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Article

Graphene quantum dot for thermoplastic nanocomposites—Scope and opportunities

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Abstract: Quantum dot can be seen as an amazing nanotechnological discovery, including inorganic semiconducting nanodots as well as carbon nanodots, like graphene quantum dots. Unlike pristine graphene nanosheet having two dimensional nanostructure, graphene quantum dot is a zero dimensional nanoentity having superior aspect ratio, surface properties, edge effects, and quantum confinement characters. To enhance valuable physical properties and potential prospects of graphene quantum dots, various high-performance nanocomposite nanostructures have been developed using polymeric matrices. In this concern, noteworthy combinations of graphene quantum dots have been reported for a number of thermoplastic polymers, like polystyrene, polyurethane, poly(vinylidene fluoride), poly(methyl methacrylate), poly(vinyl alcohol), and so on. Due to nanostructural compatibility, dispersal, and interfacial aspects, thermoplastics/graphene quantum dot nanocomposites depicted unique microstructure and technically reliable electrical/thermal conductivity, mechanical/heat strength, and countless other physical properties. Precisely speaking, thermoplastic polymer/graphene quantum dot nanocomposites have been reported in the literature for momentous applications in electromagnetic interference shielding, memory devices, florescent diodes, solar cells photocatalysts for environmental remediation, florescent sensors, antibacterial, and bioimaging. To the point, this review article offers an all inclusive and valuable literature compilation of thermoplastic polymer/graphene quantum dot nanocomposites (including design, property, and applied aspects) for field scientists/researchers to carry out future investigations on further novel designs and valued property-performance attributes.

Keywords: thermoplastics; graphene quantum dot; nanocomposite; EMI shielding; memory devices; bioimaging

1. Introduction

Continuous advancements in the field of high-performance nanocomposites have introduced wide-ranging nanoadditives to upsurge intrinsic features of matrices, especially polymers [1]. For polymeric nanocomposites, carbonaceous nanoparticles have been distinctively used to attain desired physical properties and performance [2]. In addition to unique carbon nanoparticles, like graphene, carbon dots own further enhanced physical properties and applications [3]. In polymeric matrices, adding graphene quantum dots has been documented to develop compatible matrix-nanofiller associations depending upon its surface functionalities, matrix type, and processing strategies used [4]. Particularly, polymer/graphene quantum dot nanomaterials may have physical interactions (hydrogen bonding, electrostatic, van der Waals, etc.) [5] or covalent bonding to form robust interfacial contours [6]. Thermoplastic polymers (a major polymer grouping) with graphene quantum dots have been investigated for superior microstructure, strength, heat stability, electrical/thermal conductivity, and a

number of allied properties leading to technical end uses [7]. Incidentally, thermoplastic polymer/graphene quantum dot nanocomposites revealed applied potential for devices, radiation shields, and biological arenas [8,9].

This comprehensive review article covers essential design, characteristics, and technical aspects of multifunctional thermoplastic polymer/graphene quantum dot nanocomposites prepared via facile processing techniques. In this concern, we have tried to report almost all important thermoplastic matrices explored so far in the literature with graphene quantum dots for significant structural, physical, and applied characters. Herein, the type of thermoplastic matrix and graphene quantum dot surface functionalities govern their mutual amalgamation and final structure-property profiles needed for next-level industrial applications.

2. Thermoplastic nanocomposites

The thermoplastics or thermoplastic polymers form a major cataloging of synthetic as well as naturally occurring polymers [10]. Thermoplastic polymers are either amorphous or semicrystalline in nature owing to the distribution of high molecular weight polymeric chains [11]. In semicrystalline thermoplastics, a few of the macromolecular chains may get orderly arranged to form small ordered chain fragments, rendering partial crystallinity to these polymers [12]. Conversely, amorphous polymers own nearly all randomly oriented chains throughout the polymer, so having no crystallinity in the backbone [13]. These polymers usually have restricted temperature ranges for rubbery to glassy state transformations and so show phase transformations above their glass transition temperatures [14]. Notable physical, electrical, thermal, barrier, anticorrosion, and chemical properties of lightweight and low-cost thermoplastics have been reported for methodological applications in a myriad of applications, including automotive, civil, households, toys, cosmetics, pharmaceuticals, etc. [15].

To enhance essential features and performance aspects of thermoplastic polymers, inorganic and carbon additives or nanoadditives have been used to form composite or nanocomposite materials [16]. An important use of nanoadditives in thermoplastic polymers has been observed in upsurging the intrinsically low mechanical properties of these matrices via nanoparticle dispersion and interface formation effects [17,18]. In addition, nanoparticle dispersion may form a percolation network throughout the polymers for facilitated electron, charge, or ion flow through the nanocomposite phases [19]. In these nanocomposites, the increase in physical properties seemed to be reliant upon nanofiller functionalities, amount/orientation in the matrix, and interaction/compatibility with matrices [20–22]. In this way, by optimizing the nanofiller type, amount, and surface properties, desired high-performance thermoplastic nanocomposites have been reported [23].

3. Graphene quantum dot

A quantum dot is a nanoentity having a size of a few nanometers, usually 2–10 nm, an enormously high surface area, surface features, and marvelous electronic, optical, fluorescence, and physical attributes [24]. Quantum dots can be formed using inorganic or organic materials, and so resulting nanoparticles may have distinct

features and technical significance [25]. Among organic quantum dots, carbon-based nanodots of 5–10 nm in size have been reported [26]. Carbon-structured quantum dots can be broadly listed as polymer dots, carbon nanodots, and graphene quantum dots depending upon their backbone compositions, as presented in **Figure 1** [27].

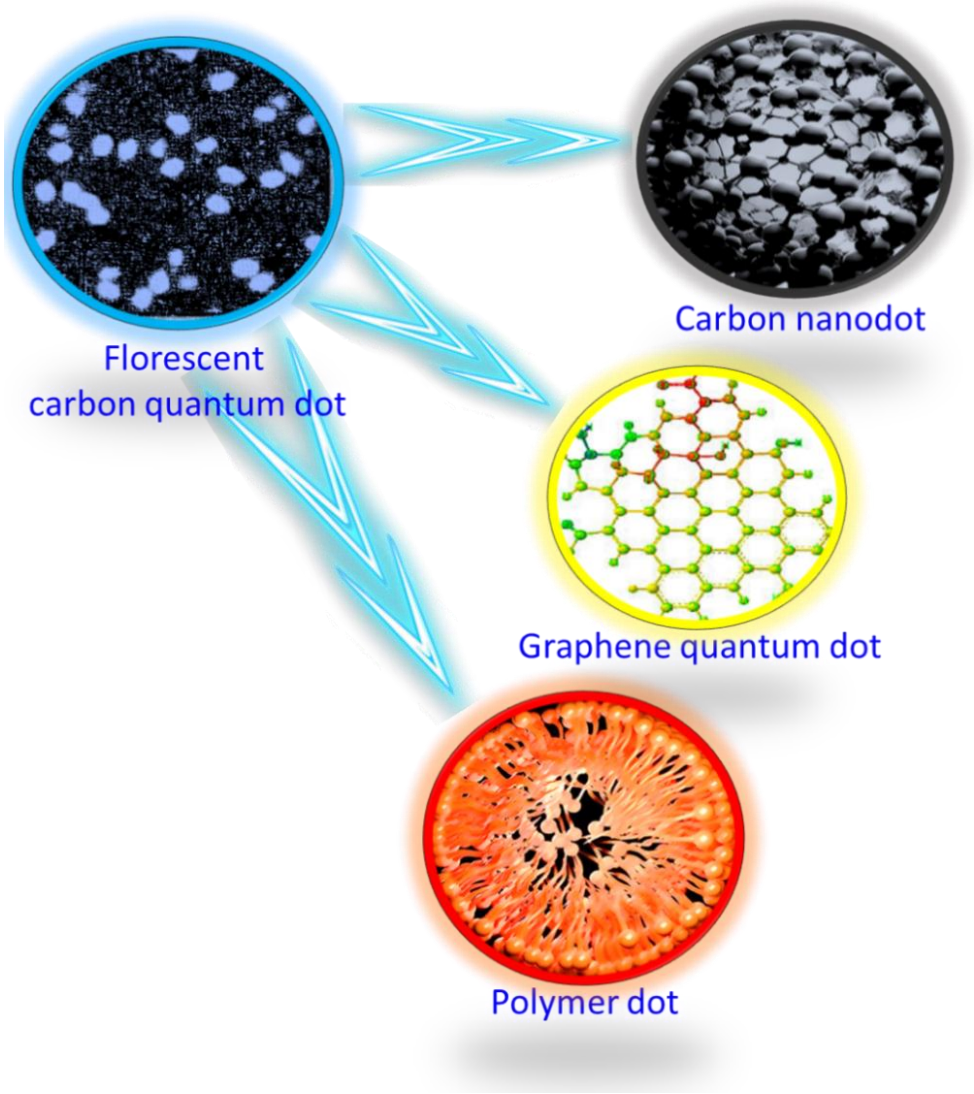

Figure 1. Carbon structured quantum dots.

