ADVANCES IN MATERIALS SCIENCE, Vol. 25, No. 1 (83), March 2025

DOI: 10.2478/adms-2025-0001

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GRAPHENE QUANTUM DOTS IN CONJUGATED AND RUBBERY MATRICES - STIPULATIONS AND PHYSICAL/TECHNICAL ATTRIBUTES

ABSTRACT

This leading-edge overview delivers an all-inclusive knowledge on fundamentals, state-of-the-art, and technicalities of two important polymer categories filled with graphene quantum dots, namely conjugated polymer/graphene quantum dots and rubber/graphene quantum dots nanocomposites. According to the literature up till now, facile and efficient fabrication methods, like in situ polymerization, solution mixing, melt blending, etc. have been reported for these graphene quantum dots derived hybrids. The ensuing graphene quantum dots based nanocomposites were inspected for microstructural, electrical conductivity, charge transportation, thermal/mechanical resistance, fluorescence properties, and allied valuable physical features. Incidentally, we notice promising applications of inimitable categories of conjugated polymer/graphene quantum dots and rubber/graphene quantum dots hybrids for gas/molecular/piezoelectric sensors, supercapacitors, and biomedical areas. Nevertheless, due to limited reports on applied sides of graphene quantum dots filled conjugated/rubbery matrices, future research attempts seem indispensable to resolve challenges of optimized/controlled processing and also to unveil structure-property-performance links and synergistic mechanisms for developing next generation industrial level conjugated polymer/graphene quantum dots and rubber/graphene quantum dots nanocomposites.

Keywords: graphene quantum dots; conjugated polymers; rubbers; nanocomposites; sensors; supercapacitors

INTRODUCTION

Among significant categories of polymeric materials, conjugated polymers and rubbers have been frequently surveyed in the literature as potential matrices for reinforcing carbonaceous nanoparticles [1, 2]. Out of vast varieties of carbon nanofillers (graphene, fullerene, carbon based quantum dots, etc.), zero dimensional graphene quantum dots have been studied as valuable nanoreinforcements due to distinct structural and physical property features [3]. In view of that, graphene quantum dots have been investigated as nanofillers for conjugated as well as rubbery polymeric matrices [4]. Subsequently, including graphene quantum dots in semiconductive polymers and elastomers revealed technically worthwhile microstructures, optoelectronic, heat/mechanical resilience, and allied physical features [5, 6]. In due course, graphene quantum dots loaded conducting/rubbery nanomaterials have been scrutinized for applications in the fields of advanced energy/electronic devices and biomedical arenas [7]. For these nanocomposites, pristine as well as modified graphene

Received: 9.11.2024; Received in revised form: 10.12.2024 ; Accepted: 28.12.2024

quantum dots, obtained by feasible synthesis routes, seem valuable to upsurge the structure-property-performance attributes for the mentioned practical uses [8-10].

Considering the scientific importance of the conjugated polymers/graphene quantum dots and rubbers/graphene quantum dots, this review has been planned to state the fundamentals as well as current crucial structure-property forecasts of these nanocomposites. Consequently, under the category of conjugated polymers, nanoreinforcement effects of graphene quantum dots for polyaniline, polypyrrole, and polythiophene/derivative matrices have been surveyed here, accordingly to the available literature till now. Regarding rubbery matrices, technical influences of graphene quantum dots loadings on synthetic rubbers (nitrile/butyl rubber or silicones) and natural rubber have been discussed in this article. Particularly, conjugated polymers/graphene quantum dots and rubbers/graphene quantum dots nanocomposites depicted distinct morphological, optical, fluorescent, thermal, and mechanical characters leading to their valuable uses in the fields of sensing and supercapacitive devices and biomedical sectors. However, current state of graphene quantum dots filled conjugated polymers and rubbery polymers demands further focused field efforts for new well defined material designs, optimized synthesis, and overcoming the structural, property, and technical challenges for their future industrial level utilizations.

