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Development of Electromagnetic Interference shielding materials over time: A review

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ABSTRACT

Excess usage of electronic devices procures electronic noise, Electromagnetic Interference (EMI) and radio-frequency interference (RFI) which could reduce the life-time of electronic components and disturb the performance of delicate devices. Metals can serve as the EMI shield, but their heavy weight, corrosion response and poor meting out conditions limit their usage in EMI shielding applications. The development of EMI shielding materials leads to certain types of nanocomposites, polymer-based nanocomposites that have better properties as compared to the conventional EMI shielding materials. The emergence of polymer-based nanocomposite replaced the conventional EMI materials due to attractive properties such as flexibility, light-weight, corrosion protection and easy processing. In this review, we will critically study different EMI materials such as metallic, nanocomposites and polymer-based nanocomposite with inclusion of different conductive fillers like metals, metallic nanoparticles (NPs), metal-oxide, CB, CF, CNF, CNTs, graphene, graphene-oxide and dielectric/ magnetic such as TiO_2, $BaTiO_3$ / γ-Fe_2O_3, $BaFe_{12}O_{19}$, $Ni_{0.8}Zn_{0.2}Fe_2O_4$.

Keywords: Shielding theory, polymer nanocomposites, EMI shielding effectiveness

1. Introduction

These days, electronics is an emerging source in information, telecommunication and signal processing [1], and with the progression of technology sideways with more efficient electronic devices and miniaturization[2] [3], electromagnetic pollution became a staid problem of environs [4] which is termed as Electromagnetic Interference (EMI) [5]. The development of scientific devices, whether for commercial or military equipment bring a threatening condition on electromagnetic pollution [6] and unwarranted radiations that will have direct influences on nervous, endocrine system and also lurking humans' health [4]. A survey made on the extensive exposure of RF radiations by the mobile phones and linked devices on users and commenced that they are suffering from cancer, neurological disorders and cardiovascular problems. Electromagnetic radiations not only influencing the humans' health but also undesirably reduces the performance and lifetime of the delicate electronic components [3] [7]. Therefore, shielding protection is needed to reduce the electromagnetic pollution and effectively isolate the devices from EMI [3, 8]. Thus, EMI shielding materials are keenly desirable [9].

EMI shielding is the marvel which works on the mechanism of reflection and absorption by the material to avert the penetration of Electromagnetic radiations into the devices [6, 10, 11]. An effective shielding material is one which has the tendency to reduce the emission of undesirable radiations along with absorption of the stray signals. So, necessary conditions for a shield are the ability to reflect radiations by the interface due to mismatch of impedance, absorption of signal

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by electro-magnetic dipoles of the shield, and also multiple reflection which is not so important but it contributes to EMI process. From this aforementioned characteristics, high conductivity plays vital role in the process of reflectivity and absorption [12].

In order to resolve this subject meritoriously, a lot of attention is made to fabricate the high performance shielding materials over the broad-band frequency region to reduce the harmful effects generated by EMI [1, 13]. Metal-based EMI materials had been discovered because of their high conductivity, permeability and owning high shielding effectiveness (SE). However, they are heavy in weight, brittle, poor diffusion, high corrosion response and difficult meting out, make them unsuitable as shielding material [6, 14]. In the modern age of nanotechnology and nanomaterials, major concerned has been inclined to the growth of thin, lightweight, flexible, corrosion resistant, cost effective, easily process materials with wide band of absorption frequency [4, 5]. Thus, a variety selection of carbon nanomaterials like carbon nanotubes (CNTS), carbon fibres (CF), carbon black (CB), graphene nanoparticles and graphene oxide [1, 8, 9, 15] are favoured for EMI shielding application owing to high thermal and electrical conductivity, lightweight, flexibility [16, 17], better tensile strength aspects ratio, skin effects which linked with broad bandwidth [18-22] and better dielectric properties. Their composites blended with intrinsically conducting polymer (ICP) with or without the doping of transition metal oxides and magnetic nanoparticles had been explored since past decades [6, 12, 20, 23, 24].

The insertion of both conducting and magnetic nanomaterials in polymer has increase the absorption of incident EM waves [25]. Modern work is shifting towards the multidimensional assemblies along with the lower percentage of conducting fillers inside polymer matrix to obtained better SE by using the aforementioned materials [4, 12]. The manufacturing of such materials used to design polymer-based foams [1, 2, 4, 8, 9, 11, 20, 23, 26, 27], papers [6, 13, 28, 29] and composites [8, 16, 21, 30] were investigated for their electrical properties such as SE, structural, morphological, surface coatings and magnetic behaviour. Major research has been done in the field of EMI for different kind of shielding materials used and their synthesis techniques. . Table 1 summarised the different materials used and Table 2 shows the synthesis techniques.

Table 1. Different materials prepared by scientific community in their experimentation

EMI Shielding materials	Polydopamine decoration on 3D graphene foam	[1]
	CNT/PDMS films	[4]
	MCMB-MWCNTs	[6]
	PVDF-Graphite/MWCNT's	[7]
	PIGF's/ Graphene	[9]
	PC/PVDF/ $\alpha - MnO_2$ doped with crossed linked GO	[18]
	MWCNT's –Inverse spinel ferrites/PVDF	[25]
	Mesocarbon microsphere composites with Fe_3O_4	[28]
	AgNW /cellulose papers	[29]
	PANI/ $SrAl_{1.3}Fe_{10.7}O_{19}$/ MWCNTs	[30]
	Cement/ GO & Ferric fluid	[31]
	Silicone Rubber / CB	[32]
	NR/ CNT	[33]
	CB/ABS, CNF/ABS & CNT/ABS based materials	[34]

	PANI coated with GO, γ-Fe_2O_3 and $BaTiO_3$ material	[35]
	Epoxy Resin/ TAGA (axial) / TAGA (radial)/ TAGA (axial)	[36]
	PU/ Fe@FeO/ Fe@SiO_2	[37]
	MCMBs/MWCNTs with Fe_3O_4	[38]
	Epoxy/3D CNT sponge/ CNT	[39]
	PC/SAN/ Graphene/ Graphene@Ni	[40]
	poly (o-toluidine)/red mud	[41]
	EP/ rGO	[42]
	SBR/ f-MWCNT/IL	[43]
	PANI/PPY/$BaFe_{12}O_{19}$/ $Ni_{0.8}Zn_{0.2}Fe_2O_4$ materials	[112]

Table 2. Different techniques used by scientific society in their researches

References	Synthesis techniques of EMI shielding materials
[8]	Extrusion foaming process
[7]	Mortar pestle method
[13]	Spray vacuum filtration method
[6, 28]	Ball milling process
[29]	Dip coating process
[1, 3, 36]	Modified Hummer method
[5, 16]	In-situ polymerization
[30]	Sonochemical method by in-situ polymerization
[15, 18]	Melt mixing process
[44]	In-situ chemical oxidative polymerization
[17]	Solvent casting method

1.1 Electromagnetic Interference (EMI) Shielding Insight

EMI shielding is the phenomenon in which a part of the incoming EM waves is reduced by the shielding material. Generally, the competency of the shield is expressed in terms of the lessening of incoming power while the signal is propagated through the shield and expressed in terms of decibel (dB). A large amount of diminution of signals corresponds to the less value of transmittance, i.e., the attenuation, may be due to surface reflection, multiple surface reflection and absorption by the shield. The term "surface reflection" corresponds to the reflection by the surface charge carriers present in the shield; multiple-reflection relates to the reflection by the conducting stacks of the structure. Absorption is link-up with certain mechanism like dielectric losses, magnetization losses and dissipation of energy by the interfacial polarization of the shield [6, 12, 18, 28, 45, 46].

1.1.1 Theory of EMI shielding

The EMI shielding effectiveness is its efficiency to attenuate the intensity of EM signal before and after the shielding mechanism. It can also be described as the ratio of the field in the presence and absence of the shield. When EM waves are incident on the shielding material, a part of the signal is reflected and absorbed, and the rest of the signal is emerges out of the shield without being reflected or absorbed [47] . This is described in terms of the co-efficient and as well in basic phenomenon of shielding as shown in Fig. 1.

