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THREE DIMENSIONAL GRAPHENE IN NANOCOMPOSITES—STRUCTURE-PROPERTY SCENARIOS AND EMI/GAMMA SHIELDING POTENTIAL

ABSTRACT

The present review article is planned to systematically unfold salient worth of three dimensional graphene based polymeric nanocomposites for radiation shielding (electromagnetic, nuclear, gamma, fast neutrons) purposes. As per literature reports so far, we discuss polymer/three dimensional graphene nanocomposites for variety of thermoplastic, thermoset, and conjugated matrices employed for related high end material designs. Accordingly, multifunctional hybrids of three dimensional graphene have been fabricated via facile/resourceful fabrication techniques, including solution processing, in situ method, melt technique, freeze drying, hydrothermal tactic, printing, foaming, and allied synthesis procedures. The ensuing three dimensional graphene based hybrids/nanomaterials have been analyzed for microstructural, structural integrity, electron/charge conduction, dielectric features, permittivity, radiation shielding effectiveness, shielding efficiency, and other features desirable for nuclear/gamma/electromagnetic radiation protection application. Besides, underlying mechanisms of radiation attenuation have also been argued, as per scientific surveys. It seems that performance of hierarchical graphene nanoassmblies relies upon nanoarchitectural adaptability, interfacial interactions/wettability, and structure-property synergies. Eventually, inimitable polymer/three dimensional graphene nanocomposites have been found promising to meet technological demands of radiation shielding in aeronautics, devices, defense, and nuclear power plant industries. Despite practical success of radiation shielding three dimensional graphene hybrids, in spite of pristine graphene, future deployment in related industrial modules seems to be connected to focused experimental/theoretical endeavors by field researchers to overcome underlying design/property/performance challenges.

Keywords: *Three dimensional graphene; nanocomposites; thermoplastics; radiation shields; electromagnetic; gamma rays; nuclear*

INTRODUCTION

In recent decades, owing to growing radiation pollution issues, continuously escalating scientific interest has been noticed towards development of competent nanomaterial shields against electromagnetic interference (EMI), nuclear, gamma, and other ecologically perilous radiations [1]. Out of carbon nanomaterials, two dimensional graphene nanosheets have been focused for radiation protection designs, due to intrinsically valuable surface properties, electron/charge conductivity, and mechanical stability features [2-4]. Particularly, high performance polymer and graphene based nanocomposites have been developed for EMI shielding application [5]. However, radiation shielding potential of polymer/graphene hybrids seemed to be restricted due to aggregation challenges of two dimensional graphene nanofiller in polymeric phases. Consequently, modified or altered graphene forms, like three

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dimensional graphene, have been considered for seamless dispersion in polymers for superior surface area, interface formation/miscibility, and structure-property consequences [6]. Such synergies in turn reveal facilitated electron/charge transfer thru polymer/hierarchical graphene architectures necessary for radiation absorption application [7,8]. Advanced EMI/gamma shields of three dimensional graphene based nanomaterials depicted noteworthy scope for energy devices, electronics, printing devices, sophisticated biomedical devices, and like so [9-11].

In view of practical worth of three dimensional graphene and derived hybrids towards radiation shielding and restricted literature reviews in the field, we document this all-inclusive manuscript as a useful guide for interested field scientists. To the best of the knowledge, this novel review article (for the first time) debate different categories of polymer/three dimensional graphene nanomaterials for relevant fundamentals, structural, morphological, processing, and EMI/gamma radiation shielding related applied attributes. Especial emphasis has been placed on key factors and mechanisms responsible for radiation defense competence of three dimensional graphene hybrids, counting nanolevel hierarchies, interfacial aspects, electrical percolation, and radiation attenuation characteristics. Herein, we see promising future applications of multifunctional polymer/three dimensional graphene nanocomposites for space/nuclear industries. Even so, future evolutions of these valuable nanomaterials depend upon new design innovations, optimum processing factors, structure-character-performance interlinks, and theoretical investigations to unveil true phenomenon and role of three dimensional graphene towards radiation protection application.

GRAPHENE AND ALLIED NANOCOMPOSITES FOR RADIATION SHIELDING

Technology growth in recent decades resulted in continuous emission of electromagnetic, gamma, nuclear, and other hazardous radiation from functional electronic, energy, and similar high tech devices and systems, as a serious environmental pollution [12]. To deal with this challenging situation, worldwide scientific are continuously struggling to design efficient radiation shields of high performance materials (metals, polymers, ceramics) [13]. In this concern, carbon nanomaterials (amorphous carbon, carbon nanofibers, carbon nanotube, graphene, etc.) also drove research interests due to facilitated electron transport features, ultimately leading to superior microwave absorption properties [14, 15].

Graphene, is one of the most exclusive discoveries among nanocarbons, has two dimensionally arranged sp^2 hybridized carbon atoms in a one atom thick nanosheet [16, 17]. In simple words, it is a single layer in stacked graphite nanostructure. Since discovery, graphene has been synthesized using wide ranging top down/bottom up fabrication techniques such as exfoliation, chemical vapor deposition, laser/plasma tactics, lithography, and chemical synthesis, to name a few [18]. Structural exclusivity of graphene can be seen in the form of noteworthy surface area, transparency, strength, charge/heat transfer, and innumerable advantageous physical characters [19]. With research advancements on graphene, altered graphene forms and derived nanomaterials have been reported depicting further superior property and performance benefits [20]. Being one of the most intensively explored nanocarbon, vast applied potential of graphene has been perceived for electronics (diodes, sensors, batteries), energy maneuvers (capacitors, photovoltaics, fuel cells), automotive/space, civil, textiles, medical, and related high tech industrial sectors [21, 22].

Particularly, adding nanocarbons in polymers demonstrated the formation of compatible nanocomposites with notable permittivity and radiation defense properties [23]. Nevertheless, as per literature reports to date, mechanism of EMI shielding seems to be a complex phenomenon involving absorption, reflection, and manifold internal reflections. For this purpose, Isari et. al. [24] investigated and proposed a comprehendible mechanism for EMI shielding, as given in Fig. 1. According to Figs. 1 a & b, EMI mechanism predominantly involves reflection of radiations and internal multiple reflections may occur due to radiation deflection inside a conducting shield. These processes of interactions of incident radiations with a shielding material usually rely upon its skin depth, i.e., intensity of incoming EM radiations decreases with increasing shield thickness (Fig. 1 c). Furthermore, depending upon the nature of shielding material (segregated, porous, or multilayered), overall internal scattering, wave transmission, and EMI defense performance may be affected (Figs. 1d-f).