GRAPHENE QUANTUM DOTS

Research up till now on carbonaceous nanostructures has resulted in the discovery of countless unique carbon nanoforms, like graphene, carbon nanofiber, carbon nanotube, nanodiamond, carbon black, etc., having different dimensionalities and nanosizes [11, 12]. Under the category of extremely small zero dimensional quantum dot nanoparticles, carbon based quantum dots, such as graphene quantum dots, polymer dots, and carbon dots, have been reported [13, 14]. These quantum dots may have nanosizes, of < 10 nm, so leading to unique fluorescence characters [15]. Among carbon based quantum dots, zero dimensional graphene quantum dots form a distinct category of nanoparticles having valuable quantum confinement and edge effects [16]. Graphene quantum dots have been designed using a number of bottom up and top down strategies, as shown in Fig. 1 [17].

The pristine graphene owns two dimensional sp^2 hybridized nanostructure, exceptionally high surface area (2630 m^2g^{-1}), zero band gap structure, and structural/physical attributes (optical, electronic, conducting, thermal, magnetic, etc.) [18]. Contrarily, graphene quantum dots have zero dimensions and non zero band gap structure along with photoluminescence emission properties [19, 20]. These tiny graphene derived nanoentities have tremendously small crystalline lattice parameters, down to 0.24 nm [21]. Other valuable characters of graphene quantum dots include their inherent non-toxicity, photoelectric effect, optoelectronic features, electron/thermal conductivity, etc. [22]. Subsequently, graphene quantum dots have gained mammoth recent scientific interests for photovoltaics, sensing devices, light emitting diodes, microelectronics, and biomedical sectors, like bioimaging, tissue engineering, and drug delivery [23-25].

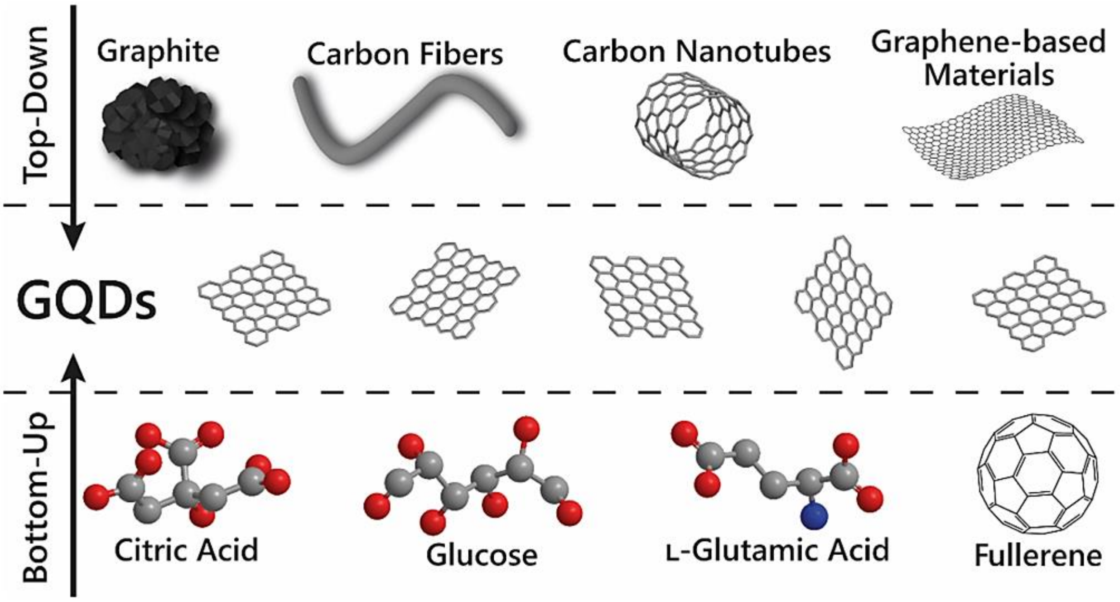

Fig. 1. Graphene quantum dot and precursors for synthesis [26]. Reproduced with permission from MDPI