Owing to the graphene backbone, the graphene quantum dot owns sp^3 and sp^2 hybridization in its structure [28]. However, unlike parent graphene, the quantum dot has zero dimensions, a size of < 10 nm, and quantum-related effects (**Figure 2**) [29]. Due to armchair or zigzag edge-related effects, valued electronic, optical, photoluminescence, magnetic, non-toxic, and biocompatible properties have been observed for graphene quantum dots [30]. Here, it is important to understand key differences between carbon quantum dots and graphene quantum dots. Firstly, carbon quantum dots have sp^3 hybridization in structure, whereas graphene quantum dots are sp^2 hybridized; secondly, carbon quantum dots are amorphous, while graphene quantum dots are crystalline in nature, having relatively superior semi-conductivity properties; third, carbon quantum dots usually have a diameter/nanosize of < 10 nm,

while on the other hand, graphene quantum dots may have a size of up to 2–20 nm to show the fluorescence phenomenon; finally, carbon quantum dots can be synthesized using any type of carbon precursor; conversely, precursors for graphene quantum dots must be some graphene-based material. Due to progressive research on graphene quantum dot, a range of top down (carbon nano-cutting, exfoliation, solution, etc.) and bottom up (hydrothermal, chemical vapor deposition, microwave, plasma, electrochemical, chemical, etc.) synthesis methods have been used [31]. Ensuing graphene quantum dots have been examined in the literature for capacitors, batteries, LEDs (light-emitting diodes), solar cells, sensors, NEMS (nanoelectrochemical systems), labs on a chip, and biomedical sectors [32]. Another worthwhile application of graphene quantum dot has been observed for the formation of nanocomposites having superior physical features and applied prospects [33].

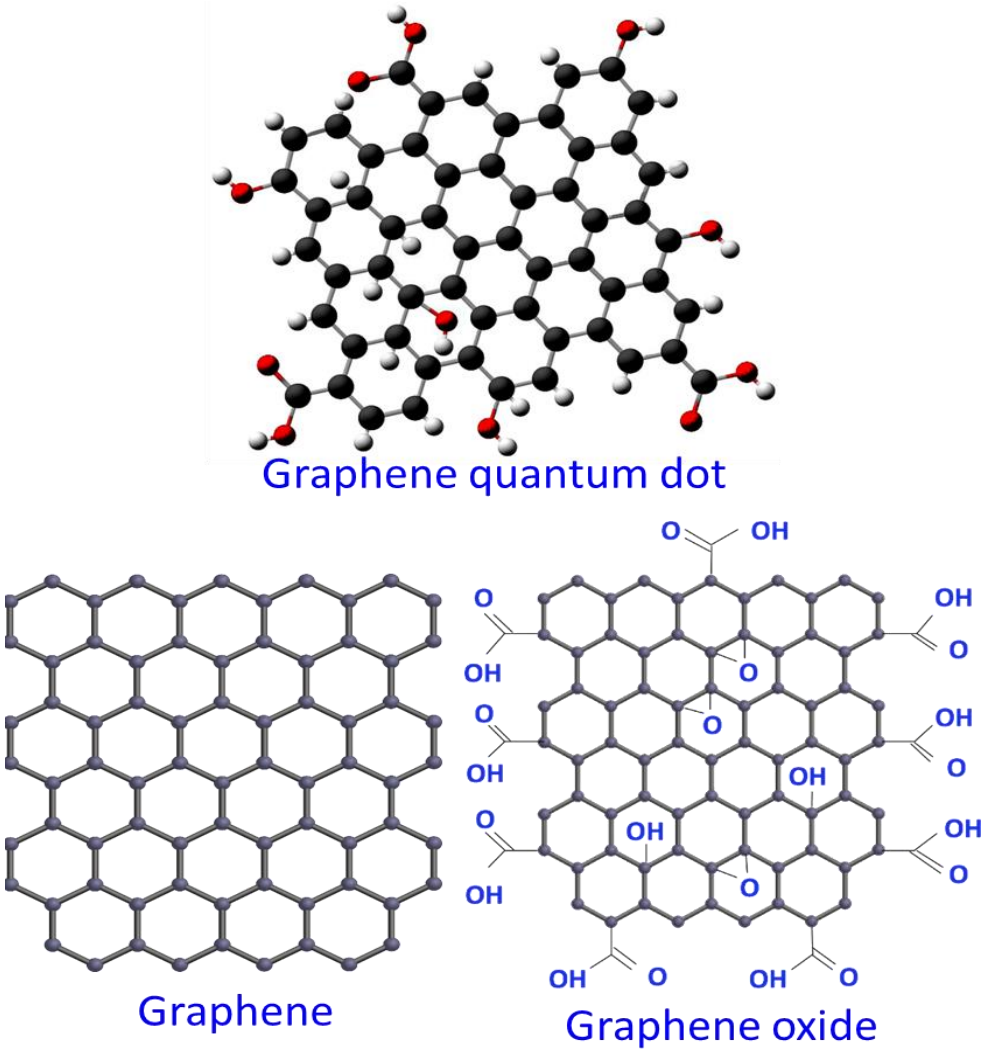

Figure 2. Structures of graphene, graphene oxide, and graphene quantum dots.

4. Thermoplastic/graphene quantum dot nanocomposites

Thermoplastic polymer/graphene nanocomposites have been frequently explored in the literature using innumerable matrices (polyethylene, polystyrene, poly(vinyl fluoride, nylon, and so on), synthesis techniques, advantageous physical properties and applications [34–36]. Similarly, graphene quantum dots have been reinforced in the thermoplastic matrices to attain versatile nanocomposite nanomaterials [37,38].

Among thermoplastics, polystyrene is one of the most recurrently used commodity polymers, having low cost, light weight, and facile processing [39]. In spite of this, brittleness of polystyrene limits its potential applications in significant engineering fields [40]. Consequently, recent researches on polystyrene suggest its processing as nanocomposite materials employing beneficial nanoadditives, like graphene, to enhance its inherent properties [41]. Hence, reports on superior mechanical, thermal, and physical features of polystyrene/graphene nanomaterials suggest further use of graphene derivatives (e.g., graphene quantum dot) to form high performance nanocomposites [42].

Ma et al. [43] formed carbon-functional graphene quantum dots via the pyrolysis technique and processed them with polystyrene using the Pickering emulsion polymerization method, as outlined in **Figure 3A**. The resulting polystyrene/carbon functional graphene quantum dot nanocomposites with 0.1–2 wt.% nanodot contents were investigated for microstructure, thermal, and flame resistance properties. **Figure 3B** (**a & b)** show scanning electron microscopy micrographs for neat polystyrene and polystyrene/carbon functional graphene quantum dot nanocomposite, respectively. As per results, pristine polystyrene microspheres formed by Pickering emulsion method had uniform surface. On the other hand, surface roughness and wrinkling of the nanocomposite microspheres can be observed in the micrographs due to the deposition of polymer chains. According to thermogravimetric analysis, neat polystyrene had one-step degradation (350 °C–450 °C), whereas 860 W g^{-1} nanocomposite showed additional weight loss steps (280 °C–380 °C and 520 °C–600 °C) due to the presence of nanodot nanoparticles (**Figure 3C**). **Figure 3D** shows heat release rate curves for pristine and composited polystyrene-based nanomaterials. Here, the decrease in the peak heat release rate of unfilled polystyrene (~860 W g^{-1}) in polystyrene/carbon functional graphene quantum dot (~520 W g^{-1}) confirmed the enhancement in the flame resistance of the nanocomposites with the nanodot addition. This fact was explained on the basis of the formation of a flame protection layer causing a barrier effect by quantum dot nanoparticles, so limiting the decomposition of polystyrene during the combustion process (**Figure 3E**). The overall enhancement in heat/flame resistance of the nanocomposites can be credited to the modified graphene quantum dot-based designs as well as the effectiveness of the fabrication technique used in this attempt. However, literature shows limited reports on polystyrene/graphene quantum dot nanocomposites.

