Review

Preparation methods for Graphene, Metal and Polymer-based Composites for EMI Shielding Materials: State of the Art of Conventional and Machine Learning Methods

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Abstract: Advancement of the novel electromagnetic inference (EMI) materials is of utmost importance and essential in various industries. The purpose of this study is to present a state-of-the-art review on the methods used in the formation of graphene, metal and polymer-based composite EMI materials. The study indicates that in graphene and metal-based composites, the utilization of alternating deposition method provides the highest shielding effectiveness. However, in polymer-based composite, the utilization of chemical vapor deposition method showed the highest shielding effectiveness. Furthermore, this review reveals that there is a gap in the literature in terms of the application of artificial intelligence and machine learning methods. The results further reveal that within the past half a decade the machine learning methods including artificial neural networks had brought significant improvement for modelling EMI materials. Research trend is identified to be in the direction of using advanced forms of machine learning for comparative analysis, research and development employing hybrid and ensemble machine learning methods to deliver higher performance.

Keywords: electromagnetic inferences; shielding, graphene; metal; polymer; traditional methods; machine learning

1. Introduction

Electromagnetic pollution is increasing in our day to day life which not only affects electronic equipment but is also harmful to human health [1]. The electromagnetic waves damage human health in various forms such as psychological disorders, affect the immune system and also causing a problem in hereditary and with time its impact is increasing which requires vital attention [2]. Electromagnetic shielding history reported back in the early 1830s when Faradays cage was discovered, which is an encircled conductive housing along with zero electric fields [5]. Therefore, there is a need for appropriate materials that acts as a shield to counter electromagnetic waves [6]. Electromagnetic shielding requires a balanced combination between electrical conductivity, dielectric permittivity, and magnetic permeability. It is also observed that the material' morphology plays an important role in electromagnetic shielding and the factors introduce are the reflection, absorption, and multiple reflection losses [7-9].

In a material, the main mechanism for electromagnetic interference attenuation are absorption, reflection and multiple reflections [10]. The reflection is a primary shielding mechanism that occurs in highly electrically conductive structures such as metals. The reflection phenomena depend on mobile charge carriers such as electron which is present

with in the material. Therefore, the shielding material is likely to be electrically conductive although it is not an essential requirement [11]. The second mechanism for electromagnetic shielding is absorption [12]. It requires the existence of electric and magnetic dipoles to interact with the electromagnetic radiations and greatly depends on the thickness. The third mechanism of electromagnetic shielding is multiple reflections which require large surface areas to interfaces within the shield [13]. The four most common methods used for the measurement of electromagnetic shielding are 1) Open field or free space method 2) Shield box method 3) Shield room method and 4) Coaxial transmission line method [14]. Over the past decade, metals were commonly used materials to overcome the electromagnetic shielding interference issue due to their good electrical conductivity and overall shielding effectiveness, however, metals have many disadvantages such as high mass density, corrosion and difficult processing [15, 16]. In order to achieve good shielding effectiveness, many other materials are introduced such as carbon, graphene and conducting polymers etc. [16]. Graphene, although is non-metal, yet exhibit properties similar to semi-conducting metals which makes it suitable for electromagnetic inferences (EMI) applications [17-19]. However, conducting polymer have a problem of poor mechanical strength and low processability. The proper distribution of carbon-based filler material within the polymer matrix can be effective to obtain good electromagnetic shielding effectiveness, where polymer-based composite materials improve the absorption and as well a reflection of the incoming radiation [20, 21].

The fabrication of materials can be accomplished by using different methods where researchers tried different methods to build a new composite, The selection of methods varies from material to material, for example for the fabrication of metals, friction stir processing and stir casting are mostly used [22]. Similarly for the other materials researcher used different methods according to the properties of materials that are suitable for the preparation of new composites [23]. The selection of an appropriate method plays a vital role in achieving the EMI shielding effectiveness by forming a homogenous sample. Various studies have been conducted on different types of available methods to obtain the maximum EMI shielding of a composite. This review aims to assess the various traditional and artificial intelligence methods to synthesize the shielding composites to deal with EMI. This paper is the perpetuation of the previous review [24] conducted on the applications of graphene, iron and polymer composites in EMI shielding, where the top materials were highlighted in each frequency range to secure good shielding effectiveness. Whereas, in this review preparation methods that help to build the EMI shielding composites have been reviewed. The method selection affects the properties of the material hence impacting the EMI shielding. The scope of this study is limited to review the research articles of Graphene and Metal-based Composites, and Graphene, Metal and Polymer-based Composites formulated through various methods (traditional and artificial intelligence). Although there are many types of carbon materials, yet not all are suitable for EMI applications. Graphene is an emerging material that shows remarkable results as a composite in EMI applications, that is why it was chosen over other types. Another reason for focusing on these materials and their combination is that graphene, metal and polymer-based composites are showing a good performance in EMI applications, therefore reviewing their methods is more pertinent. This study sets a benchmark for future researchers to select the most appropriate method in a selected composite family to formulate a new shielding material.

2. Review Methodology

The methodology of this review exhibits the extraction of those articles which were published on the composite formation via traditional methods of various materials as electromagnetic shielding materials. Popular materials such as carbon, graphene, iron and polymer were taken into consideration, as it is important to know about their manufacturing behaviour which impacts significantly on shielding effectiveness. VOSviewer software was used to make keyword analysis of Graphene and Metal-based Composites, and Graphene, Metal and Polymer-based Composites articles. Furthermore, EMI studies