CONJUGATED AND RUBBERY MATRICES

Conjugated or semiconducting polymers, mostly organic macromolecules, have intrinsically alternating double and single bonding in the main chains along with delocalized π electrons responsible for facilitated electron conductivity, and electrical and optical features [27]. Accordingly, owing to inherent backbone aromaticity, conjugated polymers are sometimes referred as the synthetic metals in the literature [28]. Consequently, conjugated or conductive matrices own facile processing tendencies for the formation of high tech materials/nanomaterials [29, 30]. Notable examples of conducting polymers include polycarbazoles, polyphenylene, polyaniline, polypyrrole, polythiophene, and countless derived forms of these polymers [31]. Owing to structure-property uniqueness of conducting polymers, advanced research efforts have focused their technical applications for nanocomposite formation, anticorrosion coatings, environmental materials, and biomedical systems, to mention as few [32-34].

Synthetic or natural rubbers mark another significant category of polymeric materials [35]. Naturally occurring rubber or natural rubber is usually extracted from latex of rubber plant using coagulation methods [36]. Natural rubber has structural formula of cis-1,4-poly(isoprene) [37]. Owing to intrinsic elastomeric backbones, processing, structural integrity, and applied possibilities of natural rubber have been improved using vulcanization technique [38]. This method is usually applied to induce three dimensional linking [39], strain triggered crystallinity [40], self-reinforcing features [41], and exceptionally high mechanical stretching [42]. Additionally, various synthetic rubbers have also been reported, e.g., butyl rubbers, nitrile rubbers, neoprene, silicones, and countless derived rubbery matrices [43, 44]. Distinguished applications of synthetic/natural rubbers have been documented ranging from elementary uses (automobile tires, automotive parts, household equipment, toys, cosmetics, etc.) to advanced industrial applications (civil/construction, defense, aerospace, radiation protection, and so on) [43, 45].

GRAPHENE QUANTUM DOTS NANOREINFORCEMENTS IN CONJUGATED MATRICES

Polyaniline/graphene quantum dots

Graphene reinforced polymeric nanocomposites have exposed momentous technological worth in wide ranging electronics, energy, and engineering applications [46]. In addition to pristine graphene, its inimitable derived forms, like graphene quantum dots, have further enhanced the technological implication of photovoltaics, supercapacitors, batteries, sensors, and biomedics, to name a few [47, 48]. For scientific utilizations of graphene quantum dots, conjugated polymeric matrices seem beneficial owing to intrinsic electron and charge transference, economical/facile processing and ecofriendliness [49, 50]. Amid conjugated polymers, polyaniline has been repeatedly searched for nanocomposite formation, however it had major drawback of poor structural strength, so restricting its technical uses [51, 52]. Luk et. al. [53] designed the polyaniline and graphene quantum dots based nanocomposites through chemical oxidation synthesis method. The ensuing polyaniline and graphene quantum dots nanocomposites showed enhanced electron/charge conductivity properties, relative to unfilled polyaniline [54]. Mondal et. al. [55] also synthesized polyaniline/graphene quantum dots nanocomposites by in situ chemical oxidation polymerization. They noticed synergetic effects between the polyaniline matrix and graphene quantum dots responsible for remarkably high specific capacitance of > 1000 Fg^{-1} for supercapacitor application. Punrat et. al. [56] prepared graphene quantum dots filled polyaniline nanomaterials using a screen printing technique. The resulting polyaniline/graphene quantum dot hybrids were studied for Cr^{6+} ion sensing in aqueous media, and superior ion recovery (of 106 %) was observed depicting their use for efficient sensors. Few literature searches have mentioned the formation and technical uses of polyaniline nanotube nanostructures [57, 58]. Breczko et. al. [59], for example, synthesized and examined sulfonated polyaniline nanotube and graphene quantum dots based hybrids. In this concern, Figs. 2 A-C portrays transmission electron microscopy images of neat graphene quantum dots, pristine polyaniline nanotubes, and ensuing polyaniline nanotube/graphene quantum dots nanocomposite. As per micrographs, neat graphene quantum dots had spherically symmetrical microstructures, of sizes ~ 2-5 nm, whereas quantum dots were seen dispersed on the polyaniline nanotube surfaces in the nanocomposite sample.