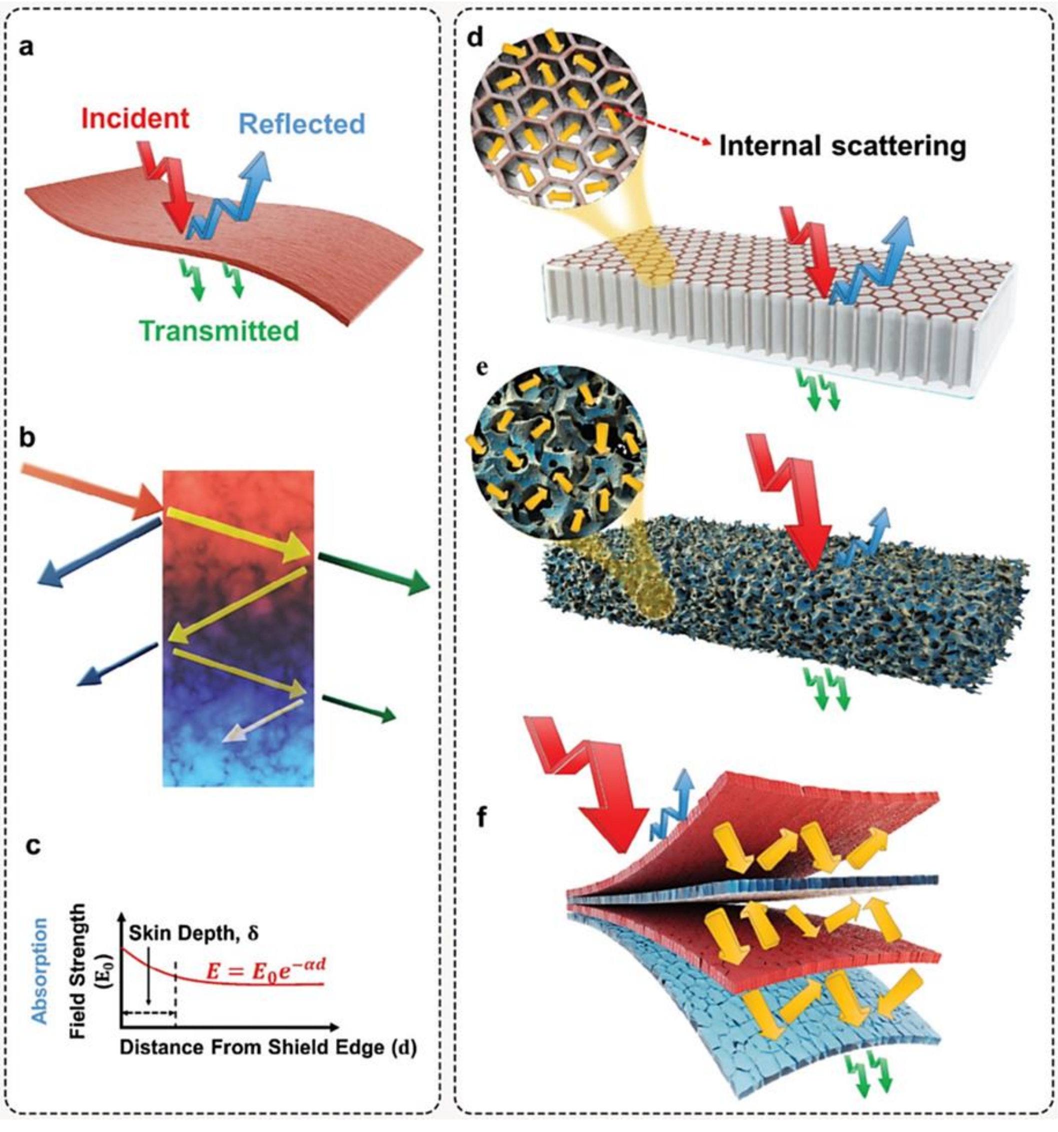

Fig. 1. (a & b) Electromagnetic (EM) interference shielding mechanisms in a highly conducting thin structure; (c) field strength vs. skin depth, i.e., distance from shield edge from where intensity of EM field decreases exponentially, 1/e, relative to initial incident value. Schematic of internal scattering in (d) segregated; (e) porous; and (f) multilayered structures [24]. Reproduced with permission from Wiley

Various carbon and polymeric EMI shielding materials/nanomaterials have been developed so far by field researchers [25, 26]. Here, electromagnetic shielding phenomenon appeared more complex in the case if multiphase materials, like nanocomposites [27]. Fig. 2 shows overtime progress of carbon nanomaterials/nanocomposites for radiation protection application, from employment of carbon black and carbon nanotube to graphene [28]. Thus, designed nanocarbon nanocomposites have been utilized for the fields of energy/electronics equipment and aerospace and defense related structures [29].

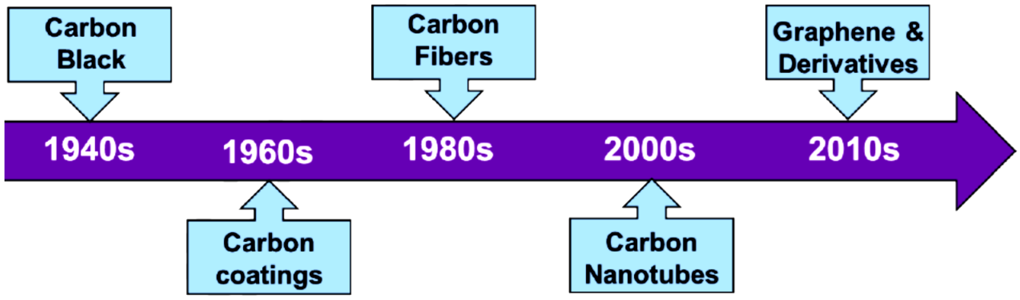

Fig. 2. History of carbon materials in electromagnetic interference shielding application [28]. Reproduced with permission from Elsevier

Among broad range of carbon nanomaterials surveyed for radiation defense purposes, graphene derived nanocomposites exhibited noteworthy potential towards shielding hazardous radiations in environment [30]. With polymers, synergistic effects of graphene or modified graphene (graphene oxide, reduced graphene oxide) have been reported via forming physical interactions, like hydrogen bonds, π-π stacking, covalent links, etc. [31][32]. Accordingly, graphene reinforced nanocomposites had high electrical/thermal conduction, thermal/mechanical stability, and allied superior physical characteristics for practical applications [33].

Out of myriad of polymer types, thermoplastics and thermosetting polymers have been reinforced with graphene to fabricate radiation defense shields [34]. As an example, Hamidinejad et. al. [35] formed polyethylene/graphene hybrids via foaming technique for radiation shielding purpose. Including 19 vol.% graphene depicted optimal EMI shielding effectiveness (~ 32 dB), probably due to structural compatibility of polyethylene and graphene. Shen et. al. [36] prepared polyurethane/graphene hybrids using a unique three dimensional saw tooth folding technique. The reported reasonably high EMI shielding effectiveness of ~ 20 dB. Pavlou et. al. [37] used chemical vapor deposition (CVD) technique for the formation of poly(methyl methacrylate) and graphene based nanocomposites having aligned nanolaminate nanostructures. A thin layer of graphene was initially deposited on copper foil using CVD technique, which was later spin coated with poly(methyl methacrylate) to form poly(methyl methacrylate)/graphene nanocomposites (Fig. 3 A). Here, lift-off method was used to attain thin films of < 100 nm thickness. Accordingly, Fig. 3 B portrays poly(methyl methacrylate)/graphene based thin samples with rising graphene contents. Fig. 3 C displays a scanning electron microscopy micrograph (cross-section) of a 0.13 vol.% nanocomposite film showing well aligned 250 nm thick graphene nanolaminates. Such microstructure indicated the development of a compatible interfacial nanostructure in these nanomaterials. Additionally, Fig. 3 D shows shielding effectiveness vs. frequency scans of 0.33 vol.% poly(methyl methacrylate)/graphene nanocomposite based shields prepared with varying thickness of 6-33 μm. Here, rising shield thickness was found to enhance the EMI shielding effectiveness of the nanocomposite, from 15-60 dB. Hence, both the graphene based nanocomposite design as well as shielding thickness were observed crucial for enhancing radiation shielding competence of nanomaterials. Lakshmi et. al. [38] studied poly(vinylidene fluoride)/graphene quantum dot hybrids, obtained by solvothermal and solution methods, for EMI defense. Here, shielding effectiveness was found to be 31-43 dB owing to electron conductivity and dielectric properties of these hybrids. Similarly, countless thermoplastic polymer/graphene systems have been designed and investigated in the literature hitherto, such as poly(methyl methacrylate) [39], poly(dimethyl sulfoxide) [40], blended forms [41, 42], and like so. Similarly, we found a number of research efforts on high end EMI shielding thermosetting polymer/graphene hybrids [43, 44].

