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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’s 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.

![Review flowchart](image)

**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.
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”, “In situ Fe3O4”, “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.
| 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. 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. | • Wrinkle-free • High-quality dispersion • Lithography-free | • Expensive procedure • Time consuming            |
| 5     | Thermal annealing method    | [42, 43]         |                                                                        | • 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 • Suitable for the materials with a high vapour pressure • Form crystalline phases | • Nanoparticles formation is slow • No access to reaction process • Expensive autoclave required |
| 7     | Hydrothermal method         | [41, 48-56]      | It involves substance crystallization at high temperature and pressure. |                                               |                                                 |
| 8     | Scalable method             | [57]             |                                                                        | Need further exploration.                     |                                                 |
|   | Method                        | Description                                                                 | Pros                                                                 | Cons                                                                 |
|---|-----------------------------|----------------------------------------------------------------------------|----------------------------------------------------------------------|----------------------------------------------------------------------|
| 9 | Solvothermal method         | 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<br>• Good control over the size and distribution of the material | • No access to reaction process<br>• Expensive autoclave required |
| 10| Filtration-assisted self-assembly method | 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. 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. | • Easy to use<br>• Cost efficient | • A high amount of diluent is required |
| 11| Wet stirring process        |                                                                           | • Cheap process<br>• Organized process | • Time consuming<br>• High cost |
| 12| Self-assembly technique     |                                                                           | • Less expensive | •  Difficult coating process |
| 13| Vacuum-assisted filtration method |                                                                           | Need further exploration. | Need further exploration. |
| 14| Solution processing method  |                                                                           | • Less expensive | • Difficult coating process |
| Step | Method                                                                 | Advantages                                                                 | Disadvantages                                                                 |
|------|------------------------------------------------------------------------|----------------------------------------------------------------------------|--------------------------------------------------------------------------------|
| 15   | Chemical oxidative polymerization [78-81]                             | Suitable for polymer synthesis                                             | Slow polymerization                                                            |
|      |                                                                         | • Suitable for polymer synthesis                                           | • Slow polymerization                                                          |
|      |                                                                         | • Simple process                                                             | • Poor control on the particle size distribution                              |
| 16   | Co-precipitation method [82, 83]                                       | It is used to synthesized iron nanoparticles.                              |                                                                                |
|      |                                                                         | • Simple process                                                             | • Slow process                                                                |
| 17   | Centrifugal mixing method [84]                                         | Need further exploration.                                                  |                                                                                |
|      |                                                                         | • Low cost                                                                  | • Difficult to control the parameters                                          |
| 18   | Citrate precursor method [85]                                          | Need further exploration.                                                  |                                                                                |
|      |                                                                         | • Low cost                                                                  | • Difficult to control the parameters                                          |
|      |                                                                         | • Less cracking in composite due to internal heat distribution              |                                                                                |
| 19   | Chemical reduction [16]                                                | It is a chelate-based method that is efficient to reduce the metal ions for | Mass production                                                               |
|      |                                                                         | nanoparticles fabrication and also stimulate reaction conditions.          | • Cost effective                                                               |
|      |                                                                         | • Effective for mass production                                             | • Not suitable for all composites.                                            |
|      |                                                                         | • Contamination risk                                                        |                                                                                |
|      |                                                                         | • Slow process                                                              |                                                                                |
| 20   | Hot-moulding process [86]                                              | Effective for the lesser amount of materials                               | High share force not suitable for graphene composite                           |
|      |                                                                         | • Effective for the lesser amount of materials                               | • Contamination risk                                                         |
|      |                                                                         | • Cheap process                                                             | • Slow process                                                                |
| 21   | Mechanical mixing [87]                                                 | Effective for the lesser amount of materials                               | High share force not suitable for graphene composite                           |
|      |                                                                         | • Cheap and easy process                                                    | • Costly for the lesser amount of materials                                  |
|      |                                                                         | • Contamination risk                                                        |                                                                                |
| 22   | Dilute polymerization [88]                                             | Effective for thermoplastic composite                                       | Not recommended on high-scale mass production                                |
|      |                                                                         | • Effective for thermoplastic composite                                      | • Not recommended on high-scale mass production                              |
| 23   | High-pressure solid-phase compression moulding [88]                   | It is an old material processing technique. In industrial methods which are | High production rate                                                          |
|      |                                                                         | are used for plastic, it was a commonly used method.                      | • Costly for the lesser amount of materials                                  |
| 24   | Injection moulding process [89]                                        | It is a high volume and low-pressure process                                |                                                                                |
which is performed with filled thermoplastics.