related to artificial intelligence were also reviewed. A summary of the review methodology is demonstrated in Figure 1.
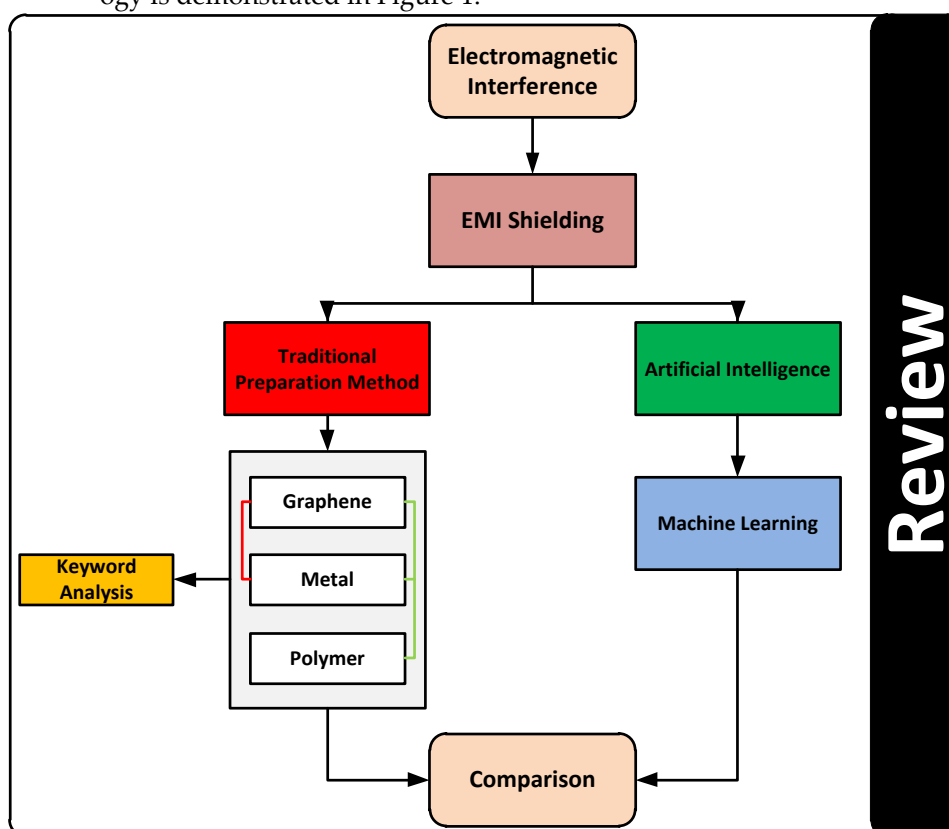

Figure 1. Review flowchart

3. Interpretation of Articles

This section covers the summary of the extracted articles (traditional methods) which were published on the preparation methods for graphene, metal and polymer-based composites for EMI shielding materials. Besides that, VOSviewer software was used to show the mapping of the extracted articles based on the keywords co-occurrence.

3.1 Summary of Extracted Articles

The English language articles were extracted using the Google Scholar search engine without applying any year limitations. The reason to use Google Scholar for articles extraction is that it covers all the published work from all the journals, either it is the Web of Science index, Scopus index or any other. Figure 2 shows the distribution of the articles of graphene and metal-based composites, and Figure 3 shows the distribution of the articles of polymer-based composites. The number of publications is limited as only those articles were taken into account which comes under the formed combination, i.e. Graphene and Metal-based Composites, and Graphene, Metal and Polymer-based Composites. It can be observed that a gradual increase in the publications occurred over time. An interesting thing which was also observed that with time the focus is more towards the polymer-based composites as it emerged as a promising shielding material.
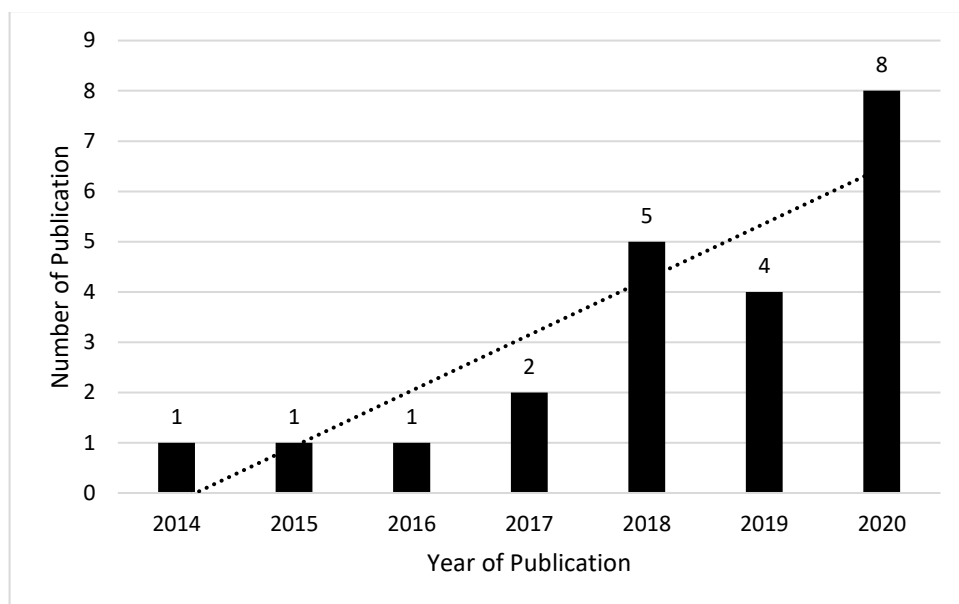
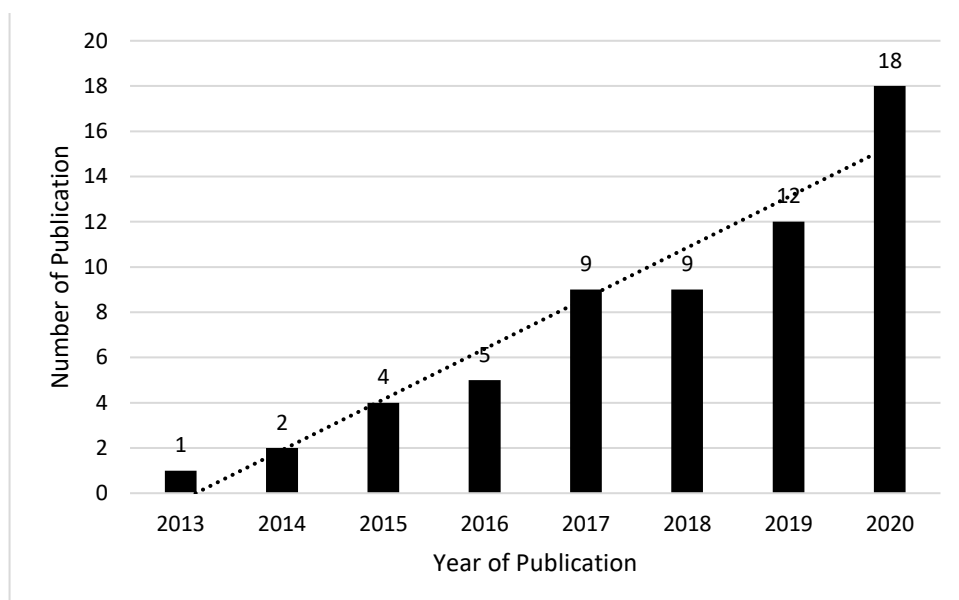**Figure 2.** Articles distribution of Graphene and Metal-based Composites.