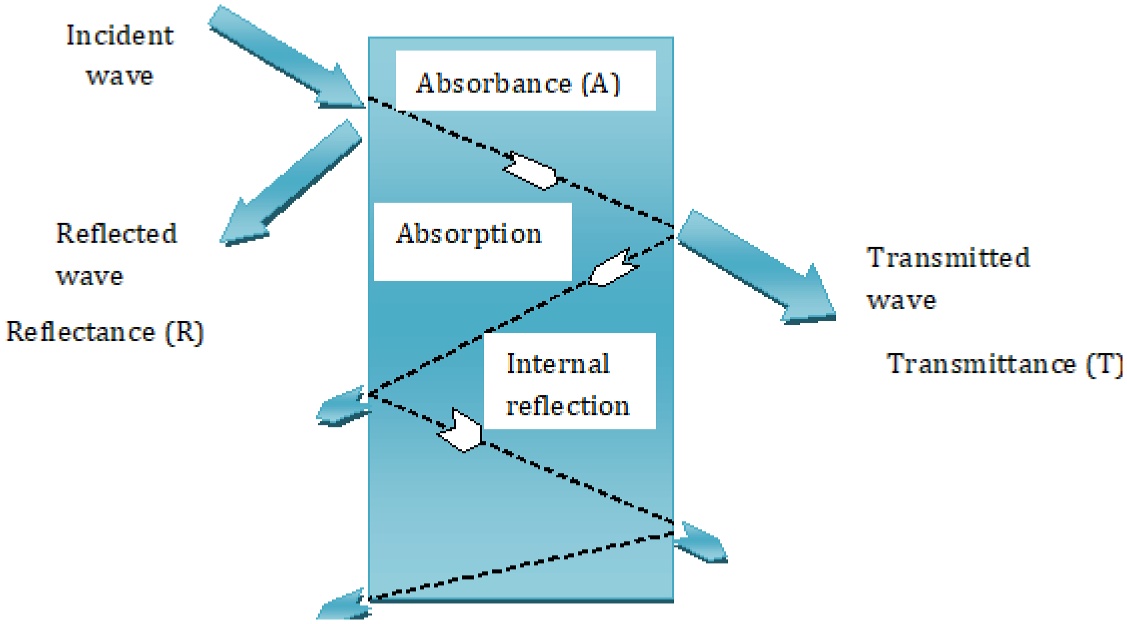


Figure 1. Mechanism of EM radiations phenomenon in terms of absorption, reflection, internal reflection and transmission

Shielding effectiveness (SE) can be expressed in terms of electric field (E), magnetic field (H) and power intensity (P), which can be expressed as the ratios of E, H and P before and after attenuation, which is expressed mathematically as follows:

$$SE = 20log\frac{E_I}{E_T} = 20log\frac{H_I}{H_T} = 10log\frac{P_I}{P_T} \text{ --------(1)}$$

where E_T, H_T, P_T, E_I, H_I and P_I are the transmitted and incident electric, magnetic fields and power intensities respectively. SE is the function of frequency so its measurement will be done in terms of frequency. The theory of Schelkunoff's showed that EMI SE can be described in terms of three shielding mechanisms namely, reflection (SE_R), absorption (SE_A) and multiple internal reflection (SE_M) losses, which is expressed mathematically as follows:

$$SE = SE_R + SE_A + SE_M \text{ -------------- (2)}$$

Reflection loss is the main mechanism in highly conducting medium, due to the misalliance of impedance of incident EM wave with the exterior of the shield which is expressed mathematically as follows:

$$SE_R = -10log_{10}\left(\frac{\sigma_T}{16\omega\varepsilon_0\mu_r}\right) \text{ -------------- (3)}$$

where σ_T is the conductivity of the shield and μ_r is the relative permeability of the shield. Equation (3) shows that reflection is directly proportional to the ratio of the conductivity and the permeability of shielding material [25, 48].

The process of absorption losses attributed from the falling-off the EM-wave passing through the shield of certain thickness. The reduction of the signal is due to dielectric and magnetic losses and not dependent on the nature of source field, which is expressed mathematically as follows:

$$SE_A = -8.86(t)\sqrt{\frac{\sigma_T \omega \mu_r}{2}} \text{ ------------- (4)} \qquad \text{where } \sigma_T = 2\pi f \varepsilon_0 \varepsilon$$

It is clearly understood from Equations 3 and 4 that SE_R depends on ($\frac{\sigma}{\mu}$) and SE_A on ($t\sigma\mu$). The dependency of these parameters shows that in solo-media (either magnetic / electric), the shielding is mainly due to absorption (magnetic) and reflection (electric) losses. But this mechanism is quite astonishing for composites that contains different micro / nano structures, produces variations in the local field which leads towards the generation of different consolidated loss parameters. So, shielding mechanism can be well understood with the help of scattering parameters in terms of permittivity (ε), permeability (μ), relative permittivity (ε_r) and permeability (μ_r), where:

$$\varepsilon_r = \varepsilon_r' - j\varepsilon_r'' \text{ and } \mu_r = \mu_r' - j\mu_r'' \text{ --------(5)}$$

Complex permeability and permittivity have a dependence on the frequency. Real parts ($\varepsilon_r{}'$, μ'_r) give information about the storage ability while the imaginary components (ε''_r, $\mu_r{}''$) related to the electric and magnetic power losses. Further, the absorption of EM waves can be increased by prolonging the routes of incident EM waves inside the shield through dissipation which is controlled only by the loss parameters [12, 17, 24, 49, 50]. Multiple reflection losses are because of continuous bouncing back of the EM signal between the two boundaries as shown in Fig. 1, which can be expressed as follows:

$$SE_M = 20 \log_{10} (1\text{-}10^{SE_A/10}) \text{ --------- (6)}$$

From Equation (6), it is clear that SE_M is dependent on SE_A. SE_M plays a major role in certain types of morphologies like porous structures and composite according to their designs and geometries. But it can be neglected for the designs in which the thickness is large, as the amplitude of the EM signals decay significantly and falls to minimum value when reaching at the second boundary. It means that when SE_A is high, the SE_M converts to a very low amount such that it can be ignored. Thus, standardized value of $SE_A \geq 10$ dB will make SE_M becomes irrelevant. SE_M is only significant at low thickness and frequencies in the range of kHz [14, 51]. For a shielding material, there are some ranges that gives information about the commercialization of the shielding material as shown in Table 3.

Table 3. Criterion for the commercialization of EMI shielding materials

SE range	Usage	% of attenuation
10 dB	-	little or no shielding
10-30 dB	commercial applications	99% of incident EM signal
≥30 dB	industrial & commercial applications	99.9% of incident EM signal

Furthermore, the standardized criterion suggested by different organization are applicable at the commercial, industrial, medical and scientific level, upon which, few standards are listed in Table 4 which operate at optimum conditions along with the nature, range and type of application which is being implemented upon the synthesized material [52].

Table 4. Standardized criterion for various specifications

Standard	Series	Specifications

CISPR	CISPR 16-1 CISPR 16-2 CISPR 16-3 CISPR 16-4	Measures radio frequency disturbance and immunity, type of EMC uncertainties
IEC	EN 61000-4-2 EN 61000-4-3 EN 61000-4-4 EN 61000-4-5 EN 61000-4-6	Involves different measuring and testing techniques regarding to Radiated, RF, EFI test
ISO	ISO 11452-1 ISO 11452-2 ISO 11452-3 ISO 11452-4 ISO 11452-5 ISO 11452-6 ISO 11452-7	Vehicles, components testing of electrical disturbance by emission of electromagnetic energy

1.2 EMI Shielding Materials - A Discussion on Different Structures

1.2.1 Metals as EMI shield

The superior conductors existed thus far are the metals. They have tendency to reflect, absorb and transmit the EM-waves compared to other materials such as rubber and plastics. Metals are highly preferred in certain applications due to their noble thermal and electrical conductivity. Primarily metals are potently used as radiators and grounds in certain machineries and electronic devices, because they have power to bleed off the static charge accumulated in devices and emerges out the heat generated during the processing of the high speed of machineries. Also, metal shields or enclosures also been used for hindering the transmission of high frequency EM signal. Most commonly used metals for fabricating the shields are mu-metal, an alloy of iron (14%), copper (5%), chromium (1.5%) and nickel (79.5%), and others such as Ag, Ni, Al, brass, metal coated plastics, metal-based fibres and conductive carbon graphite particles. A couple of obstructions enforced on the use of metals as shielding materials on account of high density, embrittled, low impact resistance and tends towards erosion that produces inter-modulation problems, which further receptive of rusty bolt effect of non-linearity. Likewise, the EMI shield produced by the non-identical metals are sensitive concerning of galvanic corrosion. That generates non-linearity and collectively reduces the SE of metals. Even so, Magnesium (Mg) enclosure or shields are favoured on the account of less weight and operative response across the whole frequency spectrum and suits better for reflection prior EMI shielding [14, 53].