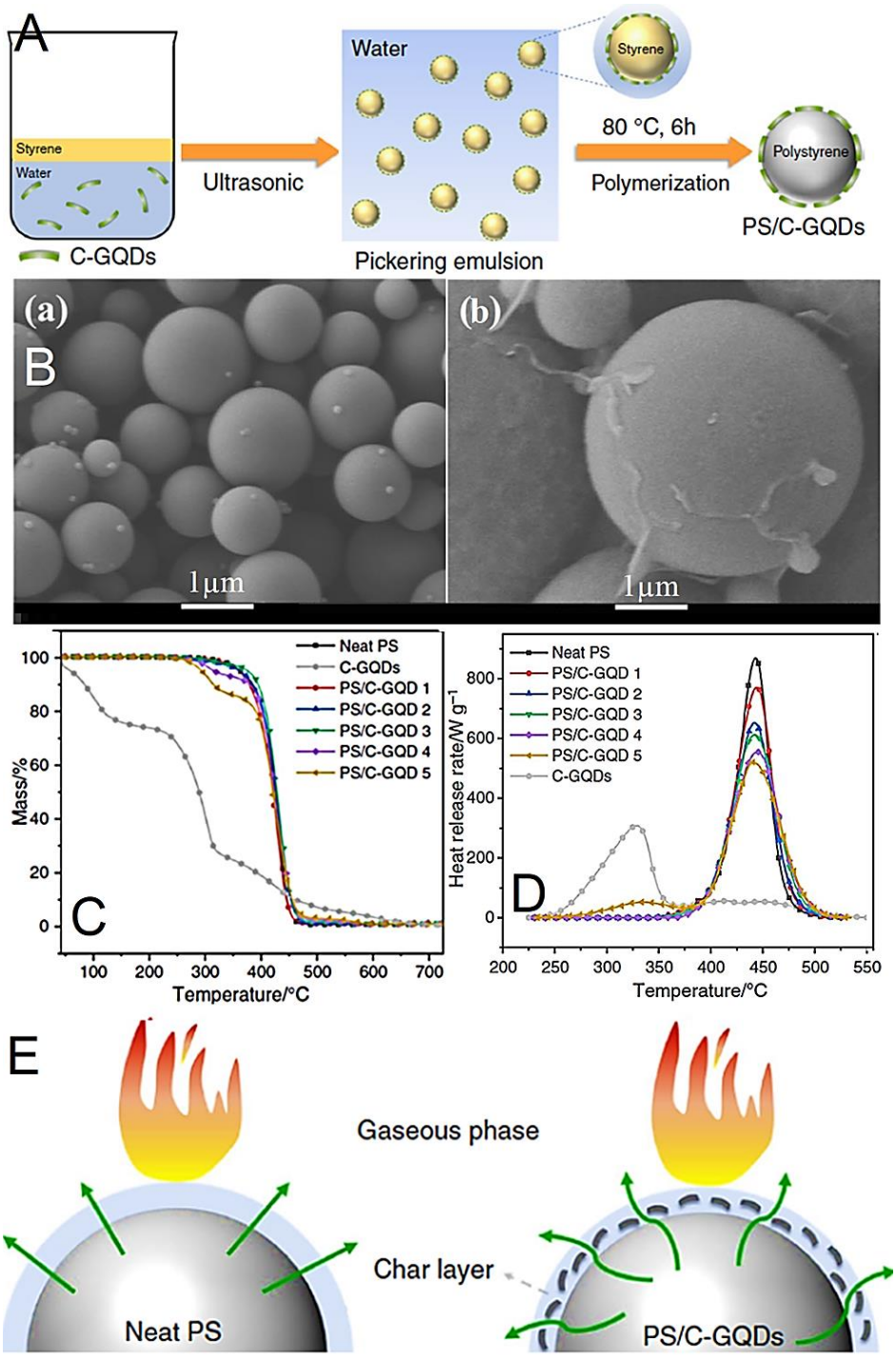

Figure 3. (A) Schematic for formation of PS/C-GQDs nanocomposites; **(B)** scanning electron microscopy images for **(a)** neat PS and **(b)** PS/C-GQD 5; **(C)** thermogravimetric analysis curves of PS and PS/C-GQDs nanocomposites; **(D)** heat release rate (HRR) curves of PS and PS/C-GQDs nanocomposites; and **(E)** mechanisms of flame retardancy of PS nanocomposites [43]. PS = polystyrene; C-GQDs = carbon-modified graphene quantum dots; PS/C-GQDs = polystyrene/carbon-modified graphene quantum dots. Reproduced with permission from Springer.

Polyethylene is another common low-price plastic material having high molecular weight and hydrophobic properties [44]. Moreover, polyethylene has fine flexibility, toughness, chemical resistance, etc., for commercial applications [45]. It is commonly practiced as low-density polyethylene, high-density polyethylene, as well as ultra-high molecular weight polyethylene forms [46]. To upsurge the industrial significance, polyethylene was composited with metal or inorganic nanoparticles as well as carbon nanoadditives, like graphene, carbon nanotube, etc. [47]. Accordingly, plenty of research can be seen for polyethylene/graphene nanocomposites [48]. Few attempts have been observed on polyethylene/graphene quantum dot hybrids in the literature to date. For example, Yin et al. [49] investigated polyethylene/carbon quantum dot/silica for microstructural and fluorescent emission spectral behavior. For this purpose, Stöber method was used to form silica microspheres and then polyethylene/carbon quantum dot/silica were designed using compression molding approach. Using this method, polyethylene chains were believed to be confined between silica nanoparticles to ensure fine dispersion and interaction towards the nanodot (**Figure 4A**). Accordingly, high resolution transmission electron microscopy of the nanocomposite depicted lattice spacing (~2.1 Å) and lattice plane (100) confirming the structural integrity and dispersion of carbon quantum dot in the nanocomposite (**Figure 4B**). **Figure 4C** illustrates fluorescent emission spectra of polyethylene/carbon quantum dot/silica hybrid with red shift (340 to 380 nm) and blue shift (380–500 nm) at lower and higher wavelengths, respectively. Such results also confirmed the surface defects and fluorescent features of carbon quantum dots. **Figure 4D** shows rectangle, triangle, and circle shapes for pristine polystyrene, polyethylene/carbon quantum dot, and polyethylene/carbon quantum dot/silica hybrids, respectively, studied simultaneously under daylight and UV excitation (~360 nm). Thus, fluorescence can be seen for the nanocomposite sample shapes under UV excitation, relative to unfilled matrix shapes.

Additionally, among polyethylene derivative thermoplastics, poly(ethylene glycol) nanocomposites reinforced with graphene quantum dot have been designed and reported [50,51]. Kim et al. [52] stated the nanomaterials based on poly(ethylene glycol) filled with fluorescent graphene quantum dot, obtained through a solution processing technique (water solvent). The poly(ethylene glycol)/graphene quantum dot nanocomposite had a higher fluorescence quantum yield of ~5% with excitation at a 320 nm wavelength, compared with pristine graphene quantum dot. In addition, low toxicity and chemical stability features of the nanomaterials have been observed. Consequently, the as-designed poly(ethylene glycol)/graphene quantum dot nanocomposites were suggested to be useful for light-emitting diode applications. The resulting device depicted luminance > 800 cd m^{-2}. Similarly, other articles can be seen in the literature regarding the synthesis and fluorescence features of poly(ethylene glycol)/graphene quantum dot nanocomposites [53,54].