Figure 3. Articles distribution of Polymer-based Composites.

3.2 Keywords Analysis

Keywords are an important component of a research article that provides useful information on paper as well as an area of interest. A comprehensive analysis of keywords in various technical fields can help demonstrate trends in research growth and differences. In many papers, co-occurrence analysis of keywords was often used to determine the extent of the relations between different keywords. The link and role of internal materials can be best grasped up in an academic domain by researching keyword co-occurrence relationships and revealing the research limits of the discipline. In the current analysis, a linking of the details based on keywords from the selected papers was generated with the help of VOSviewer software as seen in Figure 4.
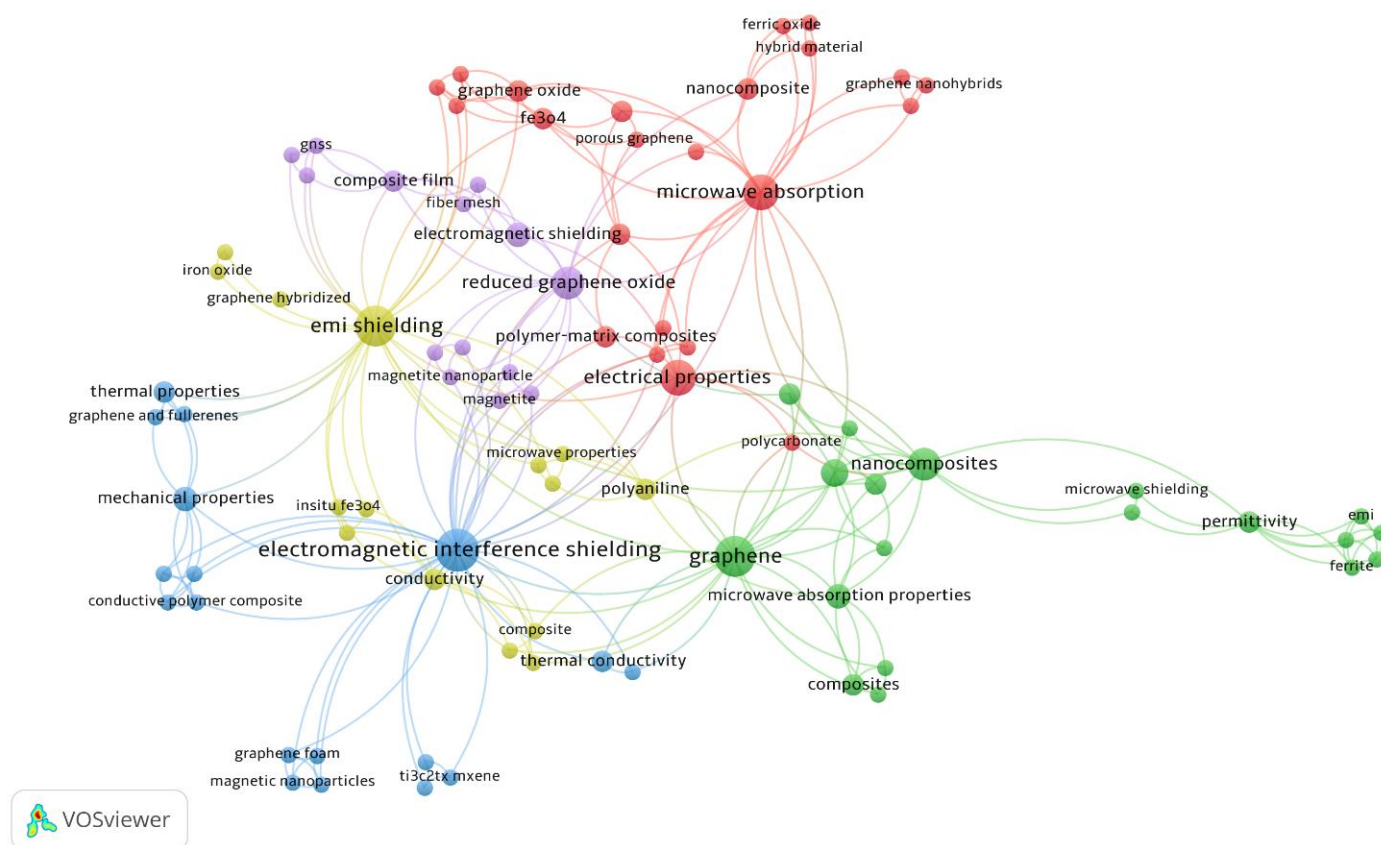
Figure 4. Mapping of Co-occurrence Keywords.

Keyword occurrence was analysed using VOSviewer's "full counting" technologies and a minimal number of keywords occurrence was set to 1. A total of 89 eligible keywords have been defined by the software that reaches the threshold. The mapping network of 89 linked recurrent keywords with five fuzzy clusters was developed by setting the cluster limit at least 13 cluster keywords. The cluster nodes represent a keyword that associates the connection with other nodes.

Blue nodes with 13 occurrences, which is the first cluster are built on the terminology "Electromagnetic interference shielding". In the same colour pattern, the terms "Mechanical properties", "Thermal properties "and "Thermal conductivity" with occurrences 3 and 2 respectively can also be seen. The following cluster also includes some other keywords and the linkage between all the keywords shows the relation of each keyword in a particular domain. Green nodes with 11 occurrences, which is the second cluster, is built on the terminology "Graphene". In the same colour pattern, few keywords like "nanocomposites", having 6 occurrences and "Microwave absorption properties" with 3 occurrences is also presented. Other keywords such as "absorption properties", "magnetic property" and "permeability" show researchers' interest in this region. Yellow nodes with 11 occurrences, which is the third cluster are built on the terminology "EMI shielding". This cluster is augmented with various polymers keywords like "Single wall carbon nanohorn, "Insitu Fe$_3$O$_4$", "Iron Oxide". The fourth prominent cluster is with red nodes around the term "Microwave absorption" and "Electrical properties" with 8 occurrences both. The fifth prominent cluster is with green nodes having the keywords "Reduced graphene oxide", with 6 occurrences.