Chi-Yuan Huang et al. [54] came up with a framework design in which a double layer metal was coated over carbon fibre (CF) reinforced acrylonitrile butadiene-styrene (ABS) composite for the protection against oxidation and de-bounding of copper with CF, as copper is thermally unstable and easily oxidized during the compounding process. This will degrade the EMI SE of copper coated CF/ABS composite. So, the electroless metals coating over the copper could increase chemical stability alongside an enhancement of conductivity property. The EMI SE of double layer electro-less metal coated/ABS composite was measured with flanged circular coaxial transmission line method calibrated in dBm. EMI SE would increase effectively to 65dBm as compared to the electro-less copper coated/ABS composite which had 37dBm. Another notable work was reported by Genaro A. Gelves et al. [55] who came up with an architecture of metal nanowire polymer nanocomposite. They synthesized copper nanowire polystyrene composite by the novel method of nanocomposite called miscible solvent mixing precipitation (MSMP). The segregated nanowire network with polystyrene were obtained with different volume concentration and beyond the threshold percolation limit which in results, increase the conductivity to $10^4 Sm^{-1}$. The EMI SE of copper NW/polymer nanocomposite exhibited more than 20 dB at X-band frequency with a volume concentration of only 1.3% of copper-NW.

1.3 Nanocomposite for EMI Shielding

A composite is a symmetry in which two or more materials combined together to form a new material that have discrete properties as compared to their entities. The matrix material can be a polymer, metal, glass, ceramic while the reinforced/filler material may be particles, fibres, flakes, tubes [14, 56, 57]. The functional features provided by the composites include stiffness, strength, ductility, energy absorption, damping and thermal stability. Though weight is not a function, it is a tremendously important part which more attention towards the lightweight structures were observed in the recent past years [58]. Scientific society are working over different nanocomposite to resolve the limitations offered by the metallic shielding materials. The developed nanocomposite should meet up the requirements for EMI shielding such as its architect design and feasibility. The individualities of EMI composite are reliant over the concentration and nature of filler with other salient features such as its conductivity, aspect ratio, geometry, magnetic and dielectric constants [14, 47, 59, 60]. For example, Yang Li et al. [2] studied the effect of composite material for electromagnetic wave absorption in ultra-thin carbon foams prepared by the pyrolysis of polyimide-graphene composite. Increment of wt. % of graphene helps in alleviating the foamed structure and aiding graphitization by the stress between filler and matrix, which resulting the distinct structure and thermal stability of the composite, showing higher average shielding effectiveness (SE) of about 24 dB compared to non-foamed corresponding item via the process of absorption over the X-band (8.2-12.4) GHz frequency region [9]. Further, Bin Shen, et al. made a comparative study on graphene films (G-films) and micro-cellular graphene foams (G-foams) in terms of EMI shielding over the broad spectrum of electromagnetic radiations (8.2-59.6) GHz. They investigated an average SE in the case of G-foams is about 26.3 dB although it has lower electrical conductivity as compared to its counter G-Films which has an average SE of (20.1) dB. The comparative results reveal that transformation of G-Films into G-Foam could improve the EMI-shielding because of the phenomenon of absorption due to improve internal multiple reflection at the boundaries.

In addition, Anisha Chaudhary et al. [6] came with MCMB-MWCNT'S composite to fabricate light weight and easily fold-able paper by a very simple and competent method. The synthesized composite had an excellent EMI-shielding in the X-Band (8.2-12.4) with very low density of 0.26 g/cm^3. They studied the electrical properties such as SE and reported the value from -31 dB to-56 dB with very low thickness ranges from 0.15 mm to 0.6 mm. The specific shielding effectiveness of the composite were recorded about -215 dB (cm^3/g), which was the highest value achieved as compared to metals and other low density carbon composites. Still, there are some problems like thickness dielectric and magnetic characteristic to be addressed, so, they progressed [28] by incorporating magnetic nanoparticles Fe_3O_4 with MCMB-MWCNT'S composite. The hybrid composite showed strong absorption due to the proper impedance matching because of the magnetic nanoparticles. Addition of magnetic nanoparticles raised the magnetic property, that in result, decrease the electrical conductivity which is further maintained by the graphitization of highly conducting MCMB at 2500°C. The maximum SE of 80 dB was obtained for the 20 wt. % loading of Fe_3O_4 with density of 0.50 g/cm^3 at thickness of 0.5mm. The excellent properties of the composite reveals that it is a novel kind of cover material for electromagnetic pollution control over high frequency.

More research on the new type of composite was done [29] where silver-nanowire-cellulose paper was prepared by an effective and simple process of dip coating. Their analysis on micro-structure concluded that the coating is dominantly performed over the cellulose paper but their partial presence inside the paper showed the decreasing density of AgNW with thickness of paper. Electrical conductivity of the analysed samples revealed that it increased from 0.34 to 67.51 Scm^{-1} with increment in number of dip coatings. Sample with highest electrical conductivity (67.51 Scm^{-1}) with 0.53 vol. % of AgNW shows the highest EMI-shielding of 48.6 dB at 1GHz and can be used as electrically conductive elements and EMI shields in advanced fields. Xing-Hua Li et al. [36] highly linked anisotropic graphene aero-gels by Freeze 3drying process. The synthesized composite exhibit different properties because of the incorporation of graphene alignment along radial and axial direction. Highly aligned graphene-epoxy composite had mechanical, electrical and excellent EMI-shielding efficiency on very small loadings of graphene. The epoxy composite with 0.8 wt. % thermally annealed GAs (TAGAs) showed EMI-shielding of 32dB due to multiple reflection and attenuation of electromagnetic waves. The aligned graphene-epoxy composite showed an increased shielding effectiveness of 32dB along radial direction and a lower value of about 25dB along axial direction.

The properties of the aforementioned composite are listed in Table 5 with host matrix, filler concentration and density thickness.

Table 5. Properties of different composite previously reported

Host matrix	filler	wt. % or vol. %	sample thickness	σ(S/cm)	EMI SE (dB)	Frequency (GHz)	Ref
PIGF's	Graphene	4 wt.%	24 µm	100	24	(8.2-12.4)	[9]
MCMB	MWCNT's	25 wt. %	0.15mm	11.2	-31	(8.2-12.4)	[6]
MCMB	Fe_3O_4	20wt.%	0.5mm	8.027	80	(8.2-12.4)	[28]
Cellulose Paper	AgNW aqueous solution	9.57wt. % 0.53 vol. %	-	67.51	48.6	1	[29]
Epoxy Resin	TAGA (axial) TAGA (radial) TAGA (axial)	0.8 wt. %	4mm	980 96 7	25 32 27	(8.2-12.4)	[36]

1.4 Conducting Polymers

The need of fabricating the best EMI shielding material motivated researchers to take advantage from polymers rather than metals. Metals have high density, heavy in weight and receptive of corrosion, making them unsuitable as EMI shields [61]. In EMI shielding, intrinsically conductive polymers (ICP's) matrices gained more attention because of their light-weight, easy processing and capability to transform their conductivity more efficiently [62]. Such polymers are preferred to make conducting composite by adding conducting fillers that would facilitate strongly to reduce the seam inside the shield [12]. Polymer matrix composites are very advantageous in EMI, because of their good electrical conductivity and less density. The properties of composite material are dependent on the nature and processibility of the polymer matrix. The non-conducting polymers such as polyvinyl alcohol (PVA), polyvinylidene fluoride (PVDF), polylactic acid (PLA), polyurethanes (PU), polyaniline (PANI), polyethyleneImine (PEI) and poly (o-toluidine) can be made conductive by doping the conducting fillers [14]. The filler might be metallic nanoparticles, metal nanowires, graphene, graphene oxide, carbon nanotubes (CNT's), carbon fibres (CF) or graphene nanoparticles graphite, added to fabricate lightweight and efficient polymer nanocomposite for EMI shielding based applications [14, 47, 59, 63, 64]. Despite of this, naturally ICP's are electrically good conductors and vastly used in EMI. Frequently used polymers for EMI involved poly aniline (PANI), poly vinyldine fluoride (PVDF) and polypyrrole (PPy) [14].