Fig. 2 D pictures a synthesis route for in situ oxidative polymerization of aniline monomers with *p*-toluene sulfonic acid to form the sulfonated polyaniline nanotubes and their later conversion to the polyaniline nanotube/graphene quantum dots nanocomposite. To understand the nature of interactions/orientations of modified polyaniline and graphene quantum dots in the hybrid system, Fig. 2 E exhibits an optimized supercell nanostructure. Accordingly, effective matrix-nanofiller interactions were proposed to be the hydrogen bonding, van der Waals links, as well as aromatic stackings in the nanocomposite [60]. Ultimately, finely interlinked sulfonated polyaniline nanotube and graphene quantum dots derived nanocomposites had superior electron conductivity and specific capacitance (245 F g^{-1}) properties for application as supercapacitor electrodes. Siddique et. al. [61] reported on polyaniline and nitrogen functional graphene quantum dots attained using hydrothermal and in situ techniques. Fig. 3 A shows high resolution transmission electron microscopy micrograph of the nitrogen functional graphene quantum dots depicting morphology and sizes of 1-7 nm, due to effectiveness of hydrothermal synthesis route. Fig. 3 B illustrates scanning electron microscopy micrograph of the polyaniline/nitrogen functional graphene quantum dots nanocomposite showing nanorods like assembly of polyaniline along with the dispersed quantum dots without any prominent aggregations.