4. Discussion of Articles

This section covers the compilation of the various methods used for the formulation of carbon, metals, graphene, iron, and polymer family materials. Based on the methods, a discussion has been provided which identify the most suitable methods in each family. The overall summary of the available traditional methods is provided in Table 1.

Table 1. Various mixing methods.

S. No	Method	Reference	Remarks	Advantage	Disadvantage
1	Chemical vapor deposition	[25-27]	It is a deposition process performed at high temperature and gas pressure and provides better optical and electrical properties in graphene-based composites.	• Recommended for coating • Gives high dispersion	• Not suitable for organic materials
2	Alternating deposition	[28]		Need further exploration.	
3	Electrophoretic deposition	[29]	Most used process for material coating	• Easy to use • The deposition rate is high • Binder elimination • Can adopt any shape	• Limited adhesion
4	In-situ growth	[30-41]	This technique is a novel way to implant graphene layers on metal without any damage to graphene. However, structural control by this technique needs further investigation.	• Wrinkle-free • High-quality dispersion • Lithography-free	• Expensive procedure • Time consuming
5	Thermal annealing method	[42, 43]	This thermal annealing method used to modify the surface morphology of materials with temperature and time. It is a mostly useable method for intrinsic, structure improving and surface roughness control in materials and is well used for stress liberation.	• Improve structure • Eliminate surface roughness	• Time consuming
6	Facile synthetic route	[44-47]	Mostly a commonly used method to synthesize porous structures.	• Cheap process • Environment friendly	• Nanoparticles formation is slow • No access to reaction process
7	Hydrothermal method	[41, 48-56]	It involves substance crystallization at high temperature and pressure.	• Suitable for the materials with a high vapour pressure • Form crystalline phases	• Expensive autoclave required
8	Scalable method	[57]		Need further exploration.	

9	Solvothermal method	[58-63]	This technique is used to form a chemical composite. The benefit of using this technique is that it involves the usage of sol-gel and hydrothermal routes, providing precise control over the shape, size and crystallinity of composites.	• Suitable for all types of materials • Good control over the size and distribution of the material	• No access to reaction process • Expensive autoclave required
10	Filtration-assisted self-assembly method	[64]		Need further exploration.	
11	Wet stirring process	[65]	It is a simple technique of stirring which deals with homogenous mixing of liquids and stir up the solid particle into liquid by using water as a solvent.	• Easy to use • Cost efficient	• A high amount of diluent is required
12	Self-assembly technique	[54, 66]	In this method without using any external direction among components the disordered system and pre-existing components make it to an organized structure or pattern, it is a low-cost approach for nanofabrication.	• Cheap process • Organized process	• Time consuming • High cost
13	Vacuum-assisted filtration method	[67]		Need further exploration.	
14	Solution processing method	[68-77]	It is a promising method to produce low-cost composites. This method is used mostly in organic materials such as polymer-based composites. It is used in different ways through high-speed shear mixing, ultrasonication and as well stirring for the formation of polymer nanocomposites where the mixing depends on the solvent. A good dispersion of carbon nanofiller in polymer	• Less expensive	• Difficult coating process

			matrixes can also be achieved.		
15	Chemical oxidative polymerization	[78-81]	In this method, oxidizing agents are used to forming a polymer-based composite.	• Suitable for polymer synthesis	• Slow polymerization
16	Co-precipitation method	[82, 83]	It is used to synthesized iron nanoparticles.	• Simple process • Low cost	• Poor control on the particle size distribution
17	Centrifugal mixing method	[84]		Need further exploration.	
18	Citrate precursor method	[85]	It is a chelate-based method that is efficient to reduce the metal ions for nanoparticles fabrication and also stimulate reaction conditions.	• Low cost • Less cracking in composite due to internal heat distribution • Good atomic-level mixing	• Difficult to control the parameters
19	Chemical reduction	[16]	This method is a cost-effective method and widely used in for mass production of reduced graphene oxide in which reduction agent used in the form of gas or liquid in graphene oxide for the elimination of functional group.	• Mass production • Cost effective	• Not suitable for all composites.
20	Hot-moulding process	[86]	This process is useful for adding thermoplastic binders to the metallic and ceramic powder to make it fluent. In all this process temperature used above then the melting point.	• Effective for the lesser amount of materials • Cheap process	• Contamination risk • Slow process
21	Mechanical mixing	[87]	It is used to form a uniform coat of the particles on the material surface where pellets are followed by a cooling process.	• Cheap and easy process	• High share force not suitable for graphene composite
22	Dilute polymerization	[88]		Need further exploration.	
23	High-pressure solid-phase compression moulding	[88]	It is an old material processing technique. In industrial methods which are used for plastic, it was a commonly used method.	• Effective for thermoplastic composite • Cheap process	• Not recommended on high-scale mass production
24	Injection moulding process	[89]	It is a high volume and low-pressure process	• High production rate • Easy to mould	• Costly for the lesser amount of materials

S. No	Method		(description)	(advantages)	(disadvantages)
			which is performed with filled thermoplastics.	• Suitable for most of the materials	
25	Ultrasonication technique	[90-92]	It is a technique used for the preparation of nanoparticles. It has good control over the structure of the material. Moreover, with this technique the size of a previously formed composite can also be reduced.	• Controlled structure of materials	• High energy consumption
26	Hummers method	[93-97]	It is a chemical process mostly used to produce graphene from graphite.	• Cheap process • Time saving • Highly efficient	• Release toxic gases during experimentation
27	Hot compressed method	[15, 98-101]	The hot compression method cannot work at room temperature like the cold compression method as it takes place by applying heat to the mould.	• Preferable for smaller production • Cheap process	• Slow process • Damaging to moulds • Contamination risk
28	3D printing method	[102]	A new method to form a shielding material followed by Object's PolyJet Matric printing technology, where a couple of materials are built simultaneously.	• Time efficient • Less parameters involved	• No mass production High cost

4.1. Traditional Methods

4.1.1 Methods for preparation of Graphene and Metal-based Composites

     Over the years, graphene has grabbed attention in the field of research due to its tremendous properties. Graphene is wrapped in the honeycomb crystal lattice and is a one atom thick planar sheet [103]. Graphene possesses optical transparency, excellent electrical conductivity, thermal conductivity, mechanical flexibility and low coefficient of thermal expansion behaviour, making it suitable for use in various fields [104]. Similarly, metals can transmit, reflect and absorb EMI and are good electrical conductors as well. Plastics and rubbers are transparent to EMI and are nonconductive as well. Whereas metals have the ability to transmit heat and electricity which makes them good for many applications [105]. Various methods have been used to synthesize the shielding composites with the combination of graphene and metals or both with some other materials as well. A brief description of such methods and the formed composites with shielding effectiveness (SE) has been presented in Table 2. The negative value in the shielding effectiveness (SE) column shows the reflection loss, whereas the positive value is the absorption/total shielding effectiveness.