ICP's major property like easy tenability with other materials as compared to the metal and carbon-based material make them an outstanding participant for EMI shielding application. Also, the shielding phenomenon of ICP's is both reflection and absorption compared to metals and carbon materials that showed superior reflection mechanism. The intrinsic conductivity of these polymers in the region of microwave and radio wave (100MHz- 20GHz) also make them favourable for EMI. Review on the existing shielding material conveyed that a single / unadulterated material does not contain all properties that are obvious for shielding mechanism. These properties involve thickness, wide absorption band, volume, tuneable electric and EM characteristics with excellent environmental and thermal stability. So, a lot of effort has been done by researchers in this regard to take maximum benefit from the properties of polymer matrix by adding organic / inorganic fillers or insulating polymer as a guest depending on the type of polymer nanocomposite [10, 51, 53, 60, 65].

1.4.1 Polymer nanocomposite matrix

Polymer matrix is an ultimate choice since it exhibits exceptional properties such as flexibility, lightweight and superficial ability to process. Yet, polymer-based nanocomposite express peculiar traits than metals, as the basic need for EM application is the electrical conductivity of the desire material [52, 66, 67]. Growing their dielectric and magnetic properties is not an easy job due to the high surface area impact of the nanofiller that in result, cause the non-uniform dispersion which will alter the desire characteristics of polymer nanocomposite [68-70]. Hence, there must be a firm mastery on the modelling approach to get the desire requirements for the shielding material. The properties of the nanocomposite are dependent on the nature of host and the guest wt. / vol. % concentration and their mutual interaction. The function of the polymer matrix is to hold the fillers and provide major interaction throughout the system. Besides this, most of the polymers do not have good properties like electric, dielectric and magnetic properties and translucent to electromagnetic radiations. So, the electromagnetic properties of the polymer nanocomposite are based on the kind of fillers. Then, the choice of specific filler is dependent on the required properties of the material. For example, when electrical properties are the major concerned, then conductive filler are used, while for dielectric and magnetic properties filler with these properties are favoured. For that purpose, diverse nanomaterials with excellent electromagnetic properties are used since past decades [71-76].

1.4.1.1 Physical insight of polymer-based nanocomposites

The nanofiller added in polymer matrix has the ability to change the electromagnetic properties of the nanocomposites though the higher loadings of fillers had adverse impact on the composite. such as it might enhance its viscosity, decrease mechanical strength and offered difficulties in processing. Thus, during the process of fabrication, care should be taken with the concentration of the filler to minimize its percolation threshold while increasing the electrical conductivity of the composite [77-80]. The concentration of the filler has an influence on the conductivity though it will not increase abruptly but slowly. At a specific point of concentration, there would be sudden and sharp increase in the conductivity, but after this limiting point, the conductivity shows no alteration with more filler concentration. This limiting point is known as the ''diafiltration threshold''. At this point, the polymer become a conductor.

The mechanism of conductivity is well understood by the two supporting theories that are: (i) conductive channel theory; and (ii) tunnel effect theory.

The conductive channel theory supports in the case of higher filler concentration by making the infinite conductive path with the particles at the diafiltration threshold. In this situation, the charge carriers are free to move in the paths to make the whole system conductive. While the tunnelling effect is considered with lower filler concentration. In this case, there are large spacing between the particles, so the conductive paths are formed by the thermal vibration of the electrons when moving inside the system to make it conductive [81]. The issue of maintaining the desire properties of the nanocomposite is still a challenge but can be resolved by keeping in mind certain cases while merging filler with polymer: (i) single filler (granular /fibrous) take in with mono-polymer; (ii) dual filler (fibrous and granular) with single mono-polymer; (iii) single filler incorporated with dual immiscible polymers; and (iv) dual filler with blend of with dual immiscible polymers. The incorporation of the above mentioned cases leads to a better dispersion of the filler inside the polymer matrix that produce desire electrical conductivity at the diafiltration threshold [14].

1.5 Types of Polymer-based Nanocomposites for EMI Shielding

1.5.1 Metal/ metal-oxide-based polymer nanocomposite

Metal or metallic nanoparticles as a filler in the polymer-matrix gained more interest due to specific surface functionalization appropriate for diverse applications, principally in the fabrication of electronic devices [14, 82-84]. A.P Singh et al. [31] came forward with a novel composite of cement / graphene oxide / Ferro fluid for observing their shielding properties. The composite was prepared by the ball milling process. Graphene-oxide and Ferro fluid were added as filler in insulating cement matrix, that in result increase the interfacial polarization along with anisotropy energy of the composite. That leads to more scattering due to its mixed morphology which resulted in the SE_T of 46dB in the X-Band (8.2-12.4) GHz region. More on the volume fraction of GO plays a vital role for increasing its absorption properties with increase of GO content up to 30 wt. % with SE_A of 36 dB. The high value of EMI shielding is due to the absorption rather than reflection. The higher value of EMI SE shows that the material is the promising candidate of microwave absorber in RF range. Another novel kind of composite polyetherimide-Graphene@Fe_3O_4 with enhanced EMI SE along with good impedance matching were prepared by Bin Shen et al. [85] by the process of phase separation. The as-prepared composite was subjected to transform to microcellular structure using WVIP's method with very low density of about (0.28-0.40) g/cm^3. The improved EMI SE of the foams are due to the increased 10 wt. % of hybrid filler Graphene@Fe_3O_4 into polymer matrix. That will generate a good matching impedance due to microcellular structure and shown an improved EMI SE of about (14.3-18.2) dB over 8.2-12.4 GHz of frequency.

Metals and metal-oxides are highly preferable due to their unique properties like permittivity and permeability, so Jiahua Zhu et al. [37] used polyurethane (PU) nanocomposites (PNC) strengthen with core-shell Fe-silica NP's for shielding the electromagnetic waves. Core-shell structure NP's were prepared by the modified Stober method. Their TGA data obtained revealed that the silica coated NP's and their relevant PNC's are thermally more stable. The coated silica layer act as an insulation that will help in improving the resistivity of the magnetic nanoparticle which in result, increase the absorption bandwidth along with the reflection loss (RL) of the PNC's. Silica shell not only improved the aforementioned parameters but also help in minimizing the eddy current losses alongside in increasing the anisotropy energy of the PNC's. The PNC's architect contains two type of polymer reinforced composite that are, Fe@SiO_2 /PU and Fe@ FeO/PU with the 71 wt. % of the filler. These PNC's were prepared with different thickness like 1.0mm, 1.8mm, 2.3mm and 3.0mm and investigated over the X-band frequency spectrum for evaluating their behaviour with changing thickness. Maximum RL of about -20 dB had been obtained at the thickness of 1.8 mm with absorption frequency of 11.3 GHz for Fe@SiO_2 / PU. Same procedure were performed with the second composite with maximum RL of < -20 dB with minimum thickness of about 3 mm and the absorption frequency were recorded to be 3.3GHz.

Polymers are doped with different kind of nanofiller for enhancing their EMI efficiency. Ranjan Singh et al. [86] used conducting fillers liked metal oxides of iron, zinc, silicon, zirconium and titanium for their improved EMI properties in the non-conducting polymer matrix of PVA. The composites were synthesized by taking the concentration of metal oxides about 0.1, 0.5, 1.0, 5.0 and 10.0 wt. % by solvent casting process. The nanocomposite was analysed for their EMI shielding properties over the C (4-8 GHz) and X (8-12 GHz) band of frequency spectrum. The results showed that the maximum RL had been obtained with 10.0 wt. % loading of metal oxides. The maximum recorded values for Fe_2O_3 based composite were -38.85 dB at 10.4 GHz; for ZnO composite, -33.65 dB at 10.4 GHz; for SiO_2 based composite, -41.90 dB at 10.4 GHz; for ZrO_2 based composite, -24.90 dB at 11.0 GHz and for TiO_2 based composite, -32.90 dB at 9.76 GHz respectively. It was also concluded that the Fe_2O_3 and SiO_2 based composite were more thermally stable and mechanically reliable and found to be more attractive material for EMI shielding application with minimum cost.