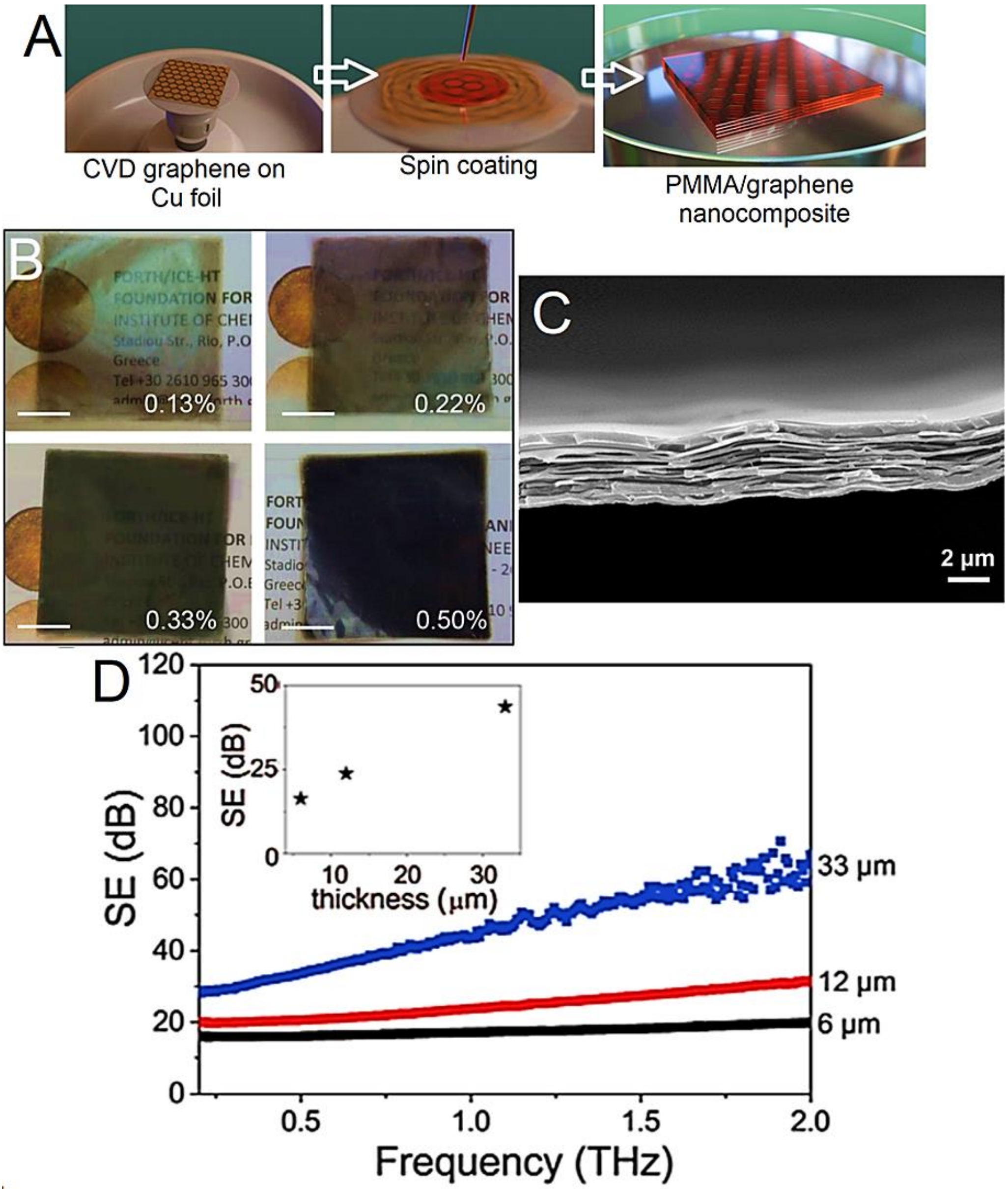

Fig. 3. (A) Chemical vapor deposition (CVD) based fabrication of poly(methyl methacrylate) (PMMA) and graphene derived nanocomposite nanolaminates; (B) photographs of nanolaminates prepared with increasing graphene contents (scale bar 1 cm); (C) scanning electron microscopy image of poly(methyl methacrylate)/graphene nanolaminates laminate (0.13 vol.% graphene) in cross-section plane; and (D) shielding effectiveness (SE) of nanolaminate with 0.33 vol.% graphene, inset: scans of SE vs. sample thickness [37]. Reproduced with permission from Nature

Besides, conjugated matrices (polyaniline, polypyrrole, polythiophene, derived forms) have been filled with graphene to manufacture radiation shields [45]. Conjugated polymers own intrinsically high electron conductivity and dielectric properties valuable for fine permittivity microwave absorption features [46, 47]. In combination with graphene or functional graphene forms, conductive polymers exhibited further superior electrical percolation features useful for radiation defense [48]. Khasim et. al [49] formed p toluene sulphonic acid doped polyaniline and graphene nanoplatelet based hybrids via in situ nanocompositing technique.

Here, doping and in situ processes were found effective to significantly enhance electron mobilization and dielectric permittivity features of the nanocomposites. Hence, remarkably high EMI shielding efficiency (> 95 %) was achieved for 10 wt.% graphene nanoplatelet loaded doped polyaniline nanocomposite.

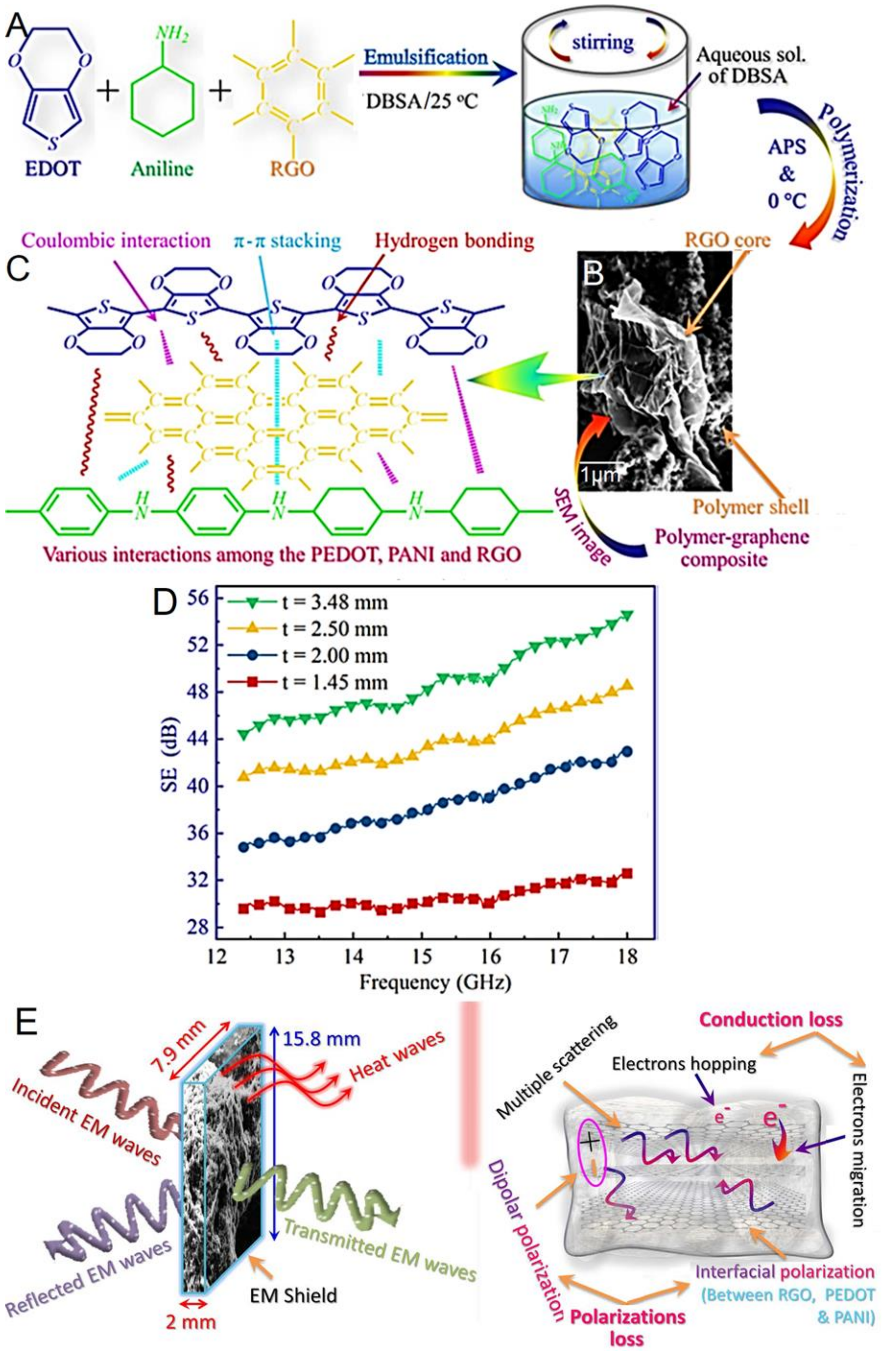

Fig. 4. (A) An in situ synthesis process for poly(3,4-ethylenedioxythiophene)/polyaniline/reduce graphene oxide hybrid; (B) scanning electron microscopy image of the nanocomposite; (C) scheme for interactions between poly(3,4-ethylenedioxythiophene) (PEDOT), polyaniline (PANI), and reduce graphene oxide (RGO) in hybrid; (D) electromagnetic interference shielding effectiveness of poly(3,4-ethylenedioxythiophene)/polyaniline/reduce graphene oxide nanocomposite having difference shield thicknesses; (E) mechanism of electromagnetic wave attenuation involving reflection, absorption, and manifold internal reflections [50]. EDOT = ethylenedioxythiophene); APS = ammonium persulfate; DBSA = dodecylbenzene sulfonic acid; EM = electromagnetic. Reproduced with permission from RSC