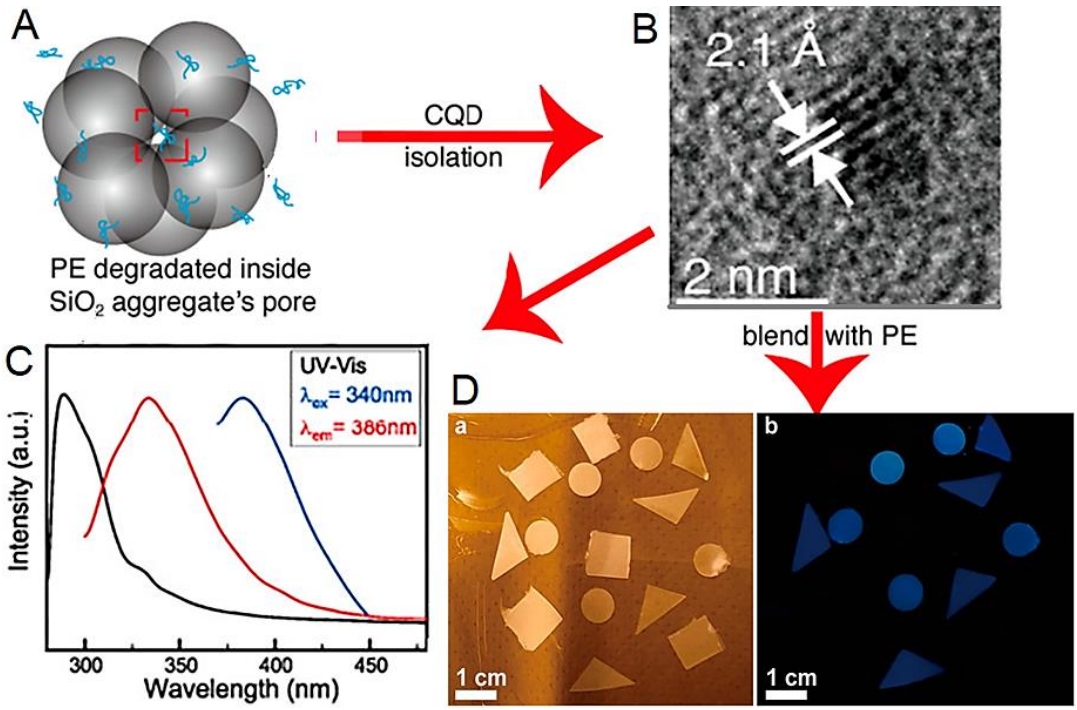

Figure 4. (A) Polyethylene in cavities/pores of silica nanoparticle aggregates; **(B)** high resolution transmission electron microscopy of polyethylene/carbon quantum dot/silica; **(C)** fluorescent emission spectra polyethylene/carbon quantum dot/silica hybrid at varying wavelengths; **(D)** photos of pure polyethylene (rectangles), polyethylene/carbon quantum dot (triangles), and polyethylene/carbon quantum dot/silica (circles) films in: **(a)** Daylight and **(b)** upon excitation with ~360 nm light [49]. Reproduced with permission from ACS.

Poly(vinylidene fluoride) appeared as a widely studied unique high performance thermoplastic, especially for the formation of nanocomposites [55]. Its important properties can be listed as optical, electronic, piezoelectric, ferroelectric, and dielectric characteristics, especially useful for energy/electronic device applications [56,57]. Out of carbon nanofillers, graphene-based poly(vinylidene fluoride) nanocomposites have been explored for technical features and performance [58]. Like graphene, its quantum dot nanostructures have been amalgamated with poly(vinylidene fluoride) to form high-performance nanocomposites [59]. Adding graphene quantum dots in poly(vinylidene fluoride) has been found to affect the piezoelectric features of the nanomaterials, e.g., β polymorph formation in the matrix [60]. Cho et al. [61] formed poly(vinylidene fluoride)/amino-modified graphene quantum dot nanocomposites. Including modified quantum dots developed hydrogen bonding and resulted in $\alpha \rightarrow \beta$ form changes of poly(vinylidene fluoride) matrix. Moreover, these nanomaterials had a high dielectric constant of ~61. Recently, Tay et al. [62] formed silver-modified graphene oxide quantum dots for a poly(vinylidene fluoride) matrix by the solution method. The poly(vinylidene fluoride)/silver-modified graphene oxide quantum dot nanomaterials had antimicrobial features against the E. coli bacterial strain. Zhang et al. [63] prepared important nanofiber designs of graphene oxide quantum dot using a poly(vinylidene fluoride) derivative, i.e., poly(vinylidene fluoride)(tetrabutyl titanate)/poly(vinyl pyrrolidone) (tetrabutyl titanate) by electrospinning method. For this purpose, graphene oxide quantum dots were synthesized using the hydrothermal method. In this way, pristine poly(vinylidene fluoride)(tetrabutyl titanate)/poly(vinyl

pyrrolidone)(tetrabutyl titanate) as well as poly(vinylidene fluoride)(tetrabutyl titanate)/poly(vinyl pyrrolidone)(tetrabutyl titanate)/graphene quantum dots nanocomposite nanofibers have been prepared. **Figure 5A** shows luminescent behavior of pristine nanodots at wavelength of 365 nm. **Figure 5B, C** show transmission electron microscopy micrographs of graphene quantum dots with uniform discrete nanoparticle dispersion. Accordingly, **Figure 5D** presents a size distribution spectrum of pristine nanodots around 1.8–3.0 nm. **Figure 5E, F** present neat polymer and graphene quantum dot-filled nanocomposite nanofibers, respectively. Relative to the smooth surface morphology of unfilled poly(vinylidene fluoride)(tetrabutyl titanate)/poly(vinyl pyrrolidone)(tetrabutyl titanate) nanofibers, the quantum dot-filled nanocomposite nanofibers had visible deposition of graphene quantum dots on the surface. These nanocomposite nanofibers were studied and found effective for photocatalytic degradation features regarding Rhodamine B.

