The incoming EM wave interacts with magnetic domains and tries to orient them in the applied field direction. With changing magnetic fields, these domains oscillate thus, providing loss due to resonance of domain wall\textsuperscript{18}. Also, there is a loss due to eddy current generation because of the interaction
between the electric and magnetic material\textsuperscript{19, 20}. But the magnetic losses become insignificant at higher frequency as their permeability decreases. Thus magnetic loss decreases drastically at higher frequency as defined by the Snoek’s limit\textsuperscript{21}.

2.2 Measurement of EMI shielding

The performance of EM shields to attenuate the incoming EM waves can be expressed using shielding effectiveness and reflection loss.

2.2.1 Shielding effectiveness (SE.)

Shielding effectiveness is defined as the logarithmic ratio of incident wave (Pi) power to transmitted wave (P\textsubscript{t}). It is expressed in terms of dB.

\[
SE_T (dB) = -10 \log \left( \frac{P_i}{P_t} \right) = SE_A + SE_R + SE_{MR} \tag{1}
\]

Where \(SE_T\) refers to total shielding effectiveness

\(SE_A\) refers to shielding effectiveness due to absorption

\(SE_R\) refers to shielding effectiveness due to reflection

\(SE_{MR}\) refers to shielding effectiveness due to multiple reflections.

Where \(SE_A, SE_R, SE_{MR}\) can be theoretically calculated using the following relations,

\[
SE_A (dB) = -8.68d \sqrt{\frac{\omega \sigma \mu_r}{2}} \tag{2}
\]

\[
SE_R (dB) = -10 \log \left( \frac{\sigma}{16\omega \varepsilon_0 \mu_r} \right) \tag{3}
\]

\[
SE_{MR} (dB) = 20 \log \left| 1 - 10^{-\frac{SE_A}{10}} \right| \tag{4}
\]

But experimentally we calculate shielding effectiveness in terms of S-parameters as follows:

\[
SE_T (dB) = |S_{12}|^2 \tag{5}
\]

\[
SE_R (dB) = |S_{11}|^2 \tag{6}
\]

\[
SE_A (dB) = SE_T - SE_A \tag{7}
\]

Where \(\omega\) is the angular frequency \((\omega = 2\pi f)\) where \(f\) is the frequency of oscillation.
\( \mu \) is the magnetic permeability, and \( \varepsilon \) is the electrical permittivity and \( d \) is the thickness of the material.

### 2.2.2 Reflection loss

Also, for the materials with magnetic permeability (\( \mu \)) and permittivity (\( \varepsilon \)), the shielding efficiency can be expressed in terms of reflection loss (RL) or reflection coefficient (RC).

\[
RL \text{ or } RC = 20 \log \left( \frac{Z_{in} - Z_o}{Z_{in} + Z_o} \right)
\]  

(8)

Where \( Z_o \) is the impedance for free space (\( Z_o = 377 \Omega \)), and \( Z_{in} \) is input impedance calculated as follow:-

\[
Z_{in} = Z_o \sqrt{\frac{\mu_r}{\varepsilon_r}} \tanh \left\{ \frac{2\pi f d}{c} \sqrt{\frac{\mu_r \varepsilon_r}{\mu_r + \varepsilon_r}} \right\}
\]  

(9)

Where \( c \) refers to the speed of light.

### 3. State of the Art

#### 3.1 PDMS based foam-like structure

Dongyi et al.²² prepared 3D graphene foam using CVD. They used nickel foam as a template over which graphene was grown. This 3D porous network was further infiltrated with PDMS under the vacuum. Thus, prepared composite showed superior conductivity of 6100 S/m with only 1.2 wt % graphene. Thus, prepared composite showed very high EMI shielding of around 40 dB at 0.25 mm thick sample.

Wu et al.²³ synthesized foam-like structure using two MXene. They fabricated MXene foam using sodium alginate. Thus, the prepared conductive foam was coated with a thin layer of PDMS. Therefore, a prepared foam-like structure showed high conductivity of 2211 S cm\(^{-1}\) and high average shielding efficiency (SE) of 70.5 dB. After 500 compression cycles, composite with 6.1 wt% MXene showed SE of 48.2 dB.