Table 2. Preparation Methods of Graphene, Metal and Polymer-based Composites.

S. No	Method	Material Composite	SE (dB)	Frequency	Reference
1	Chemical vapor deposition	3DG/Cu	32.3	Ku-band	[27]
		3D Graphene Network@PDMS	90	X-band	[25]

		MXene($Ti_3C_2T_X$)/graphene/PDMS	80	X-band	[26]
			77	Ka-band	
2	Alternating deposition	Cu/Gr	60.95	THF-band	[28]
3	Electrophoretic deposition	Cu–Ni–GNS	42	X-band	[29]
		CuNW@G	52.5	Ku-band	[39]
		GNP@PANI	-14.5	X-band	[30]
		Graphene@NiO@PANI@Ag	-37.5	Ku-band	[31]
		TiO_2/PANI/GO	-51.7	Ku-band	[34]
		Ag@Graphene/PANI	29.33	L-band	[35]
		PANI/$Li_{0.5}Fe_{0.5-x}Gd_xO_4$	42	X-band	[36]
4	In-situ growth	RGO@Hematite/PVDF	-43.97	Ku-band	[37]
		γ-Fe_2O_3/RGO/PANI	51	X-band	[33]
		PEDOT/RGO/$SrFe_{12}O_{19}$	62	X-band	[38]
		FeCo@RGO@PPy	-40.7	Ku-band	[31]
		Graphene/Ni	20	X-band	[40]
		PG-Fe_3O_4	-53	C-band	[41]
		G-PANI	32.5	Ku-band	[32]
5	Thermal annealing method	Graphene/Ni hybrid mesh	26.6	UHF-band	[43]
		CuNWs-TAGA/Epoxy	47	X-band	[42]
		RGO/Ni hybrid	52	Ku-band	[47]
6	Facile synthetic route	Polycarbonate/GNP	47	X-band	[44]
		Fe_3O_4/PANI rod/RGO	-33.3	Ku-band	[45]
		GNSs-Fe_3O_4/PVDF	52	X-band	[46]
		$ZnFe_2O_4$@graphene@TiO_2	-55	S-band	[41]
		MoS_2-RGO/$CoFe_2O_4$	19.26	X-band	[53]
		Fe_3O_4@C@Graphene	-55.02	Ku-band	[54]
		G-F	20	Ku-band	[55]
7	Hydrothermal method	$Ni_{0.5}Co_{0.5}Fe_2O_4$/graphene	-30.92	L-band	[56]
		Graphene@PANI@TiO_2	-45.4	Ku-band	[48]
		GA/PDMS	60	Ku-band	[49]
		RGO@CuS@PVDF	-25	Ku-band	[50]
		G/Polyurethane sponge	35	X-band	[51]
		PEDOT:PSS-Fe_3O_4-RGO	-61.4	Ku-band	[52]
8	Scalable method	Cellulose/reduced graphene oxide (RGO)/Fe_3O_4 aerogels	52.4	X-band	[57]
		$NiFe_2O_4$/RGO	38.2	X-band	[60]
9	Solvothermal method	Fe_3O_4@f-GNPs	25	X-band	[61]
		Ag@Fe_3O_4@RGO	-40.05	Ku-band	[62]

		Fe_3O_4@C/RGO	-59.23	Ku-band	[63]
		Hollow Fe_3O_4@GF@PDMS	70.3	X-band	[58]
		RGO-PEDOT-$NiFe_2O_4$	-45.4		
		RGO-PANI-$NiFe_2O_4$	-49.7	Ku-band	[59]
		RGO-PPy-$NiFe_2O_4$	-44.8		
10	Filtration-assisted self-assembly method	Fe_3O_4-C, C-MIL-88B/GNP	28	X-band	[64]
11	Wet stirring process	GO@CIP	-56.4	Ku-band	[65]
12	Self-assembly method	NRMG	26.4	X-band	
		PMMA/RGO	63.2	X-band	[66]
13	Vacuum-assisted filtration method	RGO/CNF@Ag-Fe_3O_4	21	X-band	[67]
14	Solution processing method	PVC/PANI/GNP	51	K-band	[74]
		Gn/SiCnw/PVDF	32.5	X-band	[68]
		PVDF/graphene	47	X-band	[69]
		Fe_3O_4@RGO/TPU	~15.51±1.6	X-band	[70]
		BaFe@TRGO@TPU	-61	K-band	[71]
		Fe_3O_4@SLGAPC@PVA	20	X-band	[72]
		PVDF/GNP-Ni-CNT	46.4	Ku-band	[73]
		PVDF/PFC	-29.7	Ku-band	[75]
		TPU/TRG	32	Ku-band	[76]
		RGO/PF/Epoxy	-10.26	Ku-band	[77]
15	Chemical oxidative polymerization	Graphene@Fe_3O_4@PANI@WO_3	-46.7	X-band	[81]
		PEDOT/RGO/$PbTiO_3$	51.94	Ku-band	[79]
		Fe_3O_4/C:PPy	>28	C-band	[80]
		Polypyrrole/BST/RGO/Fe_3O_4	48	X-band	[78]
16	Co-precipitation method	$Ti_3C_2T_x$/Fe_3O_4@PANI	58.8	X-band	[82]
		GNP/Fe_3O_4/Epoxy	37.03	X-band	[83]
17	Centrifugal mixing method	TGO/CI/Epoxy	40	X-band	[84]
18	Citrate precursor method	PANI/BF/RGO	31.1	X-band	[85]
19	Chemical reduction	RGO-CF/EP	37.6	X-band	[16]
20	Hot-moulding process	PVDF/n-Fe	40.21	Ku-band	[86]
21	Mechanical mixing	Graphene flakes@PDMS	31	THF-band	[87]
22	Dilute polymerization	Graphene@Fe_3O_4@SiO_2@polyaniline	-40.7	X-band	[88]
23	High-pressure solid-phase compression moulding	RGO@polystyrene	45.1	X-band	[88]