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.

25 Ultrasonication technique [90-92]

• Suitable for most of the materials
• Controlled structure of materials
• High energy consumption

26 Hummers method [93-97]

It is a chemical process mostly used to produce graphene from graphite. The hot compression method cannot work at room temperature like the cold compression method as it takes place by applying heat to the mould. A new method to form a shielding material followed by Object’s PolyJet Matric printing technology, where a couple of materials are built simultaneously.

• Cheap process
• Time saving
• Highly efficient
• Release toxic gases during experimentation
• Preferable for smaller production
• Cheap process

27 Hot compressed method [15, 98-101]

• Time efficient
• Less parameters involved
• No mass production
• High cost

4.1. 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) column 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.

| 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] |
| Method Description | Sample Composition | X-band | Ku-band | Ka-band | THF-band | L-band | S-band | Preprints (www.preprints.org) | doi:10.20944/preprints202107.0299.v1 |
|--------------------|--------------------|--------|---------|---------|-----------|--------|--------|-----------------------------|--------------------------------|
| **Alternating deposition** | Cu/Gr | 60.95 | | | | | | | |
| **Electrophoretic deposition** | Cu–Ni–GNS | 42 | | | | | | | |
| | CuNW@G | 52.5 | | | | | | | |
| | GNP@PANI | -14.5 | | | | | | | |
| | Graphene@NiO@PANI@Ag | -37.5 | | | | | | | |
| | TiO$_2$/PANI/GO | -51.7 | | | | | | | |
| | Ag@Graphene/PANI | 29.33 | | | | | | | |
| | PANI/Li$_3$Fe$_{0.5-x}$Gd$_{0.5}$O$_4$ | 42 | | | | | | | |
| **In-situ growth** | RGO@Hematite/PVDF | -43.97 | | | | | | | |
| | γ-Fe$_2$O$_3$/RGO/PANI | 51 | | | | | | | |
| | PEDOT/RGO/SrFe$_2$O$_9$ | 62 | | | | | | | |
| | FeCo@RGO@PPy | -40.7 | | | | | | | |
| | Graphene/Ni | 20 | | | | | | | |
| | PG-Fe$_3$O$_4$ | -53 | | | | | | | |
| | G-PANI | 32.5 | | | | | | | |
| **Thermal annealing method** | Graphene/Ni hybrid mesh | 26.6 | | | | | | | |
| | CuNWs-TAGA/Epoxy | 47 | | | | | | | |
| | RGO/Ni hybrid | 52 | | | | | | | |
| **Facile synthetic route** | Polycarbonate/GNP | 47 | | | | | | | |
| | Fe$_3$O$_4$/PANI rod/RGO | -33.3 | | | | | | | |
| | GNSs-Fe$_3$O$_4$/PVDF | 52 | | | | | | | |
| | ZnFe$_2$O$_4$/graphene@TiO$_2$ | -55 | | | | | | | |
| | MoS$_2$-RGO/CoFe$_2$O$_4$ | 19.26 | | | | | | | |
| | Fe$_3$O$_4$@C@Graphene | -55.02 | | | | | | | |
| | G-F | 20 | | | | | | | |
| **Hydrothermal method** | Ni$_{0.5}$Co$_{0.5}$Fe$_2$O$_4$/graphene | -30.92 | | | | | | | |
| | Graphene@PANI@TiO$_2$ | -45.4 | | | | | | | |
| | GA/PDMS | 60 | | | | | | | |
| | RGO@CuS@PVDF | -25 | | | | | | | |
| | G/Polyurethane sponge | 35 | | | | | | | |
| | PEDOT:PSS-Fe$_3$O$_4$/RGO | -61.4 | | | | | | | |
| **Scalable method** | Cellulose/reduced graphene oxide (RGO)/Fe$_3$O$_4$ aerogels | 52.4 | | | | | | | |
| | NiFe$_2$O$_4$/RGO | 38.2 | | | | | | | |
| **Solvothermal method** | Fe$_3$O$_4$@f-GNPs | 25 | | | | | | | |
| | Ag@Fe$_3$O$_4$/RGO | -40.05 | | | | | | | |
|   | Method                                    | Material | ΔT (°C) | X-band          | Reference |
|---|------------------------------------------|----------|---------|-----------------|-----------|
| 10 | Filtration-assisted self-assembly method | FeO$_3$-C, C-MIL-88B/GNP | 28       | X-band          | [64]      |
| 11 | Wet stirring process                      | GO@CIP   | -56.4   | Ku-band         | [65]      |
| 12 | Self-assembly method                      | PMMA/RGO | 63.2    | X-band         | [66]      |
| 13 | Vacuum-assisted filtration method         | RGO/CNF@Ag·Fe$_3$O$_4$ | 21       | X-band         | [67]      |
| 14 | Solution processing method                | BaFe@TRGO@TPU | -61    | K-band         | [71]      |
| 15 | Chemical oxidative polymerization         | FeO$_3$@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]      |