Nguyen et al.²⁴ prepared nanocomposite using ferrite (Fe\(_3\)O\(_4\)) particles intercalated MXene and graphene composite for multifunctional EMI shielding as shown in fig. 3. They dispersed
MXene in water with different concentrations (1%, 3% and 5%), followed by the addition of Fe₃O₄ nanoparticles (3 wt%). Then the formed solution was sonicated for 6 h to form homogeneous dispersion. Graphene foam (GF) was developed using CVD on a Ni substrate followed by Fe₃O₄@TiC₂Tx in nickel foam. After the addition of Fe₃O₄ particle, PDMS was introduced and cured. Finally, Ni was etched out using FeCl₃ solution. They achieved a high EM shielding value for 1 mm thick sample (SE= 80 dB in X-band and 77 dB in Ka-band) with 11.35 wt% Fe₃O₄@TiC₂Tx particle.

**Figure 3:** (a&c) Total EMI shielding of hence, prepared samples in X- and K-band respectively. (b&d) shows SEₐ, SEₐ, and SEₜ for different foams at 10.2 GHz and 27.5 GHz respectively. (e) Mechanism of EM wave interaction with the foam. [Reprinted (adapted) with permission from (Chem. Eng J., 2020, 393, 124608). Copyright (2020) Elsevier.]
Kong et al. synthesized high EM waves absorbing material using MWCNT and reduced graphene oxide (rGO). In the process, they first prepared a graphene oxide (GO) solution containing cobalt acetate (2mg/ml). The solution was freeze-dried for 24 h. Thus, obtained foam-like structure was further used to grow MWCNT using CVD. Hence, the prepared foam was coated with PDMS. The obtained foam shows SE value reaches 49 dB for 3.1 mm thick sample.

Yu et al. prepared Fe decorated porous carbon and graphene foam. They prepared Fe nanoparticle/ CNT network by carbonizing Fe(acac)$_3$ decorated zeolitic imidazole framework-8 (ZIF-8) at 950°C for 10 min under Ar atmosphere. Hence, the prepared composite shows shielding effectiveness (SE ~ 48 dB) and specific shielding effectiveness as 347.8 dB cm$^3$/g.

Lu et al. made a flexible CNT/PDMS sponge with a thickness 2 mm. They prepared a CNT sponge using a chemical vapour deposition method. They infiltrated PDMS under negative pressure for 30 min followed by curing at 55°C for 2 h. Thus, the prepared composite shows SE ~ 46.3 dB with only 1 wt% CNT content. PDMS was infiltrated into CNT sponge through vacuum impregnation.

Li et al. obtained thin layer graphite foam (GF) with controlled density (27.2-69.2 mg cm$^{-3}$) after carbonizing polyacrylonitrile foam as represented in fig.4. This is followed by the addition of PDMS, resulting in porous GF@PDMS structure with varying GF concentration from 15.9% to 31.7%. The prepared composite shows high conductivity (up to 34.3 S cm$^{-1}$) and low thermal conductivity (0.062-0.076 W m$^{-1}$K$^{-1}$) and high EMI SE (up to 36.1 dB) for 4.5 mm thick sample over the frequency range 8.2-18 GHz.
Figure 4: Preparation process, morphology, and microstructure of GF27.2 and 15.9% GF@PDMS. (a) Schematic illustration of the preparation process. Optical (b), SEM (c, d), and TEM (e, f) images of GF-27.2 and the corresponding SEM/EDX elemental mapping (g) of C, O, and N elements. Inset of (e) shows the SAED image of GF-27.2. Optical (h) and SEM images (i) of 15.9% GF@PDMS and the corresponding SEM/EDX elemental mapping.
(j) of C, O, N, and Si elements for the selected area in (i). [Reprinted (adapted) with permission from (ACS Appl. Mater. Interfaces 2018, 10, 48, 41707-41716). Copyright (2018) American Chemical Society.]