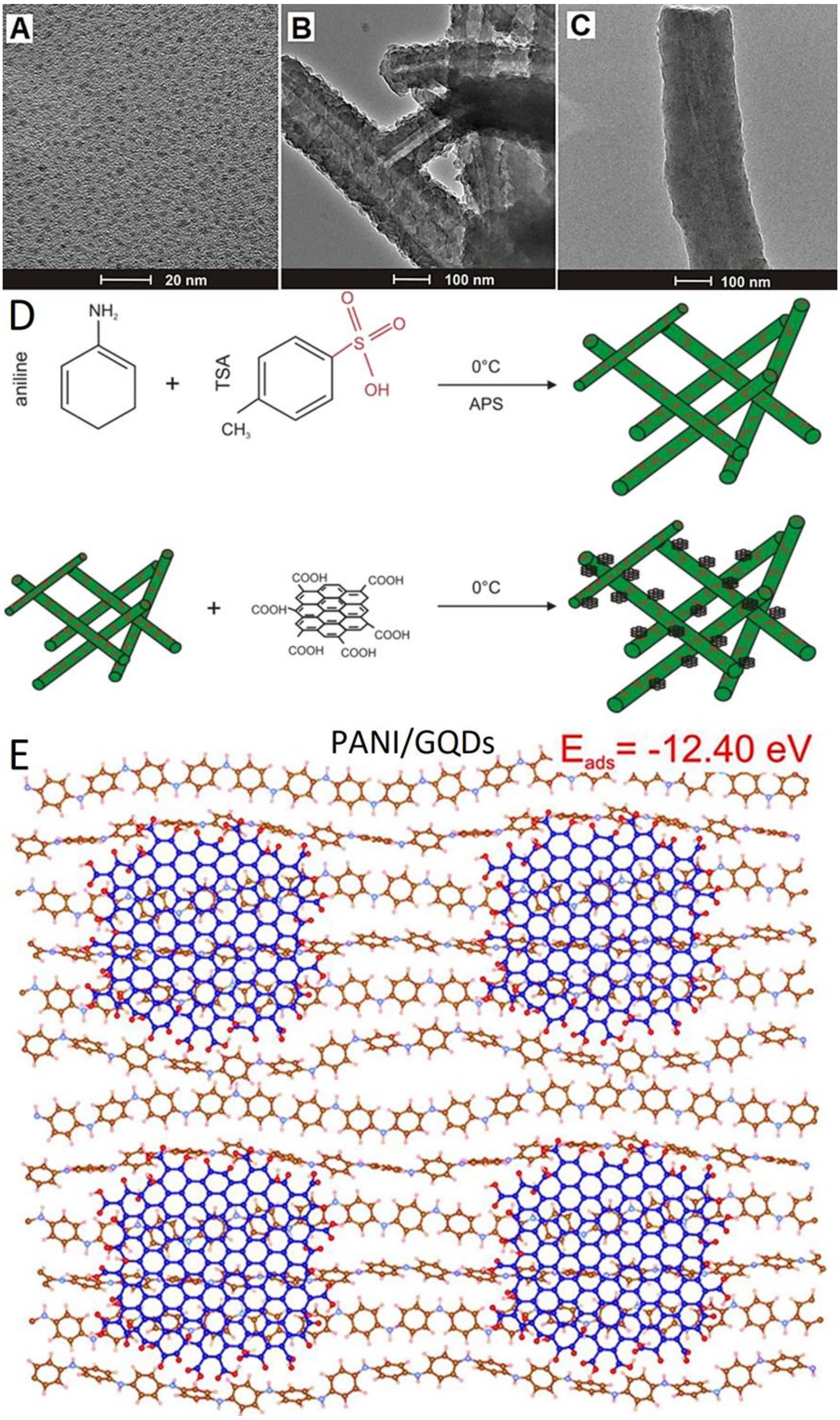

Fig. 2. Transmission electron microscopy images confirming the structure of (A) GQDs; (B) s-PANI-NTs; (C) s-PANI-NTs/GQDs; (D) synthesis of s-PANI-NTs and s-PANI-NTs/GQDs; (E) optimized structure of polyaniline/graphene quantum dots (PANI/GQDs) system [59]. GQDs = graphene quantum dots; s-PANI-NTs = sulfonated polyaniline nanotube; s-PANI-NTs/GQDs = sulfonated polyaniline nanotube/graphene quantum dots; s-PANI-NTs = sulfonated polyaniline nanotube; s-PANI-NTs/GQDs = sulfonated polyaniline nanotube/graphene quantum dots; APS = ammonium persulfate; TSA = p-toluene sulfonic acid. Reproduced with permission from Elsevier