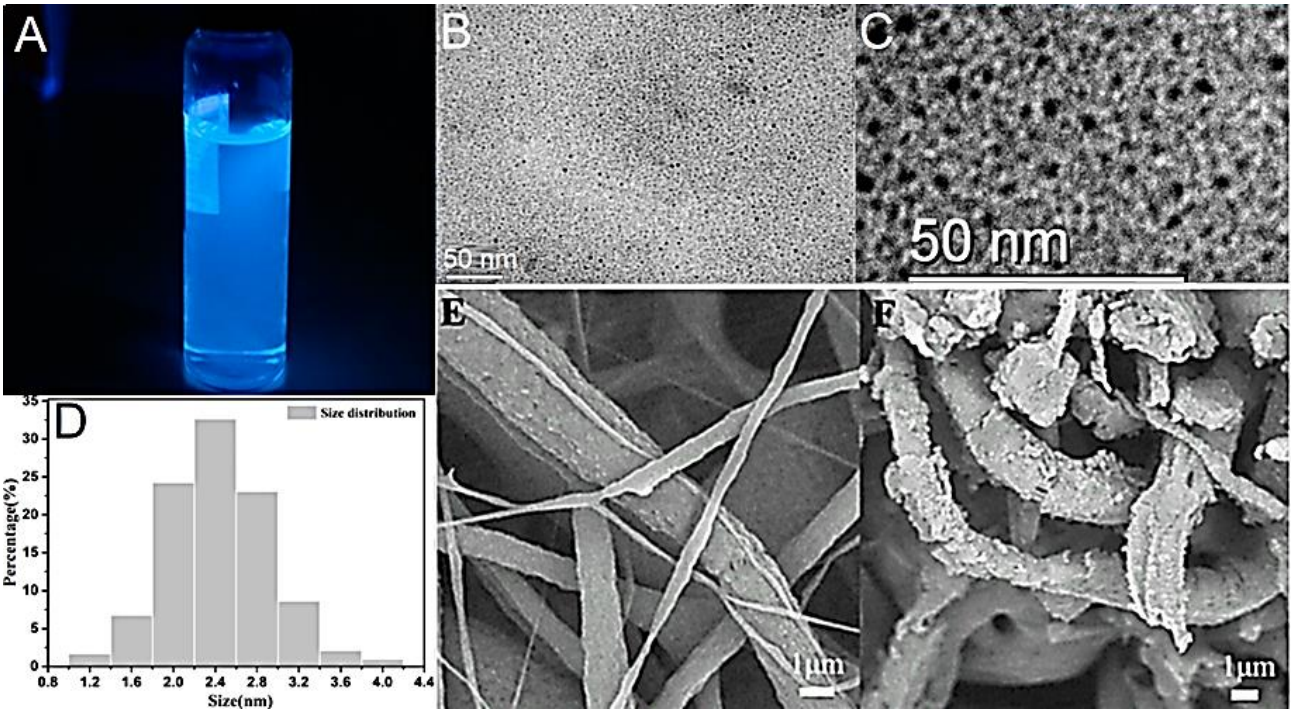

Figure 5. (A) Photograph of graphene quantum dot under 365 nm ultraviolet light; **(B,C)** transmission electron microscopy (TEM) images of quantum dot at low and high magnifications; **(D)** size distribution chart of graphene quantum dot; scanning electron microscopy of **(E)** poly(vinylidene fluoride)(tetrabutyl titanate)/poly(vinyl pyrrolidone)(tetrabutyl titanate) nanofiber; and **(F)** poly(vinylidene fluoride)(tetrabutyl titanate)/poly(vinyl pyrrolidone)(tetrabutyl titanate)/graphene quantum dot nanofiber [63]. Reproduced with permission from MDPI (open access).

Poly(vinyl alcohol) is also a common name among thermoplastics for nanocomposite formation [64,65]. It has advantageous features of water solubility, facile processing, and environmental friendliness. Owing to its abundant use as matrix material, inorganic and carbon nanofillers have been used to form the corresponding [66]. Graphene is a commonly reinforced nanocarbon in a poly(vinyl alcohol) matrix. Likewise, graphene quantum dot has been investigated for poly(vinyl alcohol) nanocomposites [67,68]. Adding quantum dots in a poly(vinyl alcohol) matrix was not

observed to repress the solubility, dispersion, luminescent properties, and quantum yield of the nanodots [69–72]. Fauzi et al. [73] developed poly(vinyl alcohol)/graphene quantum dot nanomaterial by the spin-coating method. The as-obtained nanocomposite was applied as a sensor for carbaryl detection, having a limit in the range of 0.001–0.007 ppb. Ogi and workers [74] filled graphene quantum dots in a poly(vinyl alcohol) matrix via a hydrothermal technique. These nanocomposites had remarkable photoluminescence intensity and a high quantum yield of 44%. Elumalai et al. [75] reported on solution-formed poly(vinyl alcohol)/graphene quantum dot nanocomposites. **Figure 6A** depicts a facile route for the formation of these nanomaterials having intense blue color illumination in UV light. According to the differential thermal analysis data of unfilled and filled matrix given in **Figure 6B**, the neat poly(vinyl alcohol) sample had crystalline melting peaks around 220 °C and moisture peaks at 120 °C. On the other hand, the nanocomposite depicted altogether different thermograms with a glass transition temperature of about 59 °C and exothermic peaks due to matrix-nanofiller interfacial interactions. Hence, poly(vinyl alcohol)/graphene quantum dot nanocomposites have been mostly explored for fluorescence emission properties, as per available reports so far.

5. Applications of thermoplastic/graphene quantum dot nanocomposites

As discussed in preceding sections of this review, graphene quantum dots have several structure-property advantages of extremely tiny sizes, zero dimensions, and high surface area, as well as quantum confinement effects, relative to pristine graphene nanosheets, which are in turn useful in varying technological devices and biomedical appliances [76–78]. An important application of graphene quantum dots and derived nanocomposites has been observed for electromagnetic interference (EMI) shielding [79]. For radiation shielding, literature reports on countless designs of carbonaceous nanocomposites using carbon nanotubes, graphene, carbon black, and other nanocarbons [80–82]. Generally, nano-sizes, compatibility with polymers, interface formation, and electron conduction or percolation behavior of nanocarbons were found useful for shielding the effects of ecologically hazardous electromagnetic radiations continuously emitted by functional electronics and allied equipment. The EMI defense was mainly accredited to the conducting behavior of nanocarbon. Amongst nanocarbon nanofillers, graphene is an exclusive two-dimensional nanosheet nanocarbon graphene, which has been commonly used for EMI defense applications [83–85]. Graphene quantum dot-based nanomaterials have lightweight, fine nanoparticle dispersion, electrical conductivity, dielectric, and electromagnetic features for high-end EMI shielding applications [86]. All these EMI shielding-related features of the nanocomposites rely upon graphene quantum dot dispersion as well as matrix-nanofiller associations.

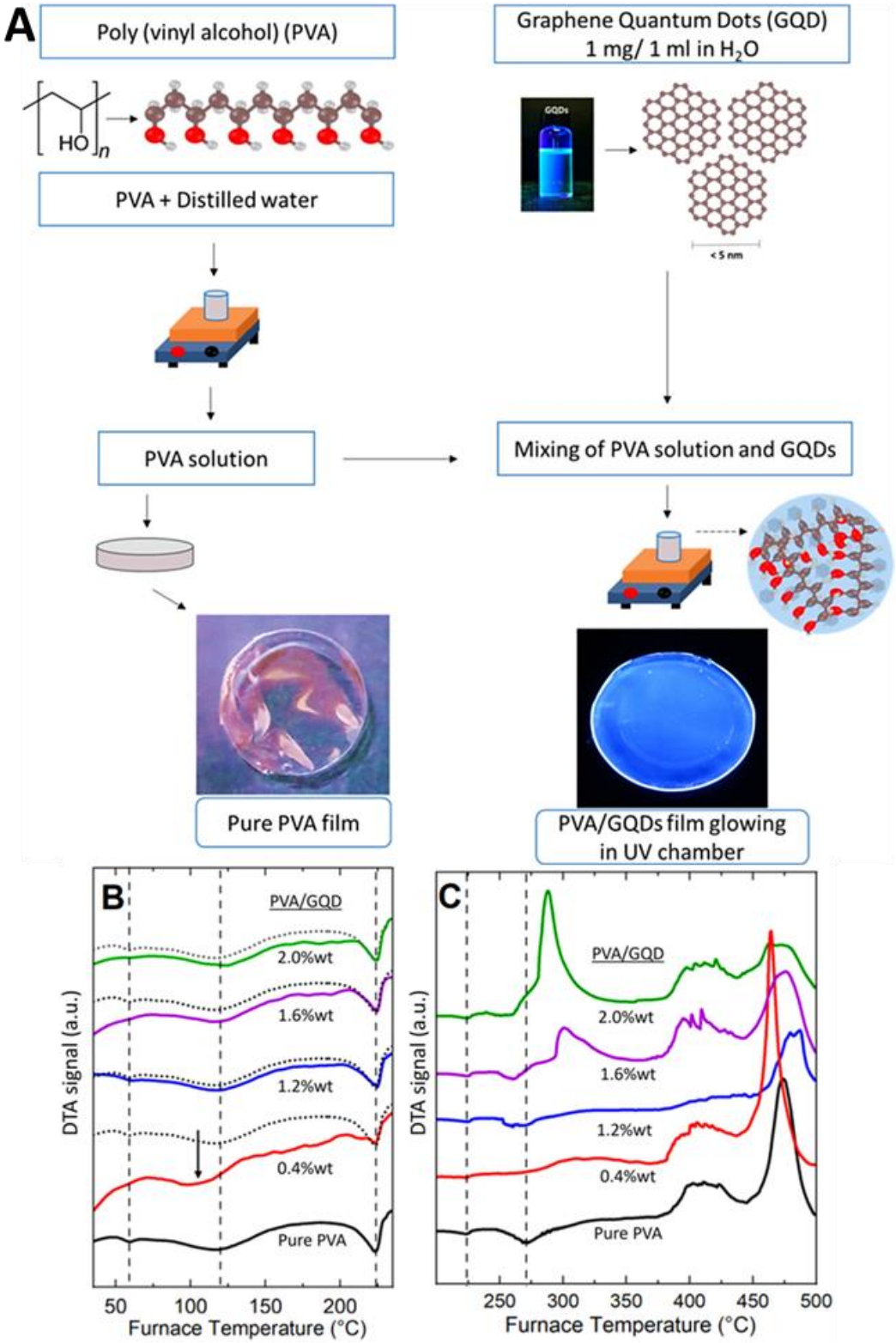

Figure 6. (A) Fabrication of pristine PVA and PVA/GQD nanocomposite films; **(B,C)** differential thermal analysis data of neat PVA and PVA/GQD nanocomposite films, respectively [75]. PVA = poly(vinyl alcohol); PVA/GQD = poly(vinyl alcohol)/graphene quantum dots nanocomposite. Reproduced with permission from MDPI (open access).