Focusing polythiophene derivative nanomaterials, Kumari et. al. [50] developed a hybrid system of poly(3,4-ethylenedioxythiophene)/polyaniline/reduce graphene oxide via *in situ* polymerization of ethylenedioxythiophene and aniline monomers using ammonium persulfate/dodecylbenzene sulfonic, in the presence of dispersed reduce graphene oxide. Fig. 4 A outlines the synthesis process used for the formation of these nanocomposites. Fig. 4 B depicts scanning electron microscopy micrograph of the as prepared nanocomposite. The wrinkled graphene nanosheet can be seen covered with thin layer of in situ formed polymers. Fig. 4 C shows nature of physical interactions, i.e., hydrogen bonding, π-π stacking, and electrostatic associations between graphene and macromolecular chains. Fig. 4 D portrays increasing shielding effectiveness of poly(3,4-ethylenedioxythiophene)/polyaniline/reduce graphene oxide nanocomposite (~ 33-55 dB) with growing material thickness (1.45-3.48 mm). These results can be attributed to effectiveness of conductive hybrid in exponentially declining intensity of incident radiation because of high electrical conductivity, in the range of ~20-74 S m^{-1}. Fig. 4 E shows dimensions of the nanocomposite shield used for EMI shielding as well as most probable radiation attenuation mechanisms involved for superior efficiency of these materials towards radiation defense. We suggest application of such high tech conjugated polymer/graphene based EMI shielding designs particularly for sophisticated electronic devices. Table 1 lists few so far reported high performance polymer/graphene hybrids for radiation shielding application.

Table 1. *Two dimensional graphene and derivative nanomaterials with electrical conductivity and electromagnetic interference (EMI) shielding competence*

Polymer	Nanoadditive	Electrical conductivity	EMI shielding (dB)	Ref.
Polyethylene	Graphene	9 orders of magnitude increase	32	[35]
Poly(vinyledene fluoride)	Graphene quantum dots	Dielectric constant 10^8-10^9	~ 31-43	[38]
Poly(dimethyl sulfoxide)	Graphene	~ 0.2 Scm^{-1}	~ 54	[40]
Polyaniline	Graphene	~ 500 Scm^{-1}	~ 21	[41]
Polyaniline	Magnetite functional graphene	~ 2.5 Scm^{-1}	~ 32	[42]
Epoxy	Reduced graphene oxide	~ 4.0 Scm^{-1}	~ 55	[43]
Polyaniline	Graphene nanoplatelet	Dielectric constant 18-21	EMI shielding efficiency > 95 %	[49]
Poly(3,4-ethylenedioxythiophene)/ polyaniline	Reduce graphene oxide	~ 20-74 S m^{-1}	33-35 dB	[50]
Poly(dimethyl sulfoxide)	Magnetite functional graphene	25 Scm^{-1}	249	[51]
Poly(vinylidene fluoride)	Graphene	0.02 Scm^{-1}	~ 32	[52]

POLYMER/THREE DIMENSIONAL GRAPHENE WITH ELECTROMAGNETIC SHIELDING COMPETENCE

In current electronic age, graphene and derived nanomaterials has attained a notable scientific stance among efficient carbon nanomaterials used to deal with environmental pollution of interfering radiations [53, 54]. Nonetheless, technical performance of graphene in polymer matrices seemed to be limiting owing to aggregation issues of high surface area two dimensional nanosheets [55-57]. To enhance the dispersion, interactions, compatibility, and overall performance with polymers, graphene has been modified in several ways, e.g., formation of three dimensional hierarchical graphene nanostructures in spite of two dimensional nanosheets [58, 59].

A three dimensional graphene nanostructure consists of hierarchically interconnected assemblies of graphene nanosheets via secondary interactions [60]. Three dimensional interlinking of graphene nanosheets has been reported to enhance aspect ratio, electron/heat/charge flow, semiconductivity, structural firmness (both mechanically and thermally), and elasticity of these porous nanoarchitectures [61]. In this regard, a number of experimental and theoretical scientific attempts have been reported stating unique structural organization, characteristic attributes, and applied potential of three dimensional graphene [60, 62]. Apropos, we note applications of three dimensional for environmental, defense, engineering, energy, and biomedical arenas [63].

Accordingly, polymer and three dimensional graphene based nanocomposites have been prepared via numerous facile means, like solution method, in situ tactic, melt blending, dipping process, etc. [64, 65]. Among notable physical characteristics of three dimensional graphene filled hybrids, mechanical/thermal constancy, electrical conductivity, percolation features, and allied advantageous characters can be seen in the literature so far [66, 67]. Here, researchers seemed to be focusing on enhancing interfacial compatibility of high surface area three dimensional graphene with different polymers, without any aggregation issues [68-70]. Consequently, optimal structure-property-performance profiles have been found responsible for next level industrial applications of high performance polymer/three dimensional graphene nanocomposites [71, 72]. Especially, electron transfer through hierarchical nanoassemblies and structural durability of three dimensional graphene seem to upsurge methodological aspects of polymers desirable for aerospace, electronics, energy, and related industries [73].

Thermoplastic matrices have been reinforced with three dimensional graphene to induce conductivity and percolation thresholds features [74]. Polystyrene is a commodity, low price, light weight, heat stable, and moisture resistant polymer [75, 76]. Among earlier attempts, Yan et. al. [77] reported on hierarchically nanoporous nanocomposites of polystyrene and three dimensional graphene using solution, compression molding, and salt leaching techniques. As given in Fig. 5 A, polystyrene microspheres and three dimensional graphene were solution processed and then foamed via mechanical mixing in presence of calcium carbonate and subsequent salt leaching.