24	Injection moulding process	Polyethylene@GNP	31	K-band	[89]
		GNP/EPDM	35	Ku-band	[90]
25	Ultrasonication technique	Ni@GNS@PVDF	51.4	K-band	[91]
		GNP/Fe/Epoxy	-78	V-band	[92]
		PANI/GO/Fe_3O_4	-53.5	Ku-band	[93]
		Cu@RGOFM@PDMS	74.2	X-band	[94]
26	Hummers method	GCF/MG_3/EP	51.1	K-band	[95]
		RGO@PEI	26	X-band	[96]
		PVA/Gr/Fe_3O_4	40.7	X-band	[97]
		PMMA/graphene	21	X-band	[99]
		PVC/graphene	31	X-band	
		Polylactic acid/Biochar/Graphite	30	K-band	[15]
27	Hot compressed method	$NiFe_2O_4$-RGO-Polypropylene	29.4	C-band	[100]
		GNP/CLF/PEEK	27.1	X-band	[101]
		Conventional RGO/PS	14.2	K-band	[98]
		Segregated RGO/PS	29.7	K-band	
28	3D printing method	Graphene/$Li_{0.35}Zn_{0.3}Fe_{0.35}O_4$/PMMA	-46.1	Ku-band	[102]

Note: Ultra high frequency (UHF) = 300 MHz - 1 GHz, L-band = 1 to 2 GHz, S-band = 2 GHz - 4 GHz, C-band = 4 GHz - 8 GHz, X-band = 8 GHz - 12 GHz, Ku-band = 12 GHz - 18 GHz, K-band = 18 GHz - 27 GHz, Ka-band = 27 GHz - 40 GHz, V-band = 40 GHz - 75 GHz, Tremendously high frequency (THF) = 300 - 3000 GHz

It can be observed that for the preparation of graphene and metal-based composites various methods have been utilized. Interestingly, graphene and metals family materials were constructed with different methods, illustrating that the structure of the material significantly depends on the selected method. Hydrothermal and solvothermal are the two most common methods that have been used extensively for these composites. The composites formed by these methods were tested up to the Ku-band frequency range, where the highest reflection loss of -55.02 dB was achieved using hydrothermal and reflection loss of -59.23 dB was achieved using the solvothermal method. The highest shielding effectiveness of 52.4 dB in X-band was achieved via using the scalable method. The higher shielding of 60.95 dB was achieved in THF-band by alternating deposition where graphene and copper were synthesized. In this case, the role of materials properties also gives significant input. Figure 5 shows the maximum shielding effectiveness achieved by utilizing the various methods in different frequency ranges.
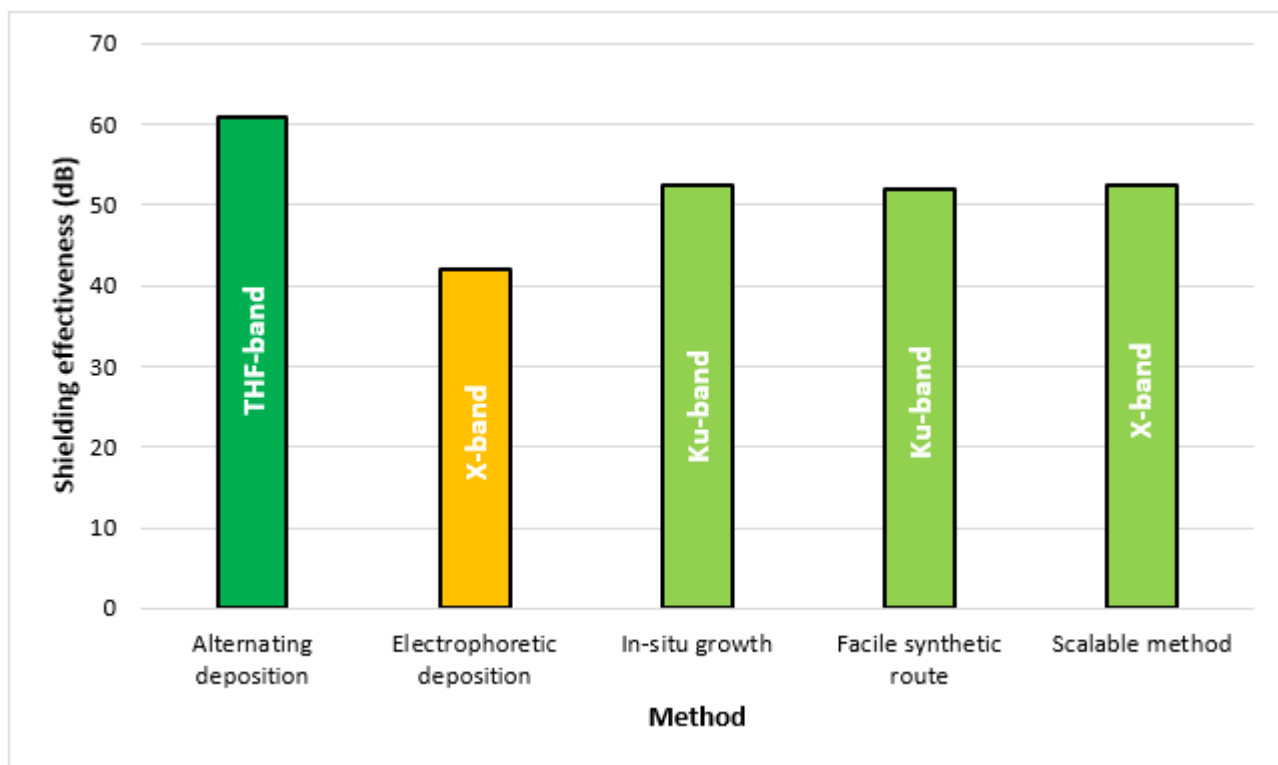
Figure 5. Top 5 methods providing higher SE in Graphene and Metal-based Composites.