Lakshmi et al. [87], for instance, explored electromagnetic shielding effectiveness of the poly(vinylidene fluoride)/graphene decorated graphene quantum dots and poly(vinylidene fluoride)/graphene decorated graphene quantum dots/silver

nanoparticles nanocomposites. According to the results for the nanocomposites having 2 wt.% nanofiller loadings, the poly(vinylidene fluoride)/graphene decorated graphene quantum dots/silver nanoparticles (with contents) had a total shielding effectiveness of 43 dB, i.e., ten times higher than that of the poly(vinylidene fluoride)/graphene decorated graphene quantum dots (31 dB) as shown in **Figure 7a**. Upsurges in the total shielding effectiveness of the graphene-decorated graphene quantum dots/silver nanoparticles-based nanocomposites seemed to be due to their mutual nanostructural effects between the quantum dots and the metal nanoparticles, therefore resulting in their uniform dispersions and an interconnecting network formation for a facilitated electron flow through the epoxy matrix. Accordingly, the graphene-decorated graphene quantum dots/silver nanoparticles were capable of shielding around 99.9% of electromagnetic radiations, relative to the neat epoxy matrix. Additionally, as shown in **Figure 7b**, both absorption and reflection of electromagnetic waves for the graphene-decorated graphene quantum dots/silver nanoparticles-filled nanocomposites were observed to be higher, as compared to the non-modified graphene-decorated graphene quantum dots nanocomposites. Herein, superior reflection/absorption from the graphene-decorated graphene quantum dots/silver nanoparticles-based nanocomposites was attributed to their better dispersion in the poly(vinylidene fluoride) matrix, therefore, facilitating more surface charges and higher conductivity of their nanocomposites, relative to the non-modified quantum dots-based system. Moreover, electromagnetic wave attenuations of the poly(vinylidene fluoride)/graphene decorated graphene quantum dots (left image in **Figure 7c**) and poly(vinylidene fluoride)/graphene decorated graphene quantum dots/silver nanoparticles nanocomposites (right-hand image in **Figure 7c**) were investigated. It was suggested that graphene-decorated graphene quantum dots/silver nanoparticles developed a denser interconnecting network in the epoxy matrix, relative to that of the non-modified graphene-decorated graphene quantum dots, therefore resulting in superior tunneling phenomena and wave attenuation characteristics of their nanocomposites.

Since pristine polymeric matrices are usually nonconductive and do not absorb radiation. Including semiconducting nanoparticles, like graphene quantum dots, in polymers has been reported to enhance the electrical conductivity and electromagnetic absorption properties of polymeric nanocomposites. The electrically conducting graphene quantum dots usually have the capability to dissipate electrical charges, thereby reducing electromagnetic field buildup. In addition, EMI shielding mechanism of polymer/graphene quantum dot nanocomposites seemed to be relying upon network formation, alignment, and selective localization of quantum dots nanoparticles at matrix-nanofiller interfaces, so contributing to EMI shielding competence. In this way, semiconducting quantum dots exhibited fine electromagnetic reflection/absorption characteristics to effectively enhance radiation shielding of nanocomposites.

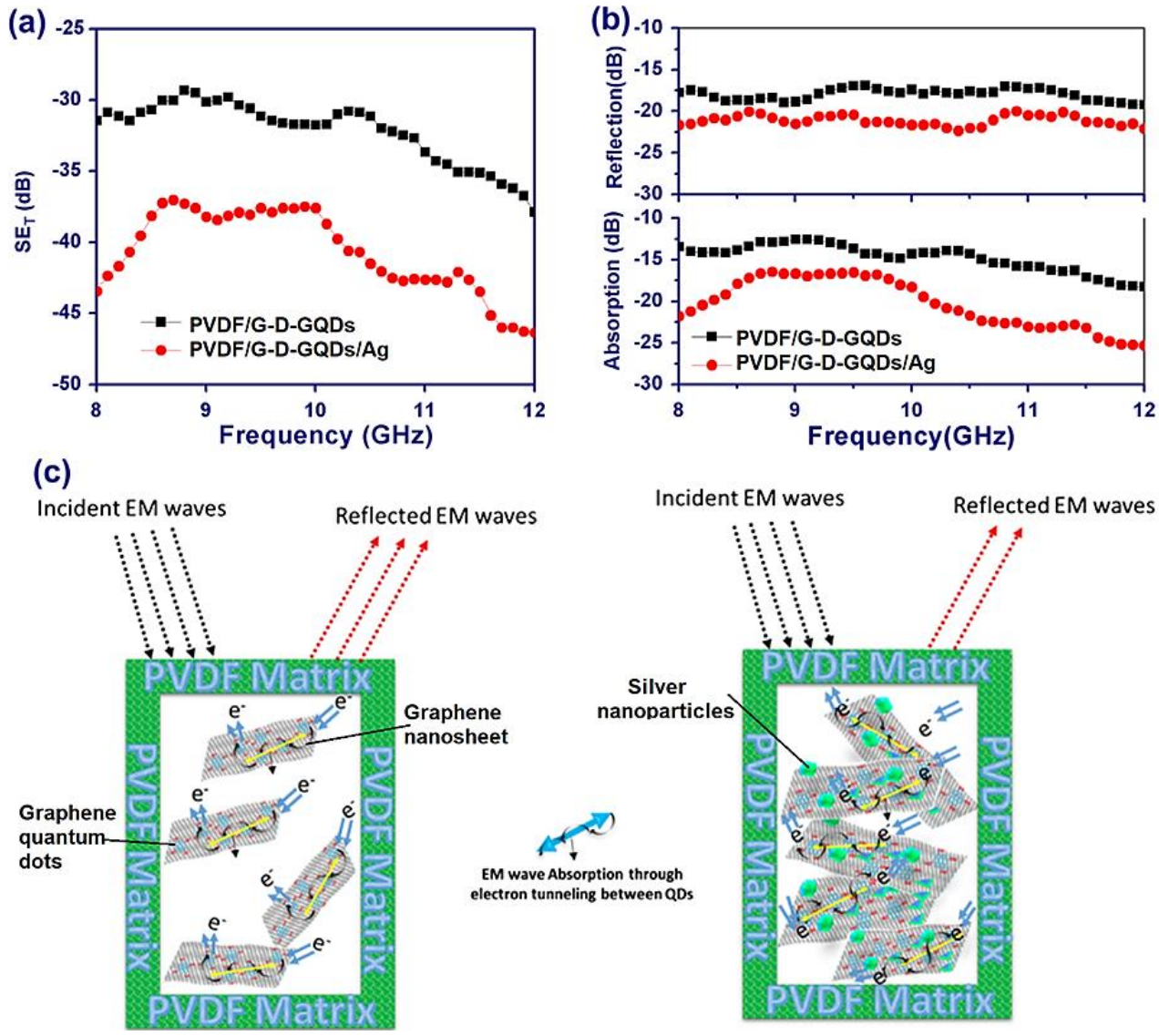

Figure 7. (a) Total shielding effectiveness of poly(vinyledene fluoride)/graphene decorated graphene quantum dots, PVDF/G-D-GQDs, and poly(vinyledene fluoride)/graphene decorated graphene quantum dots/silver nanoparticles, PVDF/GD-GQDsAg, nanocomposites in the X-band range and **(b)** reflection and absorption of the PVDF/G-D-GQDs and PVDF/GD-GQDsAg nanocomposites in the X-band range; **(c)** schematic of attenuation of electromagnetic (EM) wave of PVDF/G-D-GQDs nanocomposites (left) and PVDF/GD-GQDs/Ag nanocomposites (right) [87]. Reproduced with permission from Taylor and Francis.