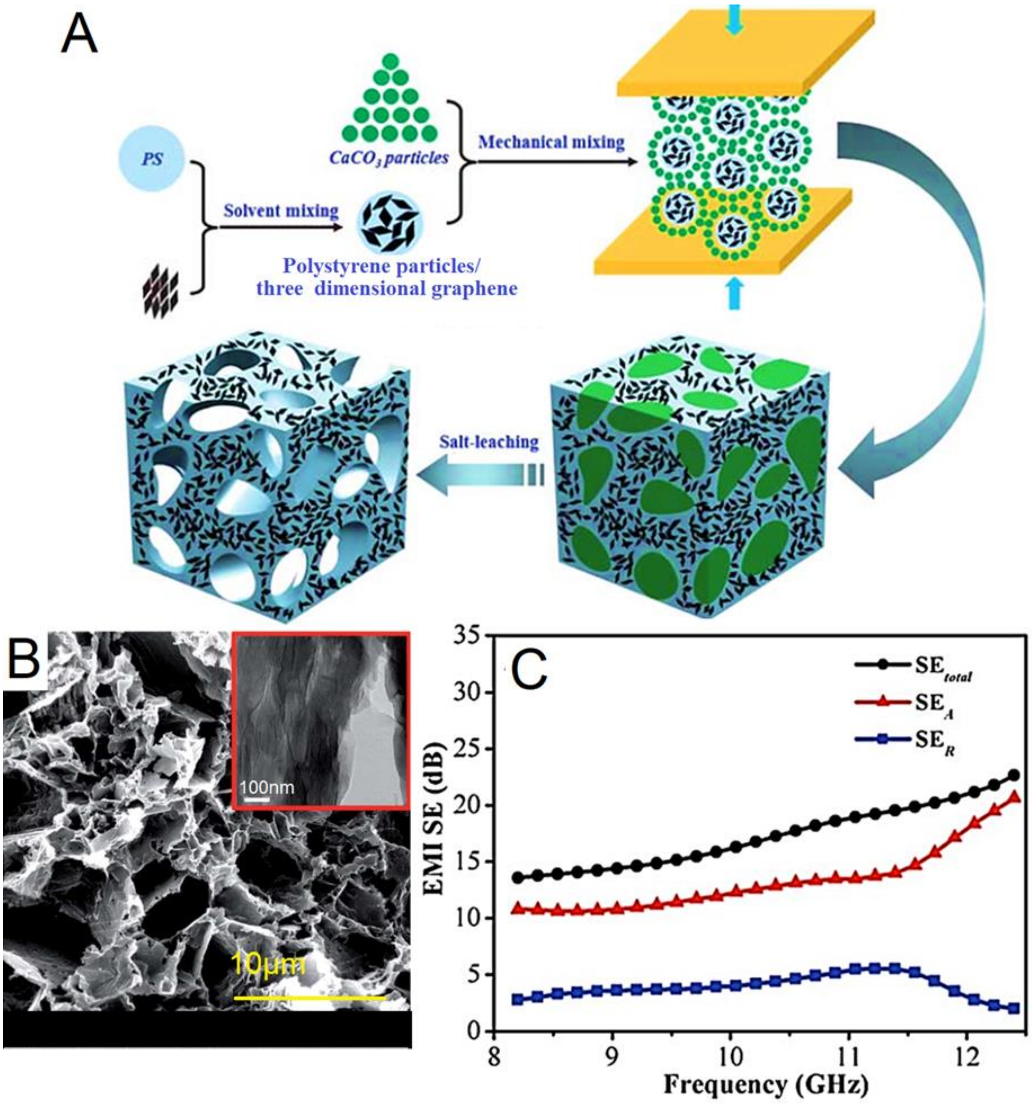

Fig. 5. (A) Manufacture of porous polystyrene (PS) and three dimensional graphene based nanocomposite through compression molding and salt leaching approaches; (B) field emission scanning electron micrograph of polystyrene/three dimensional graphene, where inset: transmission electron microscopy image of the same; (C) electromagnetic interference shielding effectiveness (EMI SE) of polystyrene/three dimensional graphene nanocomposite (8.2-12.4 GHz), where SE_{total} = total shielding effectiveness; S_{EA} = microwave absorption; S_{ER} = microwave reflection [77]. Reproduced with permission from RSC

Afterwards, compression molding (1000 MPa; 235 °C) was applied to form a 2.5 mm thick EMI shield. Fig. 5 B shows scanning electron microscopy and transmission electron microscopy micrographs of the as designed hierarchical polystyrene/three dimensional graphene hybrids. Here, three dimensional graphene seemed to be developing a compatible and well interlinked network within polymeric matrix. Such three dimensional nanoassemblies are usually beneficially for the formation of percolation networks for facilitated electron transfer through the nanocomposite system. Fig. 5 C expresses EMI shielding effectiveness of polystyrene/three dimensional graphene nanocomposite. Here, total shielding effectiveness, absorption, and reflection of ~ 17, > 13, and ~ 3.7 dB, respectively, were attained depicting microwave absorption as a dominant radiation protection phenomenon, rather than reflection. The corresponding EMI shielding effectiveness of > 64 dB cm^3 g^{-1} was observed for a 2.5 mm thick nanocomposite shield. Therefore, we note that most of microwave radiations were dissipated as heat through high performance polystyrene/three dimensional graphene nanocomposite.

We also note previous reports on three dimensional graphene filled poly(vinylidene fluoride) hybrids showing fine electrical conductivity and EMI shielding effectiveness of around 10^{-4} S m^{-1} and ~ 20 dB, respectively [78]. Amid later attempts, e.g., Sabira et. al. [79] designed low price, light weight, flexible and durable microwave absorption poly(vinylidene fluoride)/graphene nanomaterials. They documented superior EMI shielding effectiveness of ~ 47 dB, due to consist graphene nanoparticle dispersion and percolation network formation in the matrix.

Scientific literature also states three dimensional graphene filled polyurethane nanocomposites for radiation defense applications [80]. Gavgani et. al. [81] prepared three dimensional polyurethane/graphene oxide hybrids using in situ technique. The ensuing 1 wt.% nanofiller based hierarchical nanocomposites had notable electrical conductivity (> 4 S m^{-1}) and EMI shielding efficiency (> 250 dB g^{-1} cm^{-3}). Shen et. al. [82] manufactured polyurethane and graphene oxide derivative three dimensional nanocomposites via hydrothermal and dip coating processes (Fig. 6 A).

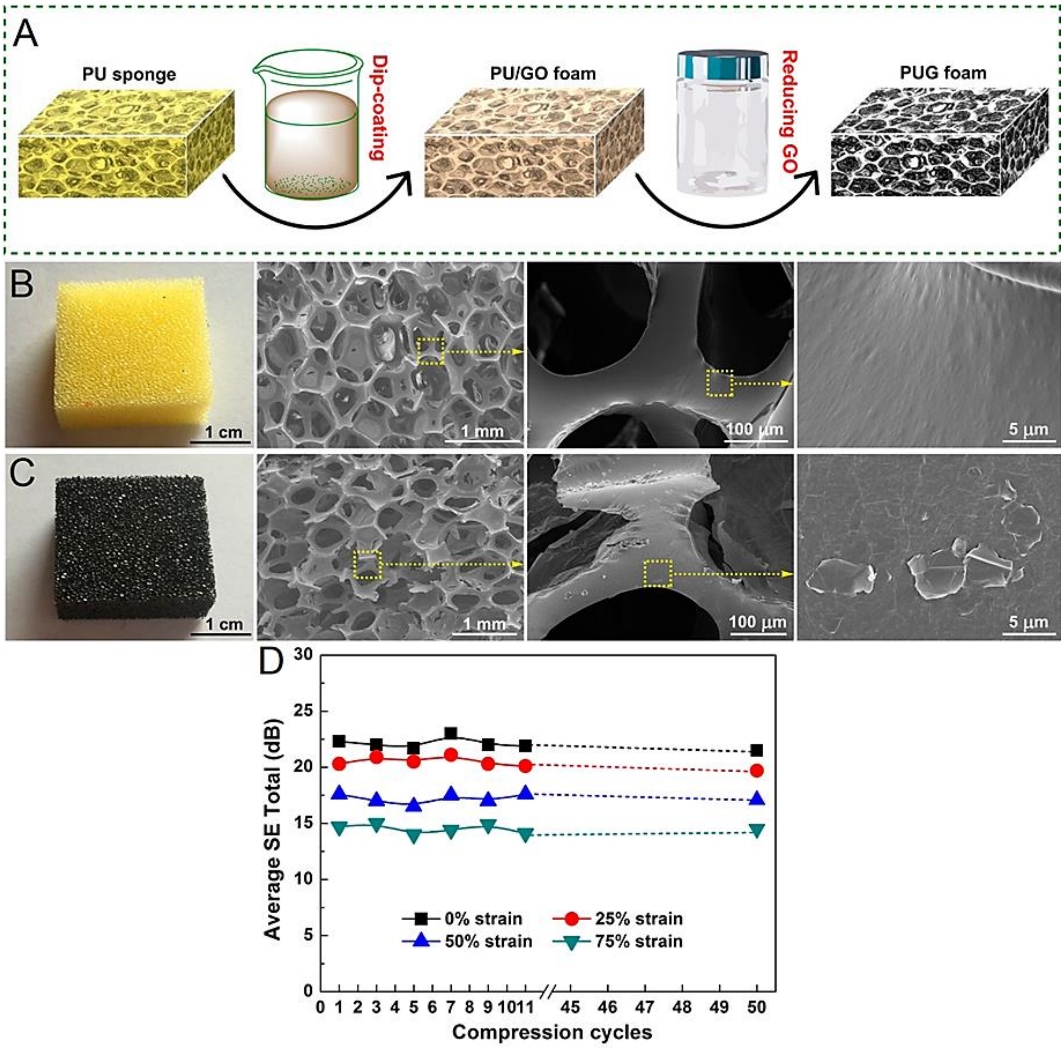

Fig. 6. (A) Manufacturing process for polyurethane (PU) and graphene oxide derived three dimensional nanocomposites; photographs and scanning electron microscopy images of (B) pristine PU nanofoam and (C) three dimensional polyurethane/graphene with 5 wt.% nanofiller; (D) shielding effectiveness (SE) total performance of three dimensional PU/GO tested under different compression cycles [82]. PU/GO = polyurethane/graphene oxide; PU/G = polyurethane/graphene. Reproduced with permission from ACS