Kong et al.\textsuperscript{29} prepared covalently bonded carbon nanowire and graphene (CNWs/graphene) architecture using a bioinspired approach from polydopamine as an interface buffer fabrication of 3D macroscopic CNWs/G sponge. A thin layer of PDMS was coated by dipping foam into PDMS and curing at 150 °C for 1 h. The sponge shows excellent EMI shielding ability of (SE~36 dB) in X-band.

Song et al.\textsuperscript{30} used three-dimensional rGO foam modified with zinc oxide (ZnO NWs) to suppress EM waves. In a typical process, they first prepared rGO foam by using the freeze-drying method. Followed by the synthesis of ZnO nanowires using seed-mediated growth. They infiltrated PDMS using a vacuum. Thus, a porous EMI shield showed minimum RC (RCmin = −31.1 dB) for ZnO NWs/RGO foam/PDMS with 0.8 mg/mL RGO at 9.2 GHz for 5 mm thick sample.

Han et al.\textsuperscript{31} synthesized composite film using rGO and zinc by dispersing zinc in an acidic solution of GO. The composite film was dialyzed for 12 h to remove acidic impurities. The obtained gel was freeze-dried for 36 h to make the foam-like structure. PDMS was infiltrated, and the sample was cured at 80°C for 4 h. For preparing a multilayer stack, several such layers were used in combination. The composite shows SE ~ 23 dB. Also, they claimed that the composite film act as an absorber and reflector in X-band. For multilayer structure, they achieved SE ~ 47dB to 53dB.

Chen et al.\textsuperscript{14} used quartz fiber cloth (QFC) reinforced MWCNTs- carbon aerogel (QMCA) by chemical route. Thus, the prepared gel was freeze-dried to form the final porous structure. The
PDMS based composite was made through vacuum infiltration followed by curing at 90 °C for 30 min. Thus, the prepared composite showed EM shielding (SE~20 dB).

Chen et al. prepared lightweight graphene foam composite with a density of 0.06 g/cm$^3$. In a typical process, they have grown graphene over Ni substrate; after that thin layer of PDMS was coated over the foam. After PDMS coating Ni was etched out by dipping into HCl solution for 24 h. The prepared sample shows SE ~ 30 dB and specific shielding effectiveness of 500 dB cm$^3$/g in the frequency range (300MHz - 1.5GHz). The foam composite showed excellent SE even after bending for 10000 cycles.

3.2 PDMS based coating material

Wang et al. prepared layer-by-layer assembly using Fe$_3$O$_4$ and MWCNT over the cotton fabric to shield the EM waves. After the deposition of nanoparticles, the fabric was coated with PDMS. Thus, robust EM shielding material shows shielding effectiveness of $\approx$ 84.5 dB in X-band with 0.96 mm thickness and good thermal conductivity.

Luo et al. developed self-cleaning composite for EMI shielding application by coating PDMS over PP fabric as depicted in figure 5. They prepared composite in three steps; firstly, they prepare PDA coating over PP surface followed by chemically reducing Ag over PP fabric. The obtained fabric was coated with PDMS layer to provide hydrophobicity. The prepared composite showed high contact angle of $\sim$ 152.3°. The coated fabric shows high conductivity of 81.2 S.cm$^{-1}$ and SE~71.2 dB (refer figure 6).
Figure 5: Schematic for the fabrication of the superhydrophobic and EMI shielding PP/PDA/AgNPs/PDMS fabric. [Reprinted (adapted) with permission from (ACS Appl. Mater. Interfaces 2019, 11, 10883-10894). Copyright (2019) American Chemical Society.]
Figure 6: (a) The photograph showing the surface resistance of the composite fabric (PP/PDA/AgNPs-25%/PDMS-40). (b) Digital photo images displaying the composite fabric in a circuit, and (c) the LED light in the circuit maintaining its original brightness during the bending. (d–h) Photographs of contact, press, and departure processes of a 5 μL water droplet on the composite fabric surface. (i) CAs of various corrosive solution droplets on the composite surface. (j) Photograph of the liquid droplets of acid (dyed in red), alkali (dyed in pink) and salt (dyed in yellow) solution sitting on fabric surface. (k–n) Photograph exhibiting the self-cleaning behavior of the fabric surface. Note that the composite fabric used in all the tests is PP/PDA/AgNPs-25%/PDMS-40. [Reprinted (adapted) with permission from *ACS Appl. Mater. Interfaces* 2019, 11, 10883-10894). Copyright (2019) American Chemical Society.]