The development of the novel and the best shielding material provoked scientist to look in several directions to take advantage from different combination and structures to obtain improved EMI SE. Such effort had been made by Yinju Chen et al. [87] so they came out with a novel kind of combination by taking the composite of PANI with graphene that is further decorated with the magnetic NP's of silver and nickel. The composite of PANI with graphene, graphene@silver and graphene@nickle were prepared by in-situ polymerization with loading of 0.5, 1.0, 3.0 and 5.0 wt. %. These composites were subjected to analysis of their electrical and EMI SE efficiencies and it was found that the best combination is of graphene@silver with electrical conductivity of about 20.32 S/cm with maximum SE_T of 29.33 dB at 1.5 GHz. It was also concluded that the addition of filler loading increases the SE and the main mechanism behind the SE was the absorption. So, graphene@silver is a promising candidate for EMI shielding application.

More on the progress in the field of EMI shielding materials, P.P Shital et al. [40] came with different strategy for getting the better results regarding to the EMI shielding. They used PC (polycarbonate) / SAN [poly (styrene-co-acrylonitrile)] blend with couple of graphene nanosheets layers that are further decorated with the nickel NP's. The growth of the nickel NP's on the graphene sheets were done by the uniform nucleation of the metal salts. For the localization of the nickel NP's on the PC phase of PC/SAN blends, a two-step methodology were adopted. The attenuation efficiency of the blends of PC/SAN with graphene and nickel decorated with graphene were analysed over the X and K_u band of the frequency spectrum. The integration of the graphene decorated with nickel NP's dramatically enhance the SE of the blends. Also, the annealing of the NP's in various environs also effects the SE efficiency of the blends, most importantly, an improved SE were observed for the case of H_2 atmosphere. For example, blend with G-Ni showed a total SE of about -29.4 dB as compared to its counterpart that contain only the graphene sheets which showed a total value of -13.6 dB with 3 wt.% loading of graphene at 18 GHz of frequency. Blends that were decorated with nickel NP's also showed an enhanced thermal transport and storage modulus properties in comparison of the blends with graphene only.

Further, S.P Gairola et al. [44] took advantage from the red mud that is available in bulk and it was a combination of different metal oxides such as Fe_2O_3, TiO_2, Al_2O_3, SiO_2, CaO and Na_2O. Due to the presence of iron oxide, it is very hazardous to the environment. So, they thought of a new way to dispose the mud. For that purpose, they prepared composite of POT / red-mud by oxidative-in/situ polymerization process. The addition of 50 % mud into POT reduced its magnetic properties. The EMI-shielding was measured in X-Band (8.2-12.4) GHz of frequency spectrum to be 8.2 dB. It was an attempt to find the application of red mud for controlling the EM pollution. The values of all the parameters like wt. %, EMI SE (dB) are tabulated in Table 6.

Table 6. EMI shielding properties of metal/metal-oxide-based polymer nanocomposite

Polymer matrix	metallic filler	wt. %	Thickness (mm)	EMI SE (dB)	Frequency (GHz)	Ref
Cement	GO & Ferric fluid	30	3.0	46	(8.2-12.4)	[31]
PEI	Graphene@Fe_3O_4	10.0	2.5	(14.3-18.2)	(8.2-12.4)	[85]
PU	Fe@FeO Fe@SiO_2	71	2.3 1.8	RL < -20 RL > -20	4.3 11.3	[37]
PVA	Fe_2O_3 ZnO SiO_2 ZrO_2 TiO_2	10.0	0.5	-38.85 -33.65 -41.90 -24.90 -32.90	10.4 10.4 10.4 11.0 9.76	[86]
PANI	Graphene@nickle Graphene@Ag	5.0	1.0	25 29.33	0.45-1.5 1.5	[87]
PC/SAN	Graphene Graphene@Ni	3.0	1.5	-13.6 -29.4	18	[40]
POT	red mud	50.0	-	8.9	(8.2-12.4)	[44]

1.6.1 Carbon-based polymer nanocomposite

The invention of the allotropic form of carbon and its nanostructure has opened new ways of progress in the field of science and technology from the biomedicine to automobile engineering. This progress not only confined to the aforementioned fields but also fascinates advantages in the field of EMI and aerospace [47]. The carbon-based polymer nanocomposite have the binary characteristics of flexibility from the polymer and the electrical conductivity from the fillers [14, 88, 89]. The next section is based on the discussion of the carbon-based nanostructures such as Carbon black (CB), Carbon nanotubes (CNTs), Carbon Fibres (CFs), Graphene and Graphene oxide.

1.6.1.1 Carbon Black reinforced polymer nanocomposite

Carbon Black is renowned as a filler that could be integrate into polymer matrix to obtain the desired characteristic regarding to the EMI SE. CB is obtained from the thermal decomposition of the Hydrocarbon during the gas phase process to get the small size carbon pigments [47]. CB as a filler has the potential to transform the electrical conductivity and antistatic properties of the polymer composite. Conductive form of the CB enhanced the σ of the polymer composite more effectively as compared to conventional CB. It is more often used due to its fair value of σ, chemical stability and slight density [14]. Owing to these properties, it found applications in the field of EMI shielding, EDS and antistatic protection agency [53, 90].

For example, M.H Al-Saleh et al. [91] studied the EMI SE characteristics of the HS-CB / PP composite over the X-band of the frequency spectrum. Composite exhibit experimentally calculated value of 43.8 dB at 12.4 GHz with the thickness of the plates 2.8 mm and 10 vol. %. They also investigated that the composite showed a considerable increase in the EMI SE of the composite with varying thickness and electrical conductivity of the composite. The major mechanism occurred while observing the EMI SE of the composite was the attenuation of the signal by the absorption than the reflection loss. Experimentally, they calculated the attenuation of the signal about 87 % of the incident signal. Results also revealed that the overall EMI SE of the composite obtained by experiment is greater as compared to the theoretically calculated that was 41.8 dB at 12.4 GHz. Raj K. Jani et al. [32] studied the effect of nanosized conducting CB on the silicone rubber elastomer matrix. They observed that percolation threshold achieved at 3 wt. % loading of CB on the composite and percolation zone occurred between (3-8) wt. % of CB loading. Further, they studied the EMI SE properties and revealed that the attenuation of the EM waves was observed to be 90 % with reflection loss > -10 dB over the X and K_u band of frequency with thickness of 1.7 mm and 2.7 mm respectively. They also investigated the EMI SE of the composite with different fillers loading and came to the conclusion that the SE of -40 dB achieved at 15 wt. % of CB with thickness of 2.8 mm at 8-18 GHz. They also tuned the SE of the composite to -15 dB to -40 dB with varying the thickness from 1 mm to 2.8 mm.

It is obvious that the polymer matrix reinforced with nanofiller of high aspect ratio display more SE as compared to the conventional micro-fillers. But it is very difficult to compare the EMI SE of the already existed polymer nanocomposite based on literature data. So, Mohammad H. Al-Saleh et al. [34] made an attempt to compare the EMI SE data of the different carboneous fillers such as high structured carbon black (HS-CB), CNF and MWCNT reinforced with acrylonitrile-butadiene-styrene (ABS). The nanocomposite was prepared under the same condition by the method of solution processing. They studied the effect of nanofiller loadings on EMI SE, electrical and dielectric properties. They concluded that among the three nanocomposite, CNT / ABS showed exceptional characteristics as compared to CB / ABS and CNF / ABS. For instance, at the nanofiller loading of 5 wt. % CNT / ABS nanocomposite showed an EMI SE of 2 times as of CNF / ABS and 7 times as of CB / ABS. It was also concluded that with only 2 wt. % loading of CNT, the EMI SE was recorded 20 dB. Further on enhancing the CNT wt. % to 15 the EMI SE approaches to 50 dB. No matter what kind of nanofiller, the dominant mechanism observed during the experimentation was absorption loss although reflection also present due to the mismatch between the incident wave and material impedance. Table 7 summarises these findings.