As per visual images and scanning electron microscopy micrographs of pristine polyurethane nanofoam and 5 wt.% graphene oxide filled three dimensional nanocomposite (Figs. 6 B & C, respectively), uniformly aligned and hierarchically interconnected nanostructures were observed. Comparatively, three dimensional polyurethane/graphene oxide nanocomposite had a slightly visible rough surface morphology due to nanoparticle dispersion, relative to unfilled polymeric nanofoam (Fig. 6 C). Moreover, shielding effectiveness of three dimensional nanocomposite was analyzed for varying applied strains under respective compression cycles (Fig. 6 D). Here, EMI shielding effectiveness (15-23 dB) seemed to be unaffected by increasing applied strain up to 75 % (50 compression cycles). Such outcomes point towards the formation of robust interracially compatible hierarchical polyurethane/graphene oxide nanostructures competent for industrial level uses in aerospace and nuclear sectors.

Regarding thermosets, Wang et. al. [83] designed epoxy/polyaniline/three dimensional graphene nanocomposite as EMI shielding nanomaterials. As shown in Fig. 7 A, initially p-phenylenediamine modified graphene oxide was used to form three dimensional graphene by hydrothermal route. Later, freeze drying and curing methods were used to obtain epoxy/polyaniline/three dimensional graphene hybrid.

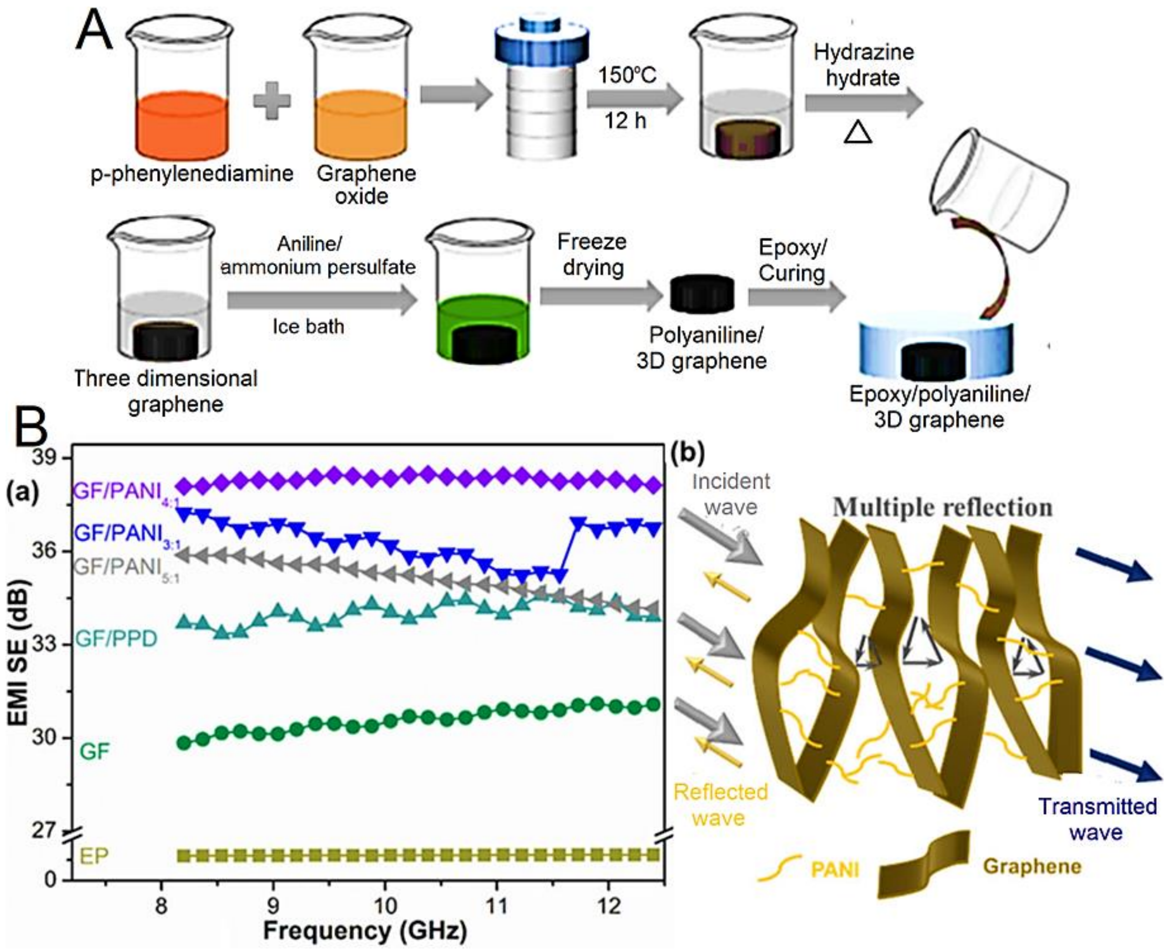

Fig. 7. (A) Synthesis of epoxy/polyaniline/three dimensional graphene nanocomposite; (B) (a) electromagnetic interference shielding effectiveness (EMI SE) of neat epoxy, three dimensional graphene, p-phenylenediamine grafted three dimensional graphene, and polyaniline and three dimensional graphene based nanocomposites (varying shield thickness); (b) schematic of electromagnetic interference shielding mechanism [83]. GF/PANI = three dimensional graphene/polyaniline; GF = three dimensional graphene; GF/PPD = three dimensional graphene grafted p-phenylenediamine; PANI = polyaniline; EP = epoxy. Reproduced with permission from MDPI

Fig. 7 B a shows EMI shielding effectiveness of pristine epoxy, phenylenediamine modified three dimensional graphene, and epoxy/polyaniline/three dimensional graphene hybrids with varying shield thicknesses. As per consequences, epoxy/polyaniline/three dimensional graphene hybrid shields had notably higher EMI shielding effectiveness (~ 36-38 dB), relative to pristine epoxy matrix (~ 3 dB). These results can be credited to compatible matrix-nanofiller interfaces and formation of hierarchically connected percolation networks in hybrids for rapid electron transport and effective microwave absorption. For these hierarchical nanoassemblies, EMI shielding mechanism was proposed to be multiple reflection and absorption occurring at compatible interfacial areas (Fig. 7 B b).