|     |                                          | Graphene@FeO$_3$@PANI@WO$_3$ | -46.7 | X-band | [81]      |
| 16 | Co-precipitation method                   | PEDOT/RGO/PbTiO$_3$ | 51.94 | Ku-band | [79]      |
|     |                                          | FeO$_3$/CPPy  | >28     | C-band         | [80]      |
|     |                                          | Polypyrrole/BST/RGO/FeO$_3$ | 48 | X-band | [78]      |
|     |                                          | Ti$_3$C$_2$T$_x$/FeO$_3$@PANI | 58.8 | X-band | [82]      |
|     |                                          | GNP/FeO$_3$/Epoxy | 37.03 | X-band | [83]      |
| 17 | Centrifugal mixing method                 | TGO/Cl/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@FeO$_3$@SiO$_2$@polyaniline | -40.7 | X-band | [88]      |
| 23 | High-pressure solid-phase compression moulding | RGO@polystyrene | 45.1 | X-band | [88]      |
| Method                        | Composite                | Frequency | Reference |
|-------------------------------|--------------------------|-----------|-----------|
| Injection moulding process    | Polyethylene@GNP         | K-band    | [89]      |
|                               | GNP/EPDM                 | K-band    | [90]      |
| Ultrasonication technique     | Ni@GNS@PVDF              | K-band    | [91]      |
|                               | GNP/Fe/Epoxy             | V-band    | [92]      |
|                               | PANI/GO/Fe3O4            | K-band    | [93]      |
|                               | Cu@RGOFM@PDMS            | X-band    | [94]      |
| Hummers method                | GCF/MG3/EP               | K-band    | [95]      |
|                               | RGO@PEI                  | X-band    | [96]      |
|                               | PVA/Gr/Fe3O4             | X-band    | [97]      |
|                               | PMMA/graphene            | X-band    |           |
|                               | PVC/graphene             | X-band    | [99]      |
|                               | Polylactic acid/Biochar/Graphite | K-band | [15] |
| Hot compressed method         | NiFe3O4-RGO-Polypropylene| C-band    | [100]     |
|                               | GNP/CLF/PEEK             | X-band    | [101]     |
|                               | Conventional RGO/PS      | K-band    | [98]      |
|                               | Segregated RGO/PS        | K-band    |           |
| 3D printing method            | Graphene/Li0.35Zn0.35Fe0.35O4/PDMA | 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 Continuous monitoring in air traffic control communication is applicable against electromagnetic interference | • Normalization and data scaling not required | • Time consuming |
| 2     | K-nearest neighbors (k-NN) algorithm | [123] | An effective approach to eliminate electromagnetic interference problems | • Training period not required • Easy implementation | • Not suitable for large and high dimension data |
| 3     | Artificial Neural Networks Backpropagation Neural Network | [124-128] [129, 130] | Use backward pass approach for parameters adjustment | • Fast and easy to use | • Problem identification is difficult • Actual performance depends on data input for problem solving |
| 4     | Self-Organizing Feature Map Neural Network | [131, 132] | Effective in evaluating the global features of electromagnetic interferences factors Effective for electromagnetic interference generated underground metallic pipelines, high voltage power lines and other problems | • Interpretation of data is easy • Grid clustering is helpful to evaluate the data similarity • Detect complex nonlinear relationship | • Slow training • Black box |
| 5     | Neural Networks | [133-137] | Effective in electromagnetic interference problems | • Flexible simulation | • Time consuming |
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 ultrasonication 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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