3.3 PDMS based nanocomposites
Zheng et al.\textsuperscript{35} used flaky iron (F-Fe) (20 wt% and 28 wt%) as a filler to prepare magnetorheological film by mixing F-Fe in dimethyl silicone oil as shown in figure 7 followed by the addition of silicone rubber (SR). This solution was thoroughly mixed using a mechanical mixer. After this, the solution was vulcanized at 40°C for 12 h. for preparing the aligned sample, the sample was cured under 200 mT magnetic field. They compared EM shielding performance of the prepared sample with a wax alternative. They got a minimum reflection loss (RL) value as -53.3 dB at 4.3 GHz for 4.3 mm (refer to figure 8).

**Figure 7:** (a) Physical mixing of flaked Fe particles in silicone rubber using combination of mechanical stirrer and ultrasonication (b) Crosslinking of silicone rubber with the help of crosslinker (c) Alignment of flaky Fe nanoparticle with the help of external magnetic field.

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Figure 8: 3D representations of calculated RL values of (a,d) F-Fe/paraffin, (b,e) I-F-Fe/MREF, and (c,f) A-F-Fe/MREF with 20 and 28 wt % loadings. [Reprinted (adapted) with permission from (Ind. Eng. Chem. Res. 2020, 59, 8, 3425-3437). Copyright (2020) American Chemical Society.]

Nallabothula et al.\textsuperscript{36} used two different processes, i.e. spin coating and compression moulding for dispersing MWCNT in PDMS matrix and studied the effect of the processing technique on the state of dispersion and EMI shielding performance of the hence prepared composite. They further formed a multilayer stack by dispersing Fe\textsubscript{3}O\textsubscript{4} particle and MWCNT with the optimized process. They found that compression moulding results in proper dispersion of MWCNT compared to spin coating. For the multilayer stack, they achieved SE $\approx$-28 dB for 0.9 mm thick sample.

Li et al.\textsuperscript{37} used cotton fibres (CTF) in addition to MWCNT to construct multiple interfaces in PDMS-based composite. They dispersed MWCNT in PDMS by ultrasonication in dichloromethane solvent, followed by degassing and post-curing at 80°C for 2 h. They found that the EM shielding effectiveness (SE) for the sample with 2 vol% and 3 vol% MWCNT
increases from \(\sim 16\) dB to \(\sim 30\) dB and \(\sim 20\) dB to \(\sim 41\) dB with the addition of 15 wt% CTF for 1.2 mm thick sample.

Zhu et al.\textsuperscript{38} prepared composite using cake-like flexible MWCNT/ graphene and PDMS. In a typical process, they oxidized MWCNT (OCNT) using Hummer’s method. Thus, the obtained OCNTs wet cake was freeze-dried and re-grinded. The OCNT/ graphene (G) were dispersed together and filtered using PTFE membrane, therefore, giving wet cake-like structure followed by drying at 60°C for 6 h and calcining at 1000°C for 2 h in Ar atmosphere. Finally, the system was infiltrated with PDMS using a vacuum. They found that composite with thermally treated graphene and OCNT shows SE \(\approx 68\) dB (X-band) for 1.2 mm thick sample.

Anooja et al.\textsuperscript{39} made composite using carbon black (CB) and reduced graphene oxide (rGO) in PDMS. They firstly solution mixed CB (2 wt\%,5 wt\%,10 wt\%, and 17 wt\%) in rGO (5 wt\% and 15 wt\%) solution and sonicated for 30 min. In the above-sonicated solution, PDMS was added and dried overnight. As obtained powder was compressed at 200°C for 20 min Hence, prepared composite with rGO 15 wt% and CB 17 wt% shows SE \(\sim 28\) dB in (8-18 GHz) range. The composite retains its EM shielding ability even after 1000 bending cycle.