Table 7. EMI shielding properties of different carbon-based polymer nanocomposite

Polymer matrix	filler	Concentration (wt. % or vol. %)	thickness (mm)	σ(Scm^{-1})	EMI SE (dB)	Frequency (GHz)	ref
PP	HS- CB	10 vol. %	2.8	44.1	43.8	8-12	[91]
Silicone Rubber	CB	15 wt.	2.8	0.57	-40	8-18	[32]
ABS	HS-CB CNF CNT	15 wt. %	1.1	- 0.67 1.23	21 35 50	8-12	[34]

1.6.1.2 Carbon Nanotubes (CNTs) based polymer nanocomposites

The advancement in the field of nanocomposite started with the discovery of the carbon nanotubes. The man who discovered the CNT was Iijma and these CNTs find application in micro and nanodevices [14, 92]. CNTs are composed of graphene sheets that are rolled in the form of concentric cylinders that create the single walled carbon nanotubes (SWCNT) and multi-walled carbon nanotube (MWNT) [67]. Their schematic can be seen in Fig. 2.
CNT enchant nano size and novel structure due to their extraordinary individualities such as ultra-high young modulus (¥ 1-1.4 TPa), outstanding room temperature thermal conductivities of the order of 3000-6000 W/mk and very high electrical conductivity of ~ 106 Scm^{-1} [93]. Both types of CNTs possessed very high surface area and also very high aspect ratio. Owing to these surprising characteristics, CNTs are most preferable filler as compared to conventional fillers. These are used for achieving the desired properties in the composite by controlling the electrical threshold limit at very low loading with high value of σ. Hence, CNTs were used in the field of EDS and EMI shielding applications [47, 67, 94, 95].

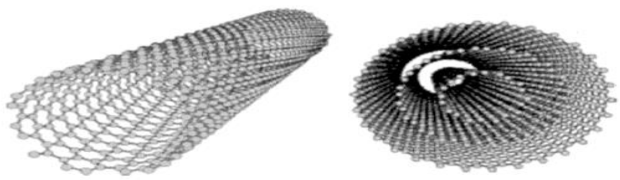


Figure 2. Schematic representation of CNTs

D.Lu et al. [4] took advantage of the flexibility, high conductivity along with the high aspect ratio of the CNTs and used them directly as EMI shielding films over the X-band frequency region. They used spongy CNT with ultra-low density of 10.0 mg/cm^3 and studied the EMI SE of about 54.8 dB at the thickness of 1.8 mm. They further increased the thickness of the CNT sponge from 0.77 mm - 2.15 mm and recorded EMI SE varied from 16.9 - 56.0 dB and also concluded that CNTs can be used as an ideal conductive network for the synthesis of nanocomposite. So, they tried to make a nanocomposite of CNT / PDMS films by simply infiltrating the PDMS over the pores of the spongy CNT with the 1.0 wt. % of CNT. Further, they investigated the composite for their EMI shielding properties and observed that composite still have an outstanding SE of 46.3 dB at thickness of 2.0 mm. More on, they bend and stretch the composite films for 1000 cycles and then again studied their shielding properties and came to conclusion that there would be no change occurred in SE. Due to these considerable properties, CNT / PDMS films found potential applications in wearable and portable electronic devices as flexible, light and ultra-high shielding material.

The design of new materials for the EMI shielding applications widespread these days and opens up many ways in this regard. Styrene-butadiene rubber (SBR) is an elastomer and commonly used in many applications due to its low cost. But its use is limited in EMI shielding due to poor electrical conductivity. But J. Abraham et al. [43] came with a novel kind of frame-work in which they used SBR/f-MWCNT composite by incorporating non-covalent functionalized MWCNT. The ionic liquid (IL) immerses in MWCNTs make it dispersible due to π- cation interaction and the results of TEM and FESEM also revealed the excellent dispersion of ionic liquid f-MWCNT in the SBR matrix. The shielding material with a thickness of 5 mm was prepared and the SE was recorded to be 35.06 dB. The analysis of the sample showed that the SE and the conductivity could be increased with changing the concentration of additives. The formation of interfacial poles inside the heterogeneous boundary of the interface plays considerable role for the absorption of the EM waves so that dominant process occurred here were absorption.

CNT segregation has the ability to develop the conductive paths and multiple interfaces inside the composite along with enhancing the flexibility. A stabilized composite of natural rubber (NR) with CNT had been prepared by Li-Chuan Jia et al. [33] and the composite shows high EMI SE after high deformation. They investigated the behaviour of the composite by long-term cycling test and came to the conclusion that the possession of the EMI SE more than 80 % after a very high deformation. The attribution of CNTs inside the NR cause the strong synergistic effect of EMI shielding and higher flexibility at the percolated threshold. The EMI SE of the composite recorded to be 43.7 dB at (8-12) GHz. This work is significant in high performance miniaturized and very compact flexible electronic devices [33]. K.K Halder et al. [7] worked on the designing of EMI shielding composite of MWCNT with graphite and insulating polymer such as PVDF. Composite were prepared by mortar-pestle method. The percolation threshold was achieved at 10 wt. % of graphite and 2 wt. % of MWCNTs. The dielectric and conductivity of the composite increased sharply at the threshold of the conductive fillers. Absorption and reflection of the radiations also increased and an EMI SE_T of 14.64 dB were recorded at (8-12) GHz of frequency spectrum.

Sourav Biswas et al. [25] worked on the physical view of the electrical properties of composite of Inverse-Spinel ferrite incorporated with CNT and insulating polymer of PVDF. Ferrite nanoparticles of Co, Ni and Fe were prepared by hydrothermal reaction technique. Further the preparation of polymer nanocomposite was done by the blend mixing technique in which PC/PVDF were mixed by 50/50 (w/w) with nanofillers. Then, they processed the compound with melt compounder with rotational speed of 60 rpm at 260 ^{0}C for about 20 min under the nitrogen gas environment. The concentration of nanoadditives and MWCNTs were taken to be 3 wt. % due to the diafiltration threshold and for achieving the better mechanical properties. The best EMI SE were achieved for the composite of PC/PVDF-MWCT and Fe_3O_4 about -31dB which was 19 % superior when compared with PC/PVDF-MWCT and $CoFe_2O_4$ and 24 % with PC/PVDF-MWCT and $NiFe_2O_4$. In addition, they changed the method of mixing the materials for only the iron ferrites; they mixed PC with Fe_3O_4 and PVDF with MWCNTs and achieved an EMI SE of about -35 dB and improved their electrical properties of DC conductivity, tangent loss and thermal conductivity. The purpose of the PC phase inside the iron ferrite to disperse the nanoadditives uniformly which account for a better thermal transport.

Reza Peymanfar et al. [30] came with notable work in which they synthesized hexaferrite nanoparticles along with doping of Al by the sol-gel method. Nanocomposites of $SrAl_{1.3}Fe_{10.7}O_{19}$ with PANI/MWCNTs had been prepared by sono-chemical in-situ polymerization process. The synthesized nanocomposites were characterized for its surface morphology, structure and magnetic properties using FE-SEM, XRD, FTIR and VSM. The shielding behaviour of the composite was analysed over K_u-Band (12-18) GHz with EMI-SE_T ~ -40.85 dB and reflection loss ~ -15.92 dB at 15.84 GHz and absorption loss of ~ -24.93 dB at 16.40 GHz with band-width of 1.66 GHz and 2.81 GHz, respectively. Another remarkable work done by Yu Chen et al. [39] in which they used the 3D CNT sponge architect with the epoxy nanocomposites to enhance the conductive network along with improving their mechanical properties such as flexural and tensile strength by using the minimum loading of about 0.66 wt. %. They observed the electrical conductivity of the 3D CNT sponge/ epoxy nanocomposites about 148 Sm^{-1} with the EMI SE of about 33 dB over the X-band of the frequency spectrum. They also prepared the sample with CNT with 20 wt. % and observed them for their electrical and mechanical properties and came to the conclusion that the 3D CNT sponge showed improved properties and could be an ideal candidate for resolving the issue of synthesis of the high performance EMI shielding material with spectacular mechanical properties. Table 8 summarised the findings.