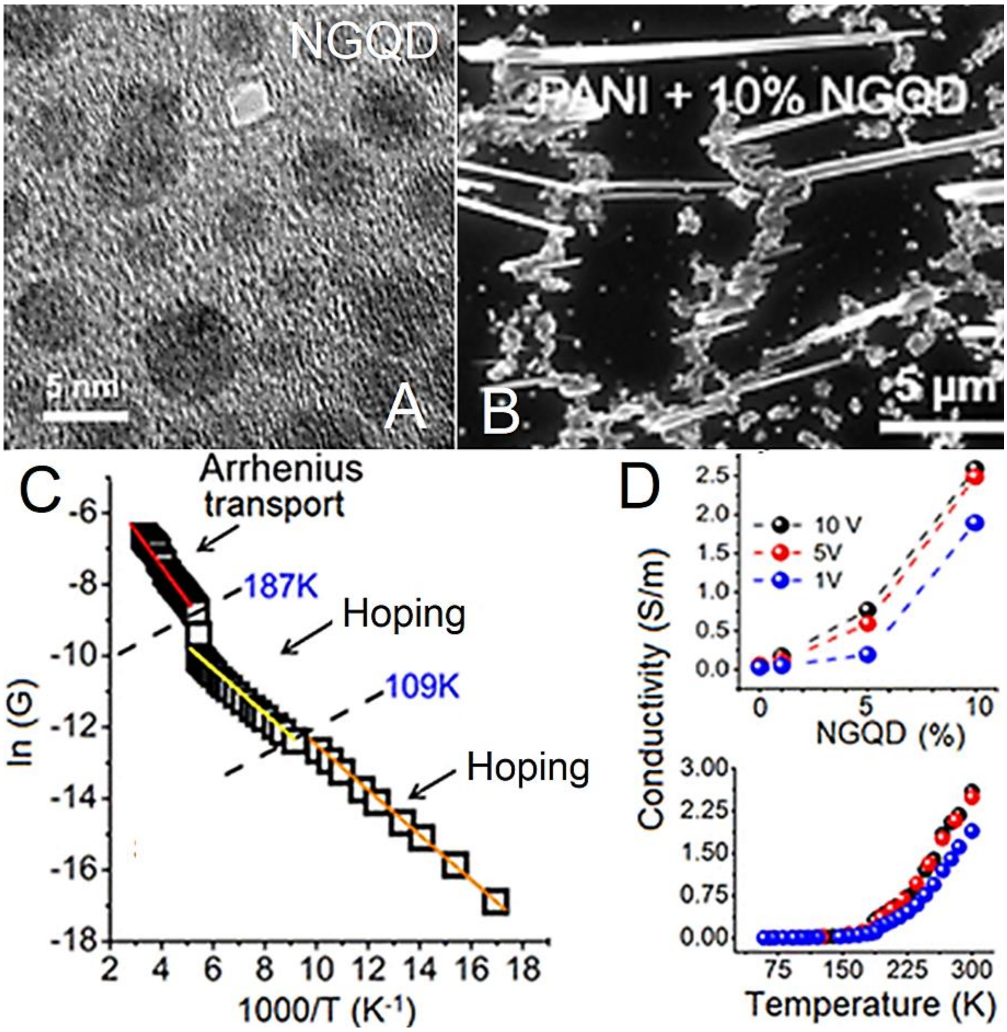

Fig. 3. (A) Bright field high resolution transmission electron microscopy micrograph of the NGQDs; (B) scanning electron microscopy micrographs of the polyaniline/nitrogen-functionalized graphene quantum dots nanocomposites; (C) variation of conductance (G) vs. 1000/T plotted on log scale for nanocomposite at bias Voltages of 10 V; (D) variation of electrical conductivity of (top) NGQD with temperature of the samples at three different bias voltages: 1, 5, and 10 V and (bottom) 10 % NGQD in PANI [61]. NGQD = Nitrogen-functionalized graphene quantum dots; PANI = polyaniline. Reproduced with permission from ACS

Fig. 3 C demonstrates Arrhenius plot to see the trend of conductance variation vs. temperature for the nanocomposite. It was observed that nanocomposite well-obeyed the Arrhenius law and foremost mechanisms for electron transportation seem to be Arrhenius transport and thermally induced hopping. Moreover, Fig. 3 D presents trends of electrical conductivity changes vs. temperature for pristine nitrogen functional graphene quantum dots and the related polyaniline nanocomposite studied for different bias voltages (1 V, 5 V, 10 V). Here, electrical conductivity had almost a linearly/exponentially rising trend with increasing temperature along with voltage, owing to conversion of benzenoid rings of polyaniline into more electrically conducting quinoid functionalities. Thus, this work pointed towards the important use of functional graphene quantum dots to beneficially alter electron transportation in polyaniline matrix against rising temperature and voltage effects, to meet the technological demands of high end photovoltaics.