Ou et al.\textsuperscript{40} developed PDMS-based composite using Galinstan (GaInSn) liquid metal. Firstly, they dispersed GaInSn into PDMS by physical mixing. They prepared two different samples with GaInSn:PDMS ratio as 1:4 and 1:8. The minimum reflection loss (RL) achieved using GaInSn/PDMS composite is \(-19.3\) dB at 14.8 GHz at a thickness of 2 mm. They also observed a shift in the RL peak from 14.8 GHz to 9.7 GHz after 30 % elongation.

Lee et al.\textsuperscript{41} prepared plaid pattern over composite made from bio- PDMS and BaTiO\textsubscript{3} (MBPBT). They made composite using MWCNT and MBPBT. Another solution made from (AgNWs) and Fe\textsubscript{3}O\textsubscript{4} nanoparticle was a deposit on MBPBT. The two composite films (with MWCNT and with AgNWs) were arranged in two different configurations (parallel and cross
configuration) Thus, the prepared composite showed a high EM shielding ability of 11 dB in cross configuration with 15 wt % BaTiO$_3$.

Bora et al.$^{42}$ prepared composite using soft magnetic Gd$_5$Si$_4$ nanoparticle and PDMS. Gd$_5$Si$_4$ nanoparticles were made from bulk composite by using an arc discharge method followed by ball milling for 2 h. Thus obtained (40 wt%) nanoparticles were dispersed in PDMS and cured in a vacuum oven at 60°C for 6 h. They tested EM shielding ability of the composite in Ka-band (12.4-18 GHz). The prepared composite showed SE ~ 69 dB for 6 mm thick sample.

Lin et al.$^{43}$ fabricated ultrathin nitrogen-doped graphene film by vacuum filtration followed by compression moulding. Thus, obtained graphene films were modified using ethylenediamine (EDA). Thus, compressed sheets were spin-coated over the PDMS substrate. For preparing wavy sheet-like structure, the GO sheets were transferred to the pre-molded PDMS substrate. Thus, the prepared sample with 6.6 µm thickness shows shielding effectiveness of 58.6 dB. They observed that the modified GO sheets showed better shielding effectiveness.

Li et al.$^{44}$ prepared highly conductive and robust 3D graphene/silver nanowires bi-continuous skeletons for EM shielding. They used the AgNWs and rGO bi-continuous network (3D bi-continuous network). With low filler concentration, 0.76 wt% possess high conductivities 10.6 S cm$^{-1}$. The sample shows SE~34.1 dB, hence, prepared sample shows excellent thermal conductivity and high compression strength.

Shao et al.$^{45}$ prepared stretchable nanocomposite using Fe$_3$O$_4$/CNT/PDMS composite film using in-situ grown Fe$_3$O$_4$ over CNT film using the solvothermal method. Thus, obtained Fe$_3$O$_4$ over CNT film was used to prepare PDMS composite in plain and wave-like structure (as shown in figure 9. Firstly, for fabricating planar composite, the Fe$_3$O$_4$/CNT film was coated over PDMS substrate and followed a thin layer coating of PDMS. They also prepared wrinkled film by coating over the pre-stretched film and releasing the stress. They found that with an
increasing number of layers, the shielding ability of the composite increases. For 5 layers, they achieved minimum RL ~ -41.3 dB at 14 GHz with a bandwidth of 10 dB (refer to figure 10).

**Figure 9:** (a and b) Schematic illustration of the fabrication of Fe₃O₄/carbon nanotube/poly(dimethylsiloxane) composites. (c) Cross-section of the carbon nanotube film. (d) Stretching–releasing process. (e) Test specimen for EM shielding test. [Reprinted (adapted) with permission from *ACS Appl. Nano Mater.* 2018, 1, 5, 2227-2236). Copyright (2018) American Chemical Society.]
Figure 10: (a) RL of multilayer flat Fe3O4/carbon nanotube/poly(dimethylsiloxane) composites. (b) RL of one-layer buckled Fe3O4/carbon nanotube/poly(dimethylsiloxane) composites for different prestrains. (c) RL of multilayer 100% prestrain buckled Fe3O4/carbon nanotube/poly(dimethylsiloxane) composites. (d) Schematic illustration of EM-wave interaction with a buckled Fe3O4/carbon nanotube/poly(dimethylsiloxane) composite. [Reprinted (adapted) with permission from (ACS Appl. Nano Mater. 2018, 1, 5, 2227-2236). Copyright (2018) American Chemical Society]