Due to the charge-storing properties of graphene quantum dots, interesting applications can be seen regarding the memory and charge-trapping devices [88,89]. Nevertheless, such applications demand low-cost and facile material fabrication on a commercial scale without environmentally harmful effects. In this concern, Kou et al. [90] hydrothermally formed poly(methyl methacrylate) filled with graphene quantum dots. A transmission electron microscopy study was used to confirm the consistent dispersion of graphene quantum dots having 7 nm in size throughout the matrix. **Figure 8A** shows the final design of a flexible memory device of poly(ethylene terephthalate)/indium-tin-oxide/poly(methyl methacrylate)/graphene quantum dot (PET/ITO/PMMA:GQDs/Al). **Figure 8B** presents comparative photoluminescence

spectra of pristine poly(methyl methacrylate), graphene nanosheet, graphene quantum dot, and PMMA/GQD nanocomposite.

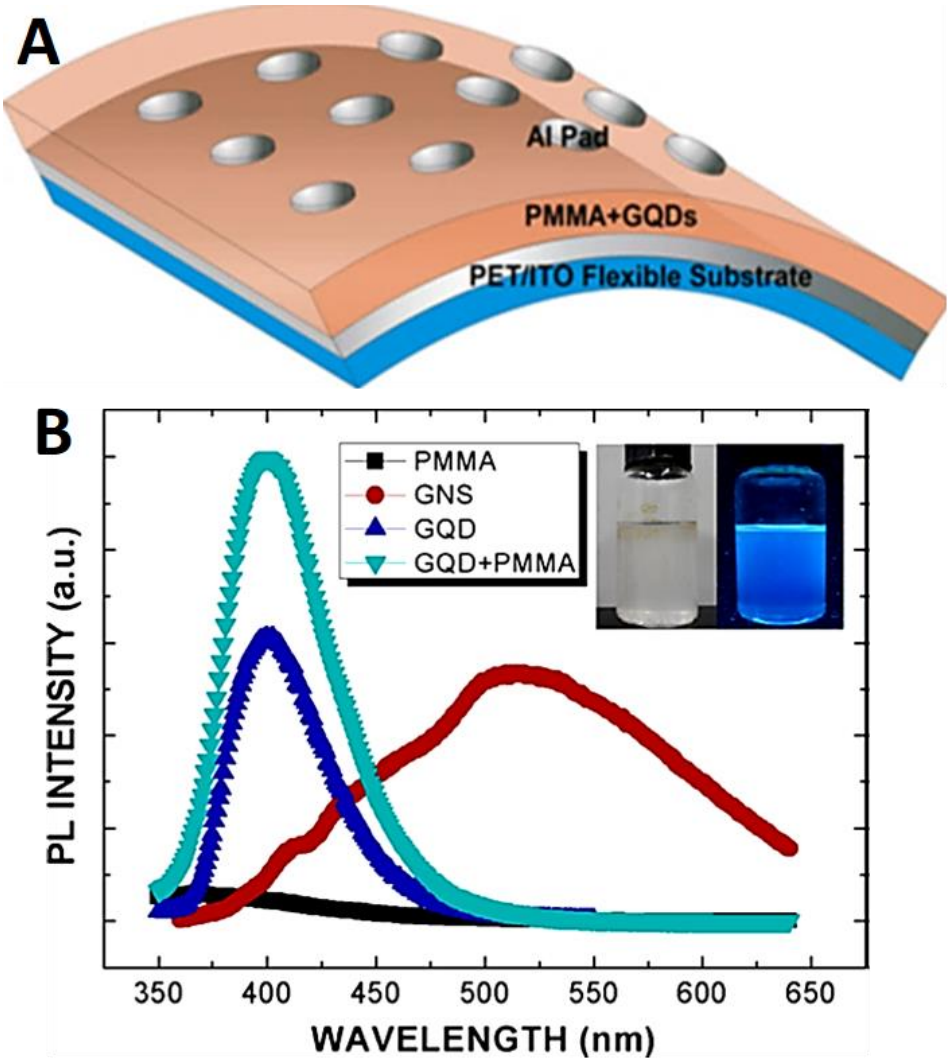

Figure 8. (A) A graphic of a flexible poly(ethylene terephthalate)/indium-tin-oxide/poly(methyl methacrylate)/graphene quantum dot (PET/ITO/PMMA: GQDs/Al) memory device; **(B)** photoluminescence spectra of poly(methyl methacrylate), PMMA, graphene nanosheet (GNS), GQD, and PMMA/GQD nanocomposite films excited at 260 nm, where Inset: Photograph of GQD chlorobenzene solution taken under visible light and 365 nm UV light, from left to right, respectively [90]. GQD = graphene quantum dots; PMMA/GQD = poly (methyl methacrylate) and graphene quantum dots-based nanocomposites; GNS = graphene nanosheet. Reproduced with permission from Elsevier.

As per results, the nanocomposite revealed a photoluminescence peak of visibly higher intensity at a wavelength of 405 nm, relative to neat graphene quantum dots. The change in fluorescence spectral intensity indicated homogeneous dispersion and compatibility of nanodots in the nanocomposite matrix, which was found useful for manufacturing the desired memory device. On the other hand, graphene showed a broad photoluminescent peak at 510 nm owing to the broad size distribution of

nanosheets. However, few researches have been seen on the use of thermoplastic polymer/graphene quantum dots for these systems, and future studies may unveil important results for designing advanced high-tech memory devices.

Regarding optoelectronic applications of thermoplastic polymer/graphene quantum dot nanomaterials, few important studies have been noticed so far on systems having fine electronic, optical, and luminescence features [91]. In this concern, Liu et al. [92] designed starch and nitrogen-doped reduced graphene oxide quantum dot-based nanocomposites having conductivity and fluorescence properties. Including 10 wt.% nitrogen-doped reduced graphene oxide quantum dot led to low resistivity of ~0.08 Ω·m, reasonably high light transmittance of 80%, and fluorescence intensity of up to 9000 CPS. These multifunctional, ecologically friendly nanocomposites were suggested for wearable optoelectronic devices. Chen et al. [93] proposed melt-processed thermoplastic starch and graphene quantum dots-based fluorescent nanocomposites. Adding 10 wt.% nanofiller depicted low resistivity and notable photoluminescence intensity properties of the nanocomposites. According to the results, these environmentally friendly nanocomposites exhibited high-end potential application as optoelectronic packaging materials.

Photocatalytic performance of thermoplastic polymer/graphene quantum dot nanocomposites has been found valuable for environmental remediation [94]. In this concern, Mafukidze et al. [95] formed solution-processed polystyrene and zinc phthalocyanine functional graphene quantum dot-derived nanocomposites and photocatalytic activity for the remediation of 4-chlorophenol from water. These nanocomposites followed second-order kinetics for photocatalytic oxidation of 4-chlorophenol. Apostolaki et al. [96] formed polystyrene and titania-modified graphene quantum dot-based photonic crystals. The ensuing nanocomposite exhibited blue luminescence at 350 nm due to n-π^* transitions. The polystyrene/titania-graphene quantum dot photonic crystals were found useful for photocatalytic degradation of salicylic acid.

Similarly, few studies on thermoplastic polymer/graphene quantum dots mentioned potential towards chemical sensing applications [97–99]. Notably, Majid Masteri-Farahani et al. [100] developed poly(methyl methacrylate)/graphene quantum dots-based fluorescent nanosensors using the molecular imprinting method for methamphetamine detection. These nanocomposites had a fine detection limit of ~1.7 μg/L for methamphetamine.