Table 8. Behaviour of different CNTs based polymer nanocomposite

Polymer	additive	wt. %	Thickness (mm)	EMI SE (dB)	Frequency (GHz)	Ref
PDMS	Spongy CNT CNT	- 1.0	2.15 2.0	56.0 46.3	8-12	[4]
SBR	f-MWCNT/IL	1/ 10	5	35	3-18	[43]
NR	CNT	10	-	43.7	8-12	[33]
PVDF	graphite/MWCNT	10 /2	-	14.64	8-12	[7]
PVDF/PC PVDF/MWCNTs	Fe_3O_4/MWCNT $NiFe_2O_4$/MWCNT $CoFe_2O_4$/MWCNT Fe_3O_4/PC	3/3	-	-31 -24 -26 -35	8-12	[25]
PANI	$SrAl_{1.3}Fe_{10.7}O_{19}$/ MWCNTs	-	6.5	-40.85	12-18	[30]
Epoxy	3D CNT sponge CNT	0.66 20	2	33 24	8-12	[39]

1.6.1.3 Carbon fibre (CF) based polymer nanocomposite

The intermesh sheets of carbon atoms or graphene with the hexagonal structure constitute to the formation of carbon fibres (CFs) [63]. CFs are potentially used in many applications such as, the material of electrodes in batteries, sensors, additives, superconductors and the energy storing and converting electronic devices. The first man who came with the use of CF was Thomas Alva Edison. He used CF as the filament of light bulb which was prepared by carbonizing bamboo and cotton. After this great achievement, the scientific society dragged their attention towards the use of the CFs in researches as well in commercial applications. The conventional CF and carbon nanofibers (CNFs) are also used but they are different from each other in many aspects, such as in size and their compatibility with the composites [14, 96].

Avanish Pratap Singh et al. [97] used a novel kind of combination of the composite for EMI shielding application. They prepared phenolic resin-based composite of RGO / γ-Fe_2O_3 / carbon fibre by compression moulding technique. The incorporation of the γ-Fe_2O_3 nanoparticles inside the RGO layers improved interfacial polarization and also enhance the anisotropy energy of the composite sheets. The purpose of the fraction of γ-Fe_2O_3 improved the microwave absorbing properties of the composite, hence maximised scattering that leads to more attenuation of EM by absorption. In this case, the EMI SE_A was recorded to be 35.42 dB and SE_T at 45.26 dB. This shows the maximum contribution is due to the absorption of the EM signal as compared to the reflection which is achieved by the magnetic filler addition inside the conducting capsule that would be significant for proper impedance matching.

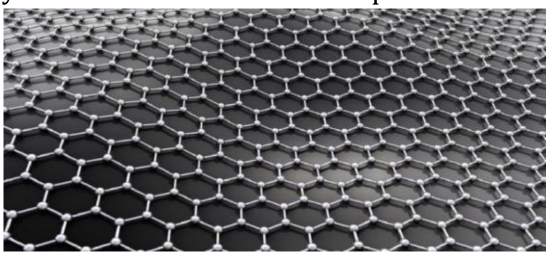
CNFs has an ability to influence the properties of the polymer nanocomposites so a higher loading of CNF inside the polymer could lead to higher EMI SE as compared to the CNTs. But the higher amount of CNF causes the poor mechanical properties and filler matrix bonding. Although CNF are cost effective as compared to CNTs, the demand of cost effective and good mechanical properties motivated Y. Yang et al. [98] to use a combination of CNFs and a small amount of CNT with the polymer matrix for EMI shielding. They used varying wt. % 1-3 for CNT and for CNFs 5-10 and investigated for their SE and obtained 20.3 dB which is the required amount for commercial applications. Juan Chen et al. [99] worked with CF for investigating its EMI SE properties. Their method of depositing GO on CF by electrophoretic process, and further GO / CF were reduced to rGO / CF through chemical reduction. The SEM results revealed the better dispersion of GO / CF and rGO / CF on the unsaturated polyester (UP) matrix. They concluded that GO / CF and rGO / CF composite showed an improved SE in UP matrix when compared to CF / UP. They observed an EMI SE of the composites having 0.75 % mass fraction of GO / CF and rGO / CF reached 34.7 and 37.8 dB at 12.4 GHz. These results showed an improvement in the SE around 6.8 – 16.3 % when compared with CF / UP composite. Table 9 summarised the results.

Table 9. CF / CN based polymer nanocomposite

Matrix	filler	wt.% or mass	thickness(mm)	EMI SE(dB)	Frequency (GHz)	Ref
PR	RGO/ γ-Fe_2O_3/CF	40/10 wt.%	-	45.26	8-12	[97]
PS	CNT/CNF	3/10 wt.%	1	20.3	12.4-18	[98]
UP	GO/CF rGO/CF	0.75 m. %	-	34.7 37.8	8-12	[99]

1.6.1.4 Graphene-based polymer nanocomposite

Graphene is an important and very fascinating 2D allotrope of carbon that constitute sp^2 bonded single layer with carbon atoms that firmly formed the planar nanostructure in which the carbon atoms made hexagonal crystal lattice. The schematic representation of graphene is shown in Fig.



3.

Figure 3. Schematic structure of graphene sheets

The men who came up with the use of single layer graphene (SLG) in 2003 were awarded with Noble Prize in 2010. Geim and Novoselov discovered graphene by using the scotch tape method [11, 100, 101]. Graphene attributes to very attractive properties, among all the carbonbased NPs. These impressive properties including high mechanical strength (Y ~ 1TPa), excellent σ (6000 S/cm), large specific area of the order of magnitude 2630 m^2g^{-1} and spectacular thermal conductivity of 5000 W/mK. Owing to excellent aforementioned properties, it is used in different areas like electronics, sensors, optoelectronics, EMI shielding and EDS [11, 47, 102]. Although graphene has impressive properties, its pure form lacks the property of uniform dispersion in polymer and it is immiscible with organic polymers [103]. It can be used conveniently with minimum filler loading with thermoplastic polymer to make it chemically and mechanically remarkable. Graphene-based polymer nanocomposite owing to the light weight, high aspect ratio and economical suitable for EMI shielding applications [104].

Liying Zhang et al. [1] fabricated 3D graphene foam by self-polymerization of Dopamine along with surface alteration of graphene. The surface modified 3D graphene showed 15 % progress on EMI-SE over the X-band frequency region, i.e., from (23.1 to 26.5) dB as compared to its counter-part (PDA / free Graphene). The presence of nitrogen in PDA act as a source to enhance the polarization effect along with the surface modification of graphene reduction. The PDA presence also made it responsible for altering the behaviour of incoming radiation upon graphene-air interface. More precisely, the EMI shielding effectiveness of PDA / rGO was grander as compared to the other graphene-polymer and carbon-based materials when considering the thickness. Sourav Biswas et al. [18] worked with the combination of PC / PVDF / $\alpha - MnO_2$ doped with crossed linked GO. The choice of the combination had direct influence on the need of requirements for EMI shielding. The doping of $\alpha - MnO_2$ of the ferrite NPs with MWCNTs ensured the intrinsic impedance matching alongside providing the conducting path ways. Ferrite doped cross linked GO helped in attenuating the EM waves. This uniquely prepared composite showed an EMI SE_T of -37 dB at 18 GHz by the absorption dominated mechanism. They also stack individual composite in multilayer manner following the absorption / multiple reflection / absorption path and evaluated the excellent SE_T at -57 dB with 0.9 mm thickness. Aforesaid, SE is very high value which indicate the attenuation of the EM wave more than 99.999 %.

Graphene sheets being highly liquid crystalline and anisotropic in nature make them an impressive candidate for multiple applications. So, lot of efforts made by researchers for preparing aligned graphene sheets in solvent and polymers. Such an effort made by Nariman Yousefi et al. [42] where they prepared self-aligned in-situ rGO / Epoxy polymer nanocomposite using an all aqueous casting method. They obtained a remarkable low percolation threshold of 0.12 vol. % with uniform dispersion of the monolayer graphene sheets having very high aspect ratio more than 30000. The self-aligned graphene sheets into the layer structure above the critical filler concentration create unique anisotropy in electrical and mechanical properties by forming the conductive paths along the aligned direction. The EMI SE of the composite having the 2 wt. % of rGO / EP about 38 dB. Yan-Jun Wan et al. [105] came with a strategy of doping the large sized graphene sheets for the preparation of the flexible light weight graphene paper (LG) for EMI shielding application. Their comparative study on iodine doped LG and un-doped LG concluded that the doped LG showed improved electrical conductivity and strength of the paper. The process of doping improved the charge carrier transport without declining the mechanical property, consequently in improving the SE of the graphene paper by the absorption dominated mechanism. They obtained the EMI SE of the doped LG of 52.2 dB at 8.2 GHz with 12.5 µm thickness, which is a large value as compared to the un-doped LG of the same thickness having SE at 47.0 dB. H.B Zhang et al. [106] prepared PMMA / graphene nanocomposite for EMI shielding application by the blending of the polymer with graphene and then foaming it through environmentally benign CO_2 technique. The microcellular graphene / PMMA foams were electrically conductive and showed an EMI SE of 13-19 dB at X-band. The dominated process of attenuation was absorption over the X-band. More on the presence of microcellular cells improved the ductility and tensile toughness of the brittle graphene/PMMA nanocomposites. Table 10 shows the findings on graphene-based polymer nanocomposite.