Xu et al.46 produced PDMS-based nanocomposite by direct infiltration into the interconnected rGO network. With the addition of graphene, the mechanical property of the composite increases. For composite with 3.07 wt%, they achieved high electrical conductivity (~103 Scm⁻¹), and the composite shows SE of 54 dB in X-band for 2 mm thick sample.
Li et al.\textsuperscript{47} used CNT film fabricated utilizing a CVD process followed by a solvothermal process to prepare CNT-Fe\textsubscript{3}O\textsubscript{4} composite with different concentrations of Fe\textsubscript{3}O\textsubscript{4}. Reduced graphene oxide (rGO) was deposited on the sample using the electrophoretic technique. Thus, prepared rGO-CNT-Fe\textsubscript{3}O\textsubscript{4} composite film was coated with a thin layer of PDMS and cured for 1 h at 100\degree C. The composite film showed minimum RL $\sim$ -50.5 dB at 16.3 GHz for four-layer (t = 1.42 mm) composite with the bandwidth of 5.7 GHz with (RL>10 dB).

Zhao et al.\textsuperscript{48} prepared PDMS/rGO/SWCNT (single-walled carbon nanotube) composite through backfilling the rGO/SWCNT aerogel. The composite showed excellent electrical conductivity of 1.2 S cm\textsuperscript{-1} and EM shielding effectiveness of 31 dB over X-band with low filler content (0.28 wt\%) for 2 mm thick sample.

Mordina et al.\textsuperscript{49} used Fe\textsubscript{3}O\textsubscript{4} embedded hollow carbon nanofiber (CNF) with PDMS to prepare efficient EM shield. In a typical process, they used co-electrospinning to prepare nanofiber from polyacrylonitrile/FeCl\textsubscript{3} and poly(methyl methacrylate) followed by carbonization at the higher temperature. Thus, obtained nanofibers were used to fabricate composite with PDMS. The composite with 25 wt\% carbon nanofiber (consisting of 5wt\% Fe\textsubscript{3}O\textsubscript{4}) gives RL $\sim$ -25 dB with absorption bandwidth of 4.33 GHz for 7.5 mm thick sample.

3.4 Others

Luo et al.\textsuperscript{10} prepared superhydrophobic and multi-responsive polypropylene fabric for EM shielding by depositing Ag nanoparticle over the ozone-treated fabric followed by spray coating with a mixture of Fe\textsubscript{3}O\textsubscript{4} and PDMS. The obtained fabric shows excellent conductivity (up to 108.8 S/cm) and EM shielding effectiveness (SE $\sim$ 56.1 dB) in X-band. The material shows excellent stability against sonication, abrasion and bending test.

Duan et al.\textsuperscript{50} prepared novel SiOC ceramic by pyrolysis of hyperbranched ferrocene-containing polysiloxane (HBPSO-VF) obtained as a product of reacting polysiloxane (PSO) with 1,1'-
Bis(dimethylvinylsilyl)ferrocene (VF) at 160°C for 2 h. Thus, the obtained product was ball-milled for 2 h and pellets were formed by cold press. The obtained pellets were pyrolyzed at 900°C in Ar atmosphere. The obtained ceramic was annealed at a different temperature from 1000 ~ 1450 °C in Ar atmosphere. The composite 1 wt% Fe and thickness of 2.5 mm calcined at 1100 °C showed RC ~ 36 dB.