Polypyrrole/graphene quantum dots

Polypyrrole has been explored as valuable conjugated matrix for graphene quantum dots, especially for their utilizations in florescent materials, optoelectronics, and heat/mechanically stable structures [62, 63]. Consequently, noteworthy combinations of polypyrrole and graphene quantum dots have been reported up till now depicting applications in sensors and supercapacitors [64, 65]. Li et. al. [66] designed the polypyrrole/sulfonated graphene quantum dots nanocomposites using in situ doping procedure and employing modified quantum dots contents of 75-160 mg mL^{-1}. Fig. 4 A shows the formation of sulfonated graphene quantum dots starting from the conversion of pyrene to 1,3,6-trinitropyrene, by nitration, and subsequent conversion to functional quantum dots via hydrothermal technique. Afterwards, introducing pyrrole monomers and in situ polymerization led to the formation of polypyrrole chains with dispersed sulfonated graphene quantum dots. Fig. 4 B shows microstructural analysis of tiny spherical pristine sulfonated graphene quantum dots with average sizes and lattice spacing, of around 5 nm and 0.21 nm, respectively. The synthesis and modification route used for quantum dots did not disturb the morphology, nanosizes, or dispersion of the quantum dots. Figs. 4 C & D depict scanning electron microscopy of the polypyrrole nanocomposite obtained with 75 mg mL^{-1} sulfonated graphene quantum dots. The resulting micrographs show uniformly dispersed polymer coated quantum dots. Fig. 4 E portrays specific capacitance retention vs. cyclic number plots for unfilled polypyrrole as well as 75 and 160 mg mL^{-1} quantum dots loaded samples. As per results, including 160 mg mL^{-1} nanofiller in polypyrrole revealed higher capacitance retention (~ 60 %), relative to that of 75 mg mL^{-1} sample (~ 50 %) and pristine quantum dots (~ 40 %), after 1000 cyclic recital. These results depicted positive effects of increasing sulfonated graphene quantum dots contents on enhancing the specific capacitance and conductivity of the polypyrrole matrix. Similarly, the electrical conductivity of the polypyrrole/sulfonated graphene quantum dots nanocomposites was observed as 7.4×10^{-3} and 2.1×10^{-2} S cm^{-1}, with 75 and 160 mg mL^{-1} doping, respectively, which was higher than neat matrix (0.5×10^{-2} $S\cdot cm^{-1}$). These results were attributed to fine electrostatic and aromatic stacking interactions between the polymeric chains and modified quantum dots, so supporting capacitance and conductivity characters for supercapacitor application. Furthermore, Zhou et. al. [67] explored the morphological properties and applied potential of polypyrrole/graphene quantum dots loaded nanocomposites via solution technique. Accordingly, Fig. 5 A shows a synthesis route for graphene quantum dots through pyrolysis, their subsequent conversion to the polypyrrole based nanomaterials, and finally interactions towards dopamine molecules for sensing purposes. As per transmission electron microscopy micrographs and high-resolution transmission electron microscopy micrographs (Figs. 5 B-D), polypyrrole/graphene quantum dots seem to have core shell nanoparticles with sizes, of 3 to 7 nm, and visibly marked boundaries due to the effectiveness of facile synthesis strategy applied. In addition, Table 1 shows sensing aptitude of the polypyrrole/graphene quantum towards dopamine detection in serum/urine samples. Herein, the nanocomposite had > 100 % efficiency for dopamine detection, both in serum and urine media. Consequently, these florescent sensors depicted useful detection limit, of 10 pM and linearity up to 8000 nM.