Table 10. Graphene-based polymer nanocomposite

Matrix	filler	wt.% or vol. %	Thickness (mm)/ μm	EMI SE(dB)	Frequency (GHz)/ MHz	Ref
PDA	rGO foam	-	0.032 mm	26.5	(8.2-12.4) GHz	[1]
PC/PVDF	$\alpha - MnO_2$/rGO/Fe	3 wt.%	0.9 mm	-36	(8-18) GHz	[18]
EP	rGO	2 wt.%	-	38	(0-5000)MHz	[42]
	Iodine doped/LG sheets Undoped/LG sheets		12.5μm	52.2 47.0	8.2 GHz	[105]
PMMA	Graphene	1.8 vol. %	2.4 mm	13-19	(8.2-12.4) GHz	[106]

1.6.3 Dielectric or magnetic additives-based polymer nanocomposites

In many applications such as radar and stealth technology, there are need of such materials that reflect very minimum radiations. To reduce the reflection loss and improving the absorption, it is a requirement of the filler to have both electric and magnetic dipoles for the interaction of the EM radiation to attenuate it [27, 65, 72]. To resolve this challenge, a lot of research were done for synthesis of the desire material, so scientific society made an attempt to add dielectric $BaTiO_3$, TiO_2 and magnetic fillers such as γ-Fe_2O_3, Fe_3O_4, $BaFe_{12}O_{19}$ within different polymer matrix [35, 107-110].

Parveen Saini et al. [107] presents a notable work in literature regarding to stealth technology as well in radar absorbing materials. They prepared $BaTiO_3$ / PANI nanocomposite through in-situ emulsion method. Their XRD and HRTEM results demonstrated the incorporation of the $BaTiO_3$ in the PANI matrix. Moreover, they investigated the nanocomposite over the K_u band (12.4- 18) GHz and observed an outstanding SE of -71 dB which is highest achieved value in the literature data. The nanocomposite showed an attenuation greater than 99.99999 % which is totally dependent on the fraction of $BaTiO_3$ NPs that provide dielectric and electrical attributes to the nanocomposite. S.W Phang et al. [108] synthesized PANI / HA / TiO_2 / CNT nanomaterial with different strategy by adding the content filler such TiO_2, SWCNT and MWCNT. The varying % of the filler directly influenced the properties like magnetization, dielectric and conductivity of polymer matrix. They investigated the nanomaterial for their EMI SE properties and concluded an attenuation of 99.2 % by absorption with wide bandwidth via the PANI / HA / TiO_2 / SWCNT with 20 % content of SWCNT. They further demonstrated that if the amount of SWCNT increased to 60%, then there would be fall of bandwidth with reflection loss less than -20 dB.

Y.E Moon et al. [35] prepared PANI coated with GO, γ-Fe_2O_3 and $BaTiO_3$ nanocomposite for improving their electromagnetic properties. The inclusion of NPs inside the PANI made a balance between the electric and magnetic dipoles which leads to the improvement of the EMI SE and thermal transport of the nanocomposite. They observed an EMI SE of the individual nanocomposite and found that PANI / GO / $BaTiO_3$ / γ-Fe_2O_3 exhibit the highest SE of~ 33 dB over 800-3000 MHz frequency spectrum. Y. Liu et al. [109] worked with the absorbing effectiveness of the different synthesized nanocomposite such as MWCNT / Epoxy, nano-Fe_3O_4 / MWNCT / Epoxy and Fe / MWCNT / Fe. The absorbing properties of the nanocomposite is dependent on the mass fraction of the MWCNT which showed a great dependence on the frequency but the incorporation of the magnetic filler is not responding the same way as MWCT. The nanocomposite MWCNT / Epoxy with 10 % of MWCNT showed an EMI SE of 56.92 dB with a bandwidth of 34.266 GHz. They also arranged the individual nanocomposite into layered architecture and observed the excellent absorbing properties shown by all. They investigate the layered structure over the testing frequency of 3.22 – 40 GHz with more than 90 % of band width and absorption of 20 dB.

Y. Wang et al. [110] prepared a nanocomposite of hard and soft ferrite $BaFe_{12}O_{19}$ / $Ni_{0.8}Zn_{0.2}Fe_2O_4$ with polymer matrix of PANI / PPY. They prepared the nanocomposite through in-situ polymerization of the individual components. They observed their electrical and magnetic properties and came to the conclusion that the insertion of the ferrite into the polymer matrix drops its electrical property and rise its magnetic property and vice versa. They recorded the EMI SE of the nanocomposite of thickness 3.0mm about -19.7dB (-12.5) dB at 7.3GHz (10.7) GHz. The inclusion of the polymer matrix improves the absorption of the nanocomposite by dielectric loss mechanism. Table 11 summarised the findings.

Table 11. Magnetic / dielectric nanofiller with different polymer

Polymer	magnetic or dielectric filler	wt. % or mass ratio	EMI SE (dB)	Frequency (GHz)/ MHz	Ref
PANI	$BaTiO_3$	2 wt.%	-71	(12.4 – 18) GHz	[107]
PANI/HA	TiO_2 /CNT	10/20 wt.%	-21.7	6 GHz	[108]
PANI	GO/ $BaTiO_3$/ γ-Fe_2O_3	3/3/3 wt.%	33	800-3000 MHz	[35]
Epoxy	Fe_3O_4/MWCNT	10 wt.%	56.92	(3.22-40) GHz	[109]
PANI/PPY	$BaFe_{12}O_{19}$/ $Ni_{0.8}Zn_{0.2}Fe_2O_4$	5:1	-19.7	12.5 GHz	[110]

1.6 Summary and Future Perspectives

In the recent time, RAMs and EMI shielding materials are used to attenuate the MW absorption in the civil and military aircrafts and control EM radiation pollution. In this review article, we have critically studied the different EMI / MW shielding materials synthesized by different techniques for high performance shielding and absorption. Here, we studied different architectures such as metallic, composites, polymer-based nanocomposites that are incorporated with metal, metal-oxides, metallic NPs, carboneous filler like CB, CF, CNF, CNTs, graphene, graphene oxide, magnetic and dielectric fillers like spinel and hexa-ferrites and barium tin oxides as EMI shield and for MW absorption.

The EMI shielding material such as metals are not used frequently due to its heavy weight, corrosion reception and poor meting out conditions for improved EMI SE. Also, the materials prepared with pure carbon doses do not meet up the commercialization standard due to lack of flexibility. So, the scientific society turns their attention towards the different polymers and elastomers such as PVDF, PC, PU, POT, PANI, PPY, EP, PMMA, PIGF and epoxy resin, NR, to take advantage of their attractive properties like light in weight, easy processing and better flexibility. Lots of literature data has been reported in this regard such as for improving the electrical conductivity of the ICP's via the insertion of the metal, carbon-based fillers, ferrites 3D and 2D architectures. Still, there are certain challenges which are requisite to be addressed such as high percolation threshold as well as the lower aspect ratio of the polymer matrices. Thus, more researches need to be done to improve the efficiency and mechanical properties of the CPC's. Existing literature data revealed that the carbon filled with polymer-based nanocomposite exhibit better shielding efficiency though their fabrication is not an easy task due to poor dispersion and inability to be added at high concentration. These issues are need to be resolved urgently. More on the dielectric / magnetic filled polymer nanocomposite possess the properties that are dependent on the consolidated loss parameters related to the filler particles, showing narrow band action and other factors like particles aggregate, particles interaction with polymer required much attention to be resolved to improve the EMI shielding / MW absorption properties. The best solution is to incorporate polymer with hybrid conductive filler. The next most important challenge is to enhance the bandwidth of the EMI shielding. There are a lot of literature data that are presented yet having narrow working bandwidth. This issue may be resolved with the use of multiple elements incorporate with polymer to improve the bandwidth by working at different broad frequency ranges. Also, the mechanism of EMI shielding with incorporation of multiple elements need to solve why different elements respond to frequency differently.

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