Bayat et al. 51 studied the effect of PDMS coating on the EMI shielding ability of the Fe$_3$O$_4$ coated carbon fiber. They used electrospinning to fabricate polyacrylonitrile (PAN) based fiber filled with Fe$_3$O$_4$ nanoparticles in a typical process. Thus, obtained fibers were carbonized at 900°C. Thus, obtained samples were coated with PDMS. They found no significant effect of PDMS coating on the EM shielding ability of the composite (SE~23 dB) for t=0.25 mm).

| S.No. | Filler | SE/RL And SSE | Thickness (mm) | % loading | Type | frequency |
|-------|--------|---------------|----------------|-----------|------|-----------|
| 1.    | MXene$^{23}$ | SE=70.5dB | 2 mm | 6.1 wt % | Foam | X-band |
| 2.    | Fe$_3$O$_4$ intercalated MXene and graphene$^{24}$ | SE=80 dB | 1 mm | 11.35 wt % Fe$_3$O$_4$@ MXene | Foam | X-band |
|       |        | SE=77 dB     |                |           |      | K-band   |
| 3.    | MWCNT and GO$^{25}$ | SE=49 dB | T=2.1 mm | GO- 100 mg MWCNT- 41.3% to 91.4% | Foam | X-band |
| 4.    | Iron nanoparticle and MWCNT$^{26}$ | SE=48 dB | T=1 mm | MWCNT(0, 41.3, 64.4, 84.2, 91.2) wt% | Foam | X-band |
| No. | Sample Description | SE (dB) | T (mm) | Material | Frequency Range |
|-----|--------------------|---------|--------|----------|----------------|
| 5.  | MWCNT             | 46      | 2      | Foam     | X-band         |
| 6.  | Graphite foam     | 36.1    | 4.3    | Foam     | 8.2-18 GHz     |
| 7.  | Carbon nanowire (CNW)/graphene | 36 | 1.6 | Foam | X-band |
| 8.  | rGO/ZnO (nanorods) | 27.8    | 4.8    | Foam     | X-band         |
| 9.  | GO aerogel film/ZnO | 43      | 1.5    | Foam     | X-band         |
| 10. | Quartz fiber cloth/MWCNT | 20      | -      | Foam     | X-band         |
| 11. | Graphene foam     | 30      | 1      | Foam     | 300MHz - 1.5GHz |
| 12. | Fe$_3$O$_4$ and MWCNT over cotton fabric | 84.5 | 0.96 | Coating | X-band |
| 13. | Ag coated PP fabric with PDMS layer | 71.2 | 1.3 | Coating | X-band |
| 14. | Flaky iron and PDMS | -53.3  | 4.3    | Composite | 2-18 GHz |
| 15. | MWCNT$^{36}$ | $SE \approx -28$ dB | $T=0.9$ mm | Ag coating (5-50%) | Composite | 12-18 GHz |
|-----|--------------|------------------|----------|------------------|-----------|-----------|
| 16. | Cotton fabric and MWCNT$^{37}$ | $SE=41$ dB | $T=1.2$ mm | 20 wt% Fe | Composite | X-band |
| 17. | MWCNT/graphene$^{38}$ | $SE=68$ dB | $T=1$ mm | 3 wt% MWCNT | Composite | X-band |
| 18. | Carbon black and rGO$^{39}$ | $SE=28$ dB | $T=2$ mm | CTF- 15 vol% | Composite | 8-18 GHz |
| 19. | GaInSn with PDMS$^{40}$ | $RL=-19.3$ dB at 14.8 GHz | $T=2$ mm | MWCNT-3 vol % | Composite | 2-18 GHz |
| 20. | BaTiO$_3$$^{41}$ | $SE=11$ dB | 200-250 µm | MWCNT-2wt% | Composite | X-band |
| 21. | Gd$_5$Si$_4$ and PDMS$^{42}$ | $SE\approx 69$ dB | $T=1$ mm | CB-2,5,7,10,15,17,20 wt % | Composite | (12.4-18 GHz) |
| 22. | N-doped graphene$^{43}$ | $SE=58.6$ dB | $T=9.35$ µm | rGO-10, 15, 17 wt% | Composite | X-band |
| 23. | Silver nanowire/rGO$^{44}$ | $SE=34.1$ dB | $T=2$ mm | GaInSn:PDMS 3.3:2 and 3.3:4 | Composite | X-band |
| 24. | Fe$_3$O$_4$ over MWCNT$^{45}$ | $RL=41.3$ dB at 14 GHz | $T=2$ mm | BaTiO$_3$- 15 wt% | Composite | 2-18 GHz |
| 25. | Graphene$^{46}$ | $SE=54$ dB | $T=2$ mm | Gd$_5$Si$_4$ -40 wt % | Composite | X-band |
|   | Material Description | RL (dB) | T (mm) | SE (dB) | RL (dB) | T (mm) | SE (dB) | RC (dB) | T (mm) | SE (dB) | RL (dB) | T (mm) | Notes |
|---|----------------------|---------|--------|---------|---------|--------|---------|---------|--------|---------|---------|--------|-------|
| 26. | rGO-Fe₃O₄ grown over MWCNT | -50.5 | 1.42 | 31 | -25 | 7.5 | 56.1 | -36 | 2.5 | 23 | 0.7 | Composite 2-18 GHz |
| 27. | rGO/SWCNT | 31 | 2 | - | 2 | 2 mm | AgNWs-0.43wt% | Composite X-band |
| 28. | Fe₃O₄ embedded hollow CNF | -25 | 7.5 | 31 | - | 7.5 | Ag-5%, 10%, 15%, 20%, 25% | Other X-band |
| 29. | Ag and Fe₃O₄ | 56.1 | 7.5 | 36 | 2.5 | Graphene-3.07wt% | Other X-band |
| 30. | SiOC fibers | - | 2.5 | 36 | 2.5 | | | |
| 31. | Fe₃O₄ coated carbon fiber | 23 | 0.7 | - | 0.7 | With 0.2, 0.3, 0.4, 0.5 and 0.06M FeCl₃ | Other X-band |