It can be observed that in-situ growth, facile synthetic route, and scalable method gives SE greater than 50 dB in X-band and Ku-band frequency range, whereas, with electrophoretic deposition, the SE was in the range of 40 dB in X-band. Looking into the combinations of the materials, a scalable method provides better shielding in X-band, while both in-situ growth and facile synthetic route come as the most suitable methods for Ku-band frequency range materials. The highest shielding effectiveness greater than 60 dB was achieved by alternating deposition in THF-band, however, this method requires further exploration.

4.1.2 Methods for preparation of Polymer-based Composites

Graphene and metals although are the most suitable composites for EMI shielding but have some limitations [106, 107]. Due to the advancement in electronic applications, the demand for an effective shielding material has also boost up where thermal expansion, material design flexibility, and non-corrosive properties play a significant role. Besides these properties, the weight-to-strength ratio of EMI shielding materials is also important from the inertia and structural perspective. Moreover, to be part of the electronic system, the material should be lightweight where the polymer composite materials emerge as the most promising materials [108]. Forming a polymer composite, various methods have been used as shown in Table 2.

As shown in Table 2, to form a polymer-based composite, various methods have been used where the most adopted methods are solution processing method, in-situ growth, hydrothermal method, hummers method and solvothermal method. In the X-band frequency range, the highest total shielding effectiveness was achieved up to 90 dB by utilizing chemical vapor deposition. In Ku-band, the highest total shielding effectiveness was achieved up to 60 dB with a reflection loss of -61.4 dB by making a composite with the hydrothermal method. In the K-band frequency range the maximum total shielding effectiveness of 51.4 dB, 51.1 dB and 51 dB by using ultrasonication technique, hummers method and solution processing method. While a reflection loss of -61 dB was achieved via the solution processing method. In the Ka-band frequency range, the highest total shielding effectiveness of 77 dB was achieved by using chemical vapor deposition.

Overall, this method gave better shielding in both X-band and Ka-band. A high reflection loss of -78 dB was also observed in the V-band frequency range by the ultrasonication method. A comparison of all the methods has been drawn in Figure 6 which gives shielding effectiveness greater than 50 dB in their respectable frequency ranges.
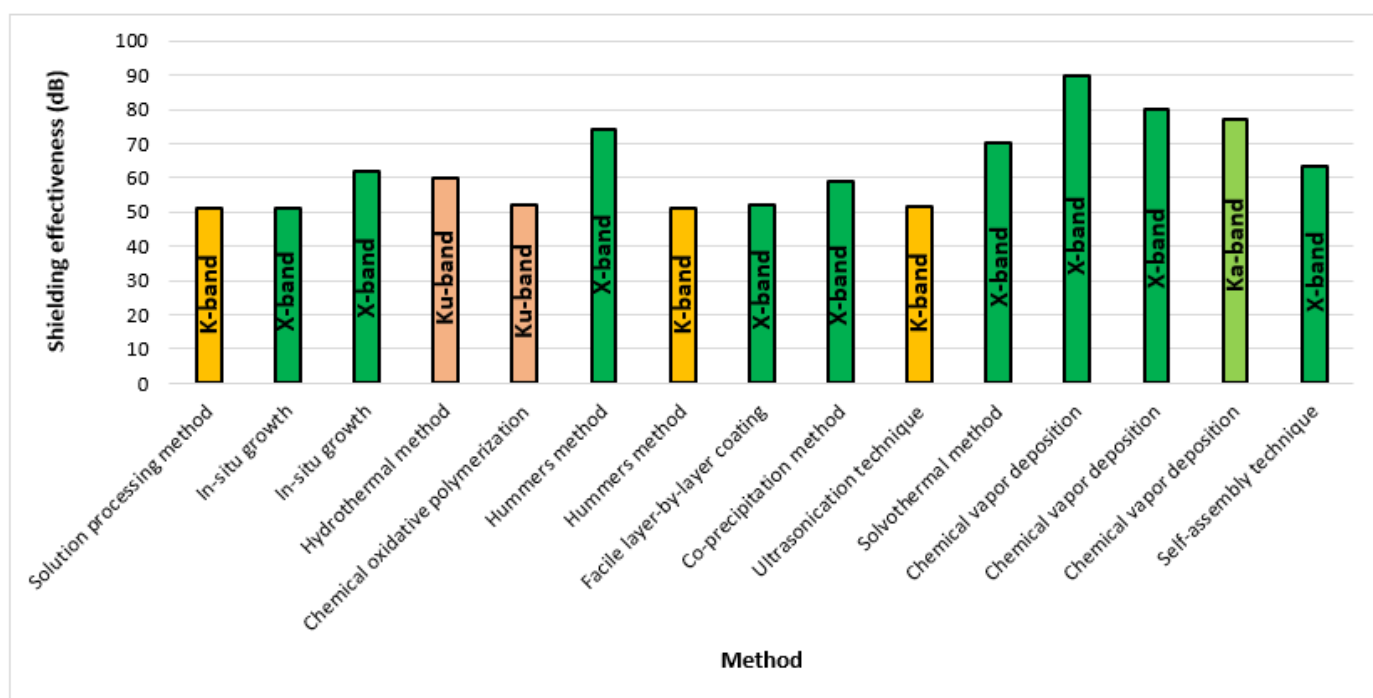

Figure 6. Top methods providing higher SE in Polymer-based Composites.

It can be observed that in the X-band frequency range the highest shielding effectiveness was attained by forming the polymer composite via chemical vapor deposition. In the Ku-band frequency range, the hydrothermal method was more efficient as compared to chemical oxidative polymerization. In the K-band frequency range, solution processing method, hummers method and ultrasonication technique, all performed efficiently. While in Ka-band chemical vapor deposition gives better shielding effectiveness. The overall maximum shielding was achieved by chemical vapor deposition; however, more combinations need to be tested formed by this method.

In this review, the authors tried to draw a comparison of the composite's formation methods for better understanding for future researchers, while performing the review it was observed that this area still needs exploration in terms of comparison of the same composite formed via different methods. In this way, the direction for each composite will be clearer for researchers.