In the biomedical sector, carbon nanodots, especially graphene quantum dots, attained significance owing to their biocompatibility and non-toxic nature [101,102]. Furthermore, these nanoparticles have remarkable fluorescent and surface/edge-related quantum effects for biomedical uses [103,104]. Besides, graphene quantum dots have been reported for their biologically inert nature, leading to technical utilization in fluorescent bioimaging probes [105]. Zhu et al. [106] formed fluorescent graphene quantum dot-based bioimaging probes using the solvothermal method. The obtained graphene quantum dot had a high quantum yield of > 11%. Nurunnabi et al. [107] reported on graphene quantum dots coated with polydopamine obtained by oxidation/exfoliation techniques. The as-formed polydopamine/graphene quantum dot nanocomposites depicted notable properties, like in vivo stability, non-toxicity, photoluminescence, etc. Such nanocomposite architectures were discovered to be

advantageous for drug delivery as well as optical imaging applications [108]. As per literature, **Figure 9** illustrates the use of bioactive graphene quantum dots for malignancy diagnosis via the bioimaging method [109]. For designing fluorescent bioimaging probes, photoluminescence properties of graphene quantum dots were found valuable [110,111]. Similarly, Sheng et al. [112] fabricated polyvinylpyrrolidone/nitrogen-doped graphene quantum dot hybrids via hydrothermal means. The ensuing nanocomposites have high quantum efficiency (> 64%), which was found beneficial for employment in fluorescence probes for chromium (VI) detection. Thus, bioactive graphene quantum dots have been effectively explored in manufacturing fluorescent bioimaging probes [113]. Nonetheless, future attempts in this direction may lead to the formation of reproducible commercial designs of fluorescent graphene quantum dots for bioimaging applications.

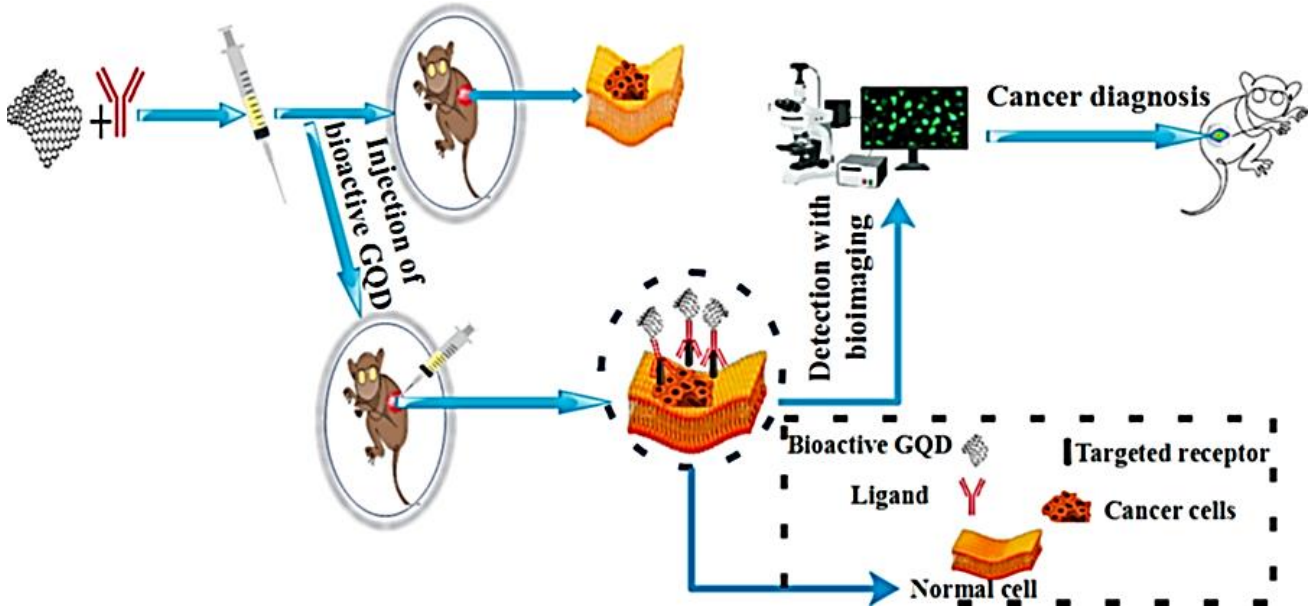

Figure 9. Use of bioactive graphene quantum dots (GQD) for malignancy diagnosis via bioimaging [99]. Reproduced with permission from MDPI.

Furthermore, scientific attempts have been observed on the antimicrobial activity of graphene quantum dots-based nanomaterials [114]. In this concern, Rajendiran et al. [115] functionalized graphene quantum dots with poly(ethylene glycol), polyethyleneimine, and poly-l-lysine polymers. These non-toxic polymer/graphene quantum dot nanocomposites showed antibacterial and antifungal activities. Liu et al. [116] formed polyethylenimine/graphene quantum dot/zinc oxide nanocomposites using sol-gel and solution methods. These nanomaterials showed fine antimicrobial activity towards the E. coli bacterial strain. However, limited literature has been seen so far in this important biomedical area of graphene quantum dots.

6. Conclusions and views

In short, thermoplastic polymers have been decisively reviewed with graphene quantum dot nanofillers, considering different matrices, fabrication techniques, physical features, and applied prospects. Amalgamation of thermoplastic polymer

with graphene-structured quantum dots revealed interesting structural, morphological, and wide-ranging physical attributes. The upsurges in these properties of thermoplastic polymer/graphene quantum dot can be credited to the quality of these polymers to consistently disperse the nanodot for robust interfacial interactions. Here, functional graphene quantum dots seem to effectively interact with thermoplastics for the formation of compatible nanocomposite nanostructures. Among prominent application zones, thermoplastic polymer/graphene quantum dot nanocomposites revealed success for radiation shielding, memory devices, optoelectronics, photocatalysts, sensing, antibacterial, and bioimaging. For future high-performance designs and applications, challenges regarding polymer and nanodot functionalities, synthesis, mutual compatibility, and large-scale production need to be technically resolved.

Broadly speaking, graphene quantum dots have been found to develop fine interfacial miscibility with thermoplastic polymers, thereby leading to unique microstructures, electron/charge transference, fluorescence, heat stability, nonflammability, biomolecular sensing, catalytic, and antibacterial properties. All these key advantageous characteristics and applied aptitudes of polymer/graphene quantum dot nanomaterials depend upon seamless nanoparticle dispersion and network formation in matrices, which can be attained via appropriate processing techniques to develop strong matrix-nanofiller interactions.

For synthesizing thermoplastic polymer/graphene quantum dot nanocomposites, most studies focused on facile solution processing and Pickering emulsion polymerization techniques. Few reports have also been observed on melt mixing or compression molding methods for the formation of graphene quantum dots filled nanomaterials. Beside conventional solution and melt techniques, sophisticated approaches, like spin coating and hydrothermal/solvothetmal have also been used to integrate quantum dots in polymeric nanocomposites. Here, selection of an appropriate technique seems to directly influence the physical features of the as-prepared polymer/graphene quantum dot nanocomposite. All these techniques used so far have certain advantages and disadvantages, which must be considered before nanocomposite synthesis. Out of these, solution processing has been considered a low-cost and environmentally friendly method for the formation of polymer/graphene quantum dot nanocomposites. This technique has been used to attain fine nanofiller dispersion in the matrices. The melt method has also been reported for fine nanofiller dispersion in polymeric matrices; however, it has its own limitations of using extreme temperature/shear conditions. Relative to solution/melt methods, the spin coating method has been found more effective for uniform quantum dot dispersion in polymers. Consequently, homogeneous nanofiller dispersion may ensure strong interfacial bonding with polymers, thereby leading to desirable enhancements in physical properties (microstructure, structural integrity, heat stability, electrical percolation, etc.) of the nanocomposites. Along with the choice of an appropriate technique, control of processing parameters seems indispensable for large-scale processing to attain high-performance polymer/graphene quantum dot nanocomposites.

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