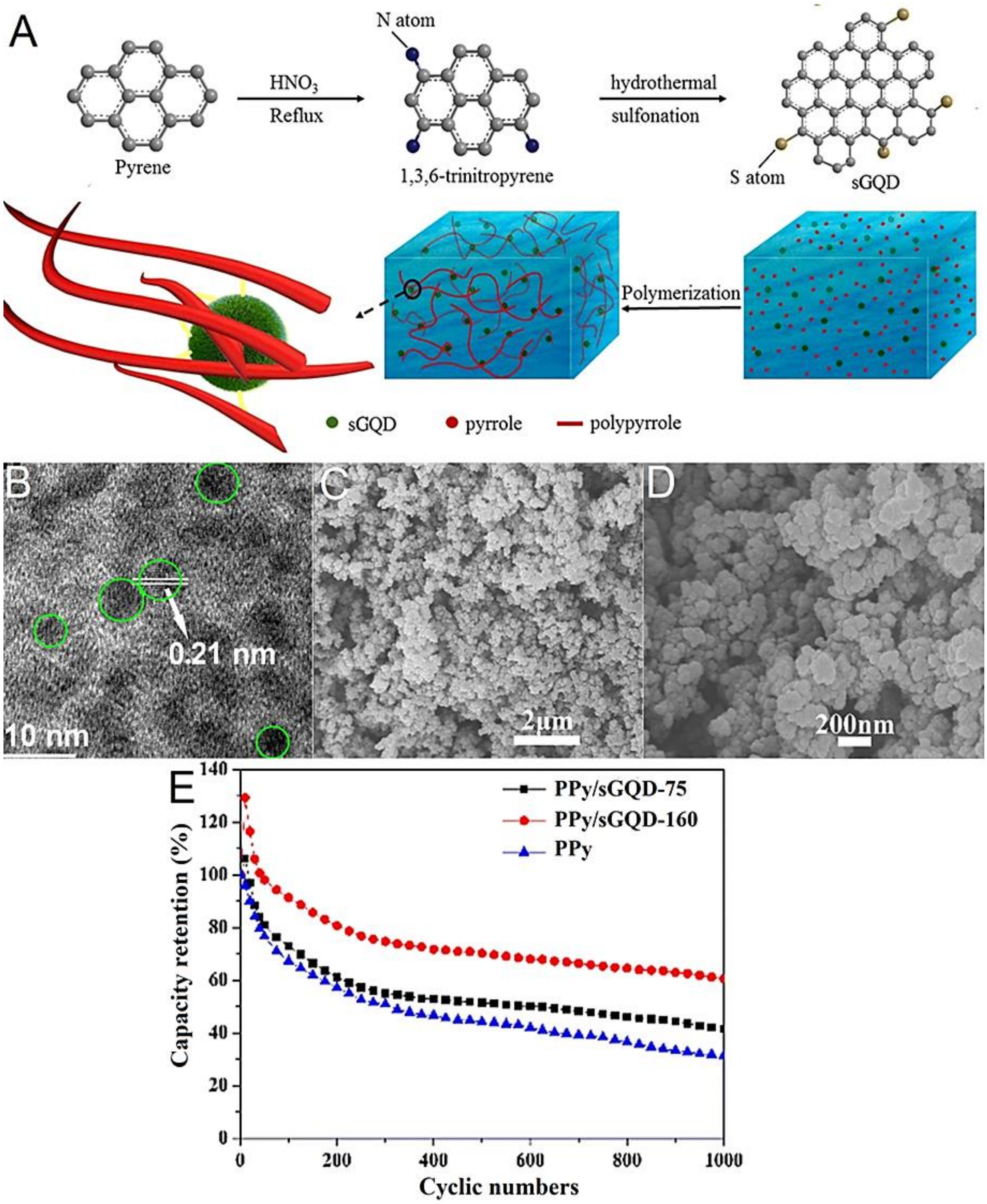

Fig. 4. (A) Schematic of PPy/sGQD nanocomposite formation; (B) high resolution transmission electron microscopy image of sGQD; (C & D) Scanning electron microscopy images of PPy/sGQD-75; (E) specific capacitance retention of neat polypyrrole and nanocomposites [66]. sGQD sulfonated graphene quantum dots; PPy = polypyrrole; PPy/sGQD = polypyrrole/sulfonated graphene quantum dots; where sGQD amount used 75 mg mL^{-1} and 160 mg mL^{-1}, in PPy/sGQD-75 and PPy/sGQD-160, respectively. Reproduced with permission from Springer