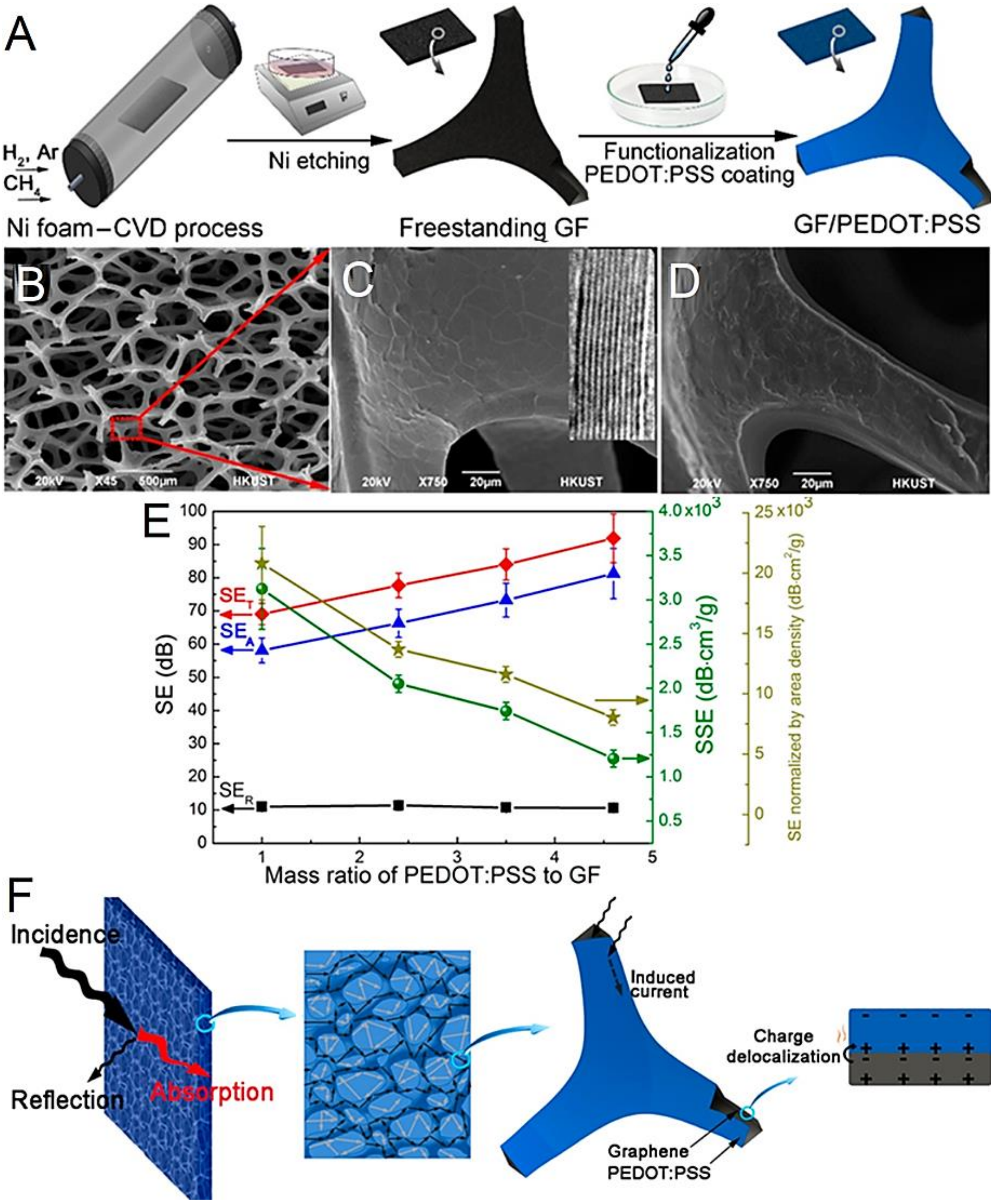

Fig. 8. (A) Manufacturing of three dimensional graphene/poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (GF/PEDOT:PSS) nanocomposites; (B & C) scanning and transmission electron microscopy micrographs, respectively, of three dimensional graphene (GF); (D) transmission electron microscopy micrograph of GF/PEDOT:PSS; (E) shielding effectiveness (SE) and specific shielding effectiveness (SSE) of PEDOT:PSS/GF; (F) a snapshot of electromagnetic shielding mechanism [87]. CVD = chemical vapor deposition; PEDOT:PSS = poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate). Reproduced with permission from ACS

Besides thermoplastics or thermosets, conductive matrices have also been amalgamated with three dimensional graphene to form microwave absorption materials [84-86]. As mentioned earlier, intrinsic electrical conductivity and percolation features of conjugated polymers have been found promising to synergistically enhance dielectric permittivity and radiation defense properties with graphene, especially three dimensional graphene nanostructures. Wu et. al. [87] fabricated EMI shields of poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) and three dimensional graphene derived hybrids. Fig. 8 A shows chemical vapor deposition synthesis of three dimensional graphene on a nickel foam template and then in situ deposition of poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) to form the nanocomposites. According to scanning and transmission electron microscopy micrographs in Figs. 8 B-D, pristine three dimensional graphene can be seen as free standing graphene hierarchical nanoassemblies, whereas uniform conductive polymer coating was observed over nanofoam surface, in the case of hybrid. Fig. 8 E shows shielding effectiveness and specific shielding effectiveness properties (> 90 dB and 3124 dB cm^3 g^{-1}, respectively) of poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate)/three dimensional graphene shield. Such high EMI shielding performance of these hybrids seemed to be due to their intrinsically superior surface area (1206 cm^3 g^{-1}) and electrical conductivity (> 43 Scm^{-1}) features. Fig. 8 F sketches probable EMI shielding mechanism for microwave absorption/dissipation thru high performance poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate)/three dimensional graphene based hybrid shield.

GAMMA/NUCLEAR RADIATION SHIELDING POTENTIAL OF POLYMER/THREE DIMENSIONAL GRAPHENE NANOCOMPOSITES

In addition to EMI shielding, promising materials have been devised for radiation protection against nuclear, gamma, and fast moving neutrons to be applied in nuclear power plants, defense, aerospace/automotive, and similar high tech industrial sectors [88, 89]. Initially, metals and ceramic materials were proposed for nuclear/neutron absorption purposes [90]. Later, surveys led to the deployment of high performance polymeric materials and nanomaterials to deal with hazardous gamma or nuclear radiations based environmental pollution [91-93]. In this concern, nanocarbon based designs employing graphene, modified graphene, or carbon nanotube depicted structural/thermal robustness in addition to nuclear shielding performance [94, 95]. Like EMI shielding, hierarchical graphene nanostructures have been designed and tested for nuclear/gamma shielding application [96]. For this purpose, Wang et. al. [97] formed three dimensional graphene and polyacrylamide derived nanocomposite shields using facile solution method. Here, remarkably high electrical conductivity of > 3000 S m^{-1} was attained owing to percolation effects in hierarchically porous nanostructures. Consequently, polyacrylamide/three dimensional graphene nanocomposites showed superior γ radiation shielding effectiveness of ~ 10-25 dB due to interfacial wettability and structural synergies. These gamma shields of three dimensional graphene based nanocomposites can be suggested for nuclear power plant application. In another research attempt, Wang et. al. [98] fabricated epoxy and three dimensional graphene derivative nanomaterials via resin transfer molding and annealing (200-800 °C) methods. Adding three dimensional graphene lead to 50-80 % enhancements in compressive/flexural strength of the thermosetting nanocomposites. Moreover, the nanocomposites depicted > 90 % gamma ray shielding competence under intense gamma irradiation environment. Hashemi et. al. [99] proposed epoxy and lead oxide functional graphene oxide based hybrids, prepared by vacuum shock technique, for radiation shielding. The 4-6 mm thick nanocomposite shields had > 100-120 % superior radiation attenuation rate, relative to pristine epoxy resin. Guo et. al. [100] used freeze drying and extrusion based three dimensional techniques for the formation three dimensional graphene oxide based aerogels. With shield thickness of 3 mm, superior electron conductivity and radiation shielding effectiveness (> 700 S m^{-1} and ~ 70 dB, respectively) have been observed. A recent report by Orikasa et. al. [101] depicted high performance three dimensional graphene based nanostructures for gamma shielding. These nanomaterials had notable neutron radiation shielding competence with mass absorption coefficient ~ 15 cm^2 g^{-1}. The versatile neutron shielding nanomaterials can be recommended for space exploration purposes. Despite the success of three dimensional graphene based nanomaterials in gamma/nuclear radiation shielding, research efforts seemed to be limiting in this area, as compared to their abundant use in EMI shielding application. Therefore, we suggest focused and comprehensive future investigations on novel three dimensional graphene nanocomposites revealing commercial scale gamma/nuclear radiation defense performance.