Table 1: Summary of all the PDMS-based nanocomposite used for EMI shielding.

4. Perspective

The journey of PDMS-based composites from bench to translational research for EMI shielding application is reviewed here in detail. In the last decade, various forms of PDMS-based nanocomposites for the targeted application have been designed to cater to different applications like flexible gaskets, reinforced sheets, encapsulating critical biomedical devices, contact lenses etc. In addition to making various components, PDMS-based coatings over porous structures obtained by carbonization of various foams resulting in foam-like structures have also been attempted. Since, PDMS is transparent to EM waves, various functional nanoparticles have been added to render the composites with key properties.
attributes like conducting, magnetic and/or dielectric properties. However, the presence of sulphur inhibits the catalytic activity of Pt-based catalyst; therefore, in most cases, researchers have used tin-based catalyst for crosslinking, which is key to realizing the exceptional properties that PDMS has to offer\textsuperscript{52}. This further leads to preparing composite using hydrosilylation crosslinking mechanism, which is more environmentally friendly. Recently, researchers have also explored click chemistry to enhance, the crosslinking in PDMS.

From the current state-of-the-art literature, it is well understood that PDMS-based foams are the most popular choice when it comes to designing lightweight EMI shielding materials. Of the various particle systems, 2D nanomaterials (like MoS\textsubscript{2} and WS\textsubscript{2} and other) need to be explored as the huge specific surface area of these 2D nanomaterials can help shield through multiple internal reflections/scattering. In addition, nanoporous materials like metal-organic framework (MOF) and covalent organic framework (COF) when blended with PDMS and offer a range of functional particles that can be further explored for EMI shielding materials. Due to their high thermal stability, these polymer composite can be used to shield electronic devices operating at high temperature and in addition can also dissipate heat, thereby protecting the precise electronics. More research should be focused on enhancing the shelf life of these composites by incorporating functional particles that can block both UV as well as EMI.

The use of PDMS based nanocomposite are more favourable as PDMS is biocompatible and hence, less harmful to the environment. Such, nanocomposite can find their potential use in a healthcare industry. Also, owning to its high thermal stability these nanocomposites can find their application in devices operating at higher temperatures. Also, these composite or coatings are mostly demanded in moisture sensitive equipment as PDMS shows very high hydrophobicity.
This review article highlights the journey of PDMS-based composites designed for EMI shielding applications. Various case studies have been presented to help guide the researchers working in this area from both industry and academia. Given the surge in the operating frequencies (higher GHz), more research should focus on PDMS based composites with hybrid and/or core-shell structures, multi-layered architectures and both rigid/soft foam-like structures.

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