4.2. Artificial intelligence methods

Development of artificial intelligence, mainly in machine learning, a variety of reforms have been made in materials formation by exploring new materials and their combinations, along with their properties. This approach is a trending topic as a lot of work is still ongoing [109-111]. The machine learning method has the potential to discover the properties of new composites [112]. However, its benefits are still unrevealed, especially in polymer science [113, 114]. Various properties of polymers depend on the degree of crystallinity. Machine learning-based methods are competent enough to forecast crystallinity, which can counter the deficiency of traditional methods. With the help of machine learning, melting temperature can also be predicted for new polymers, as its one of the difficult parameters to be controlled in traditional methods [115]. For specific applications, machine learning models are used to identify the properties of the polymer, such as dielectric constant [116] which is a parameter for attaining efficient EMI shielding.

The composite synthesis requires materials recognition to attain the desirable properties required for specific applications. The traditional methods have been used extensively to evaluate the required properties, based on which further assessment is performed [117]. However, there are various drawbacks of these traditional methods such as time and material consumption, selection of appropriate method for a specific material etc. [118, 119]. In this manner, machine learning is a tool that can be utilized for quick decisions [120]. Not only it helps in finding the properties, but new materials formation is also possible by its adaptation [113, 121]. Various machine learning approaches that have been used in EMI are presented in Table 4.

Table 3: Machine learning methods for EMI

S. No	Machine learning Method	Reference	Remarks	Advantage	Disadvantage
1	Association rule learning and decision tree algorithm	[122]	Effective in dealing with electromagnetic interference in high power line communication and helps to eliminate the troubleshooting	• Normalization and data scaling not required	• Time consuming
2	K-nearest neighbors (k-NN) algorithm	[123]	Continuous monitoring in air traffic control communication is applicable against electromagnetic interference	• Training period not required • Easy implementation	• Not suitable for large and high dimension data
3	Artificial Neural Networks	[124-128]	An effective approach to eliminate electromagnetic interference problems	• Fast evaluation	• Problem identification is difficult
4	Back-propagation Neural Network	[129, 130]	Use backward pass approach for parameters adjustment	• Fast and easy to use	• Actual performance depends on data input for problem solving
5	Self-Organizing Feature Map Neural Network	[131, 132]	Effective in evaluating the global features of electromagnetic inferences factors	• Interpretation of data is easy • Grid clustering is helpful to evaluate the data similarity	• Slow training
6	Neural Networks	[133-137]	Effective for electromagnetic interference generated underground metallic pipelines, high voltage power lines and other problems	• Detect complex nonlinear relationship • Suitable for multiple training	• Black box
7	Monte Carlo Method	[138, 139]	Effective in electromagnetic interference problems	• Flexible simulation	• Time consuming

solving especially in power lines	

It is evident from the above table that various machine learning approaches are available to deal with EMI problems, however, this area requires extensive work as most researchers are focusing on the traditional methods, regardless of their time and cost consuming drawbacks instead of adopting the artificial intelligence. Machine learning methods to evaluate the composite properties and formation needs vital attention as it is the future in material science and can bring major reforms by constructing new composites. With the help of machine learning, materials properties can be pre-tested, which can be helpful in the construction of the best-suited combination before its experimental formation. Another approach that can be utilized with the help of machine learning is the formation of 3D shielding materials constructed via 3D printing. Such 3D printing can be a time and material saver. The results further reveal that within the past half a decade the machine learning methods including artificial neural networks had brought significant improvement for modelling EMI materials. Research trend is identified to be in the direction of using advanced forms of machine learning for comparative analysis, research and development employing hybrid and ensemble machine learning methods to deliver higher performance. ANN, and advanced forms of neural networks and optimized ANN are the most dominant machine learning methods used.

5. Future Direction

The dependency of the traditional method is mainly on the structure of the material which has been taken to form a composite material. Moreover, many new methods have been introduced which needs further investigation to prove their needfulness purposes. Although there are many methods available for the formation of shielding material, very few effective in giving desirable results. However, the area is still unexplored in terms of methods comparison. The opinion of methods selection can be biased as most researchers present their findings as extraordinary without making the comparison. The most suitable way to observe the efficiency of a particular method for any composite is to make the same composite with different available methods and then perform the same analysis, which will give the true picture of the adopted methods. Moreover, the inclusion of machine learning in EMI applications can bring reforms. This is still a gap in this area of knowledge and the investigation will be benchmarking for new researchers as it is time and cost consuming if a wrong method ever adopted. The results further reveal that within the past half a decade the machine learning methods including artificial neural networks had brought significant improvement for modelling EMI materials. Research trend is identified to be in the direction of using advanced forms of machine learning for comparative analysis, research and development employing hybrid and ensemble machine learning methods to deliver higher performance.

6. Conclusions

This study is a review of the formation of graphene, metal, and polymer-based composites via various traditional and artificial intelligence methods. The working on graphene and metal-based composites as shielding material exists for a long time, where the addition of polymer-based composites is new and remarkable results have been seen in the field of electromagnetic shielding. An extensive literature was conducted where it was revealed that in graphene and metal-based composites, the alternating deposition method, which is still less explored provide the maximum shielding effectiveness in THF-band. In Ku-band, in-situ growth, while in X-band scalable method utilization provides better shielding effectiveness. In polymer-based composites, the highest shielding effectiveness came when the composite was formed from chemical vapor deposition in X-band. While in Ku-band hydrothermal method, in K-band solution processing method, hummers method and ultra-sonication technique and in Ka-band chemical vapor deposition utilization provided better shielding effectiveness. However, there is still a gap in the implication of machine learning in EMI applications. The review was conducted with the purpose to highlight the best-suited method for the formation of the composites; however, it is concluded that it is too early to declare any method as

the best as still there is a gap in this area of knowledge that needs to be filled by making extensive research in which a comparison of the methods should be made for a single composite. The formation of a single composite via various methods upon shielding effectiveness will provide the true picture of the scenario that which is the most suitable methods among the available list in providing the highest shielding effectiveness. The results further reveal that within the past half a decade the machine learning methods including artificial neural networks had brought significant improvement for modelling EMI materials. Research trend is identified to be in the direction of using advanced forms of machine learning for comparative analysis, research and development employing hybrid and ensemble machine learning methods to deliver higher performance.

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