FUTURE PREDICTIONS AND CONCLUSIONS

Combination of unique hierarchical three dimensional graphene nanostructures with polymers resulted in innumerable technological advantages of these nanomaterials, including EMI shielding effectiveness and nuclear/gamma and neutron shielding potential (Table 2) [102]. In the field of ecologically hazardous radiation defense applications, research surveys up till now showed noteworthy design versatilities, processing feasibilities, and desirable radiation attenuation and absorption performance (Fig. 9). In this concern, we note myriad of polymeric matrices, like polyethylene, polyurethane, poly(vinyl fluoride), epoxy resin, polyaniline, and polythiophene derivatives (to mention a few), employed with graphene aerogels using simplistic processing methods [103]. Basically, three dimensional hierarchies of graphene aerogels seemed to be capable of inducing facilitated electron percolation pathways in the macromolecular phases [104, 105]. For this, compatibility of three dimensional graphene with polymers rely on the extent of interfacial wettability via physical or covalent association between the matrix-nanofiller phases [106]. Consequently, microstructural synergies facilitated electron/charge transfer thru polymer/three dimensional graphene hybrids [107]. Subsequently, as per literature, superior electrical conductivity corelates to significant permittivity, radiation attenuation, and radiation absorption effectiveness [108]. Here, manufacturing technique, conditions, and parameters have also been considered as critical factors influencing electromagnetic and gamma/nuclear shielding competence of polymer/three dimensional graphene nanomaterials [109]. Looking at the radiation attenuation performance of physically/chemically compatible interfacial hierarchies, we suggest use of modified three dimensional graphene, instead of pristine graphene aerogels [110]. Furthermore, we note shield thickness as a key factor for enhancing electromagnetic and gamma radiation effectiveness, due to exponentially interlinked rising trends between them [111]. At this point, we noticed identification of both the optimal nanofiller contents as well as material thickness significant for designing advanced polymer/three dimensional graphene based radiation shields. Last of all, for future advancement in this field and to meet technological demands of next level aerospace, defense, and nuclear sectors, we suggest green or biodegradable polymers as well as green synthesis tactics adopted to attain sustainable three dimensional graphene base hierarchical nanomaterials.

Table 2. *Aspects of three dimensional graphene hybrids for radiation shielding*

Polymer	Nanoadditive	Processing	Physical features/EMI shielding	Ref.
Polystyrene	Three dimensional graphene	Solution method; compression molding; salt leaching	2.5 mm thick EMI shield; total shielding effectiveness, absorption, and reflection ~ 17, > 13, and ~ 3.7 dB, respectively; EMI shielding effectiveness > 64 dB cm^3 g^{-1}	[77]
Poly(vinylidene fluoride)	Three dimensional graphene	Solution processing	Electrical conductivity and EMI shielding effectiveness 10^{-4} S m^{-1} and ~ 20 dB, respectively	[78]
Poly(vinylidene fluoride)	Three dimensional graphene	Solution processing	EMI shielding effectiveness ~ 47 dB; percolation network	[79]
Polyurethane	Three dimensional graphene oxide	In situ technique	Electrical conductivity > 4 S m^{-1}; EMI shielding efficiency > 250 dB g^{-1} cm^{-3}	[81]
Polyurethane	Three dimensional graphene oxide	Hydrothermal; dip coating	EMI shielding effectiveness 15-23 dB; EMI shielding effectiveness vs. applied strain/ compression cycles	[82]
Epoxy/polyaniline	p-phenylenediamine modified three dimensional graphene	Hydrothermal; solution processing; curing	Higher EMI shielding effectiveness ~ 36-38 dB, than pristine epoxy (~ 3 dB)	[83]
Poly(3,4-ethylenedioxythiophene): poly(styrene sulfonate)	Three dimensional graphene	Chemical vapor deposition; nickel foam template; in situ technique	Surface area 1206 cm^3 g^{-1}; electrical conductivity > 43 Scm^{-1}; shielding effectiveness and specific shielding effectiveness > 90 dB and 3124 dB cm^3 g^{-1}, respectively	[87]
Polyacrylamide	Three dimensional graphene	Solution processing	Electrical conductivity > 3000 S m^{-1}; γ radiation shielding effectiveness ~ 10-25 dB	[97]
Epoxy	Three dimensional graphene	Resin transfer molding; annealing (200-800 °C)	50-80 % enhancements in compressive/flexural strength; > 90 % gamma ray shielding efficiency	[98]
Epoxy	Lead oxide functional graphene oxide	Vacuum shock technique	4-6 mm thick shields; > 100-120 % nuclear radiation attenuation rate	[99]
Thermoset	Three dimensional graphene	Freeze drying; extrusion	Shield thickness 3 mm; conductivity and gamma radiation shielding effectiveness > 700 S m^{-1} and ~ 70 dB, respectively	[100]
Thermoset	Three dimensional graphene	Solution	Neutron radiation shielding; mass absorption coefficient ~ 15 cm^2 g^{-1}	[101]

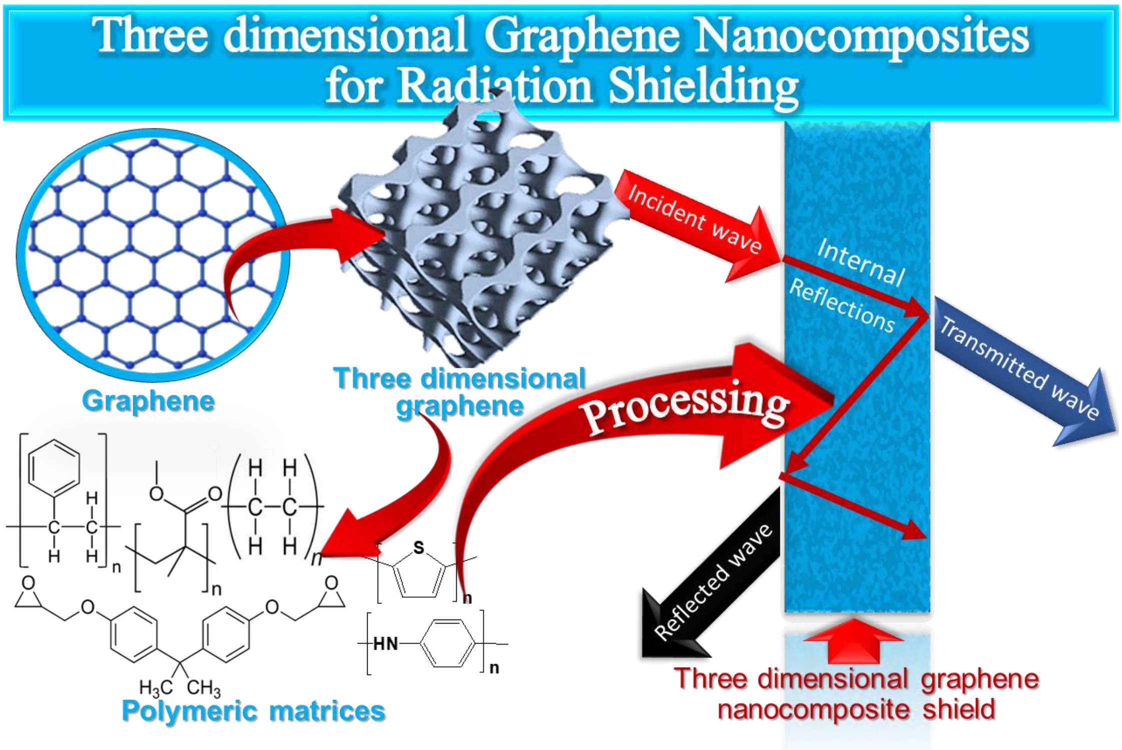

Fig. 9. Three dimensional graphene nanocomposites for radiation shielding

This review article is aimed to recite potential of three dimensional graphene and derived nanocomposites for radiation shielding (both EMI shielding and nuclear/gamma ray defense). Three dimensional graphene has been reinforced in different polymeric matrices (thermoplastics, thermosets, conductive polymers) to form hierarchical nanostructures having anomalous light weight, rapid electron transport, mechanical stability, dielectric permittivity, microwave absorption, and nuclear radiation attenuation characteristics. In this concern, a number of facile processing techniques have been practiced towards compatible and interracially well interlinked nanostructures. Numerous advantageous structural and physical characteristics of graphene aerogels have been analyzed in the literature so far, relative to two dimensional graphene. Notwithstanding the success of hierarchical graphene nanostructures as radiation absorption shields, scientific knowledge seems limiting on crucial structure-property aspects of these nanocomposites. Nevertheless, future progress of polymer/three dimensional graphene nanomaterials relies on exploring novel structural combinations and uses of experimental and theoretical methods to investigate microstructural, interfacial, physical, conducting, dielectric, and EMI and nuclear radiation shielding effectiveness. In short, concentrated future studies regarding structure-property-performance prospects of polymer/three dimensional graphene nanoarchitectures may unveil high tech designs for commercial scale utilizations in aeronautical/military and nuclear power plants related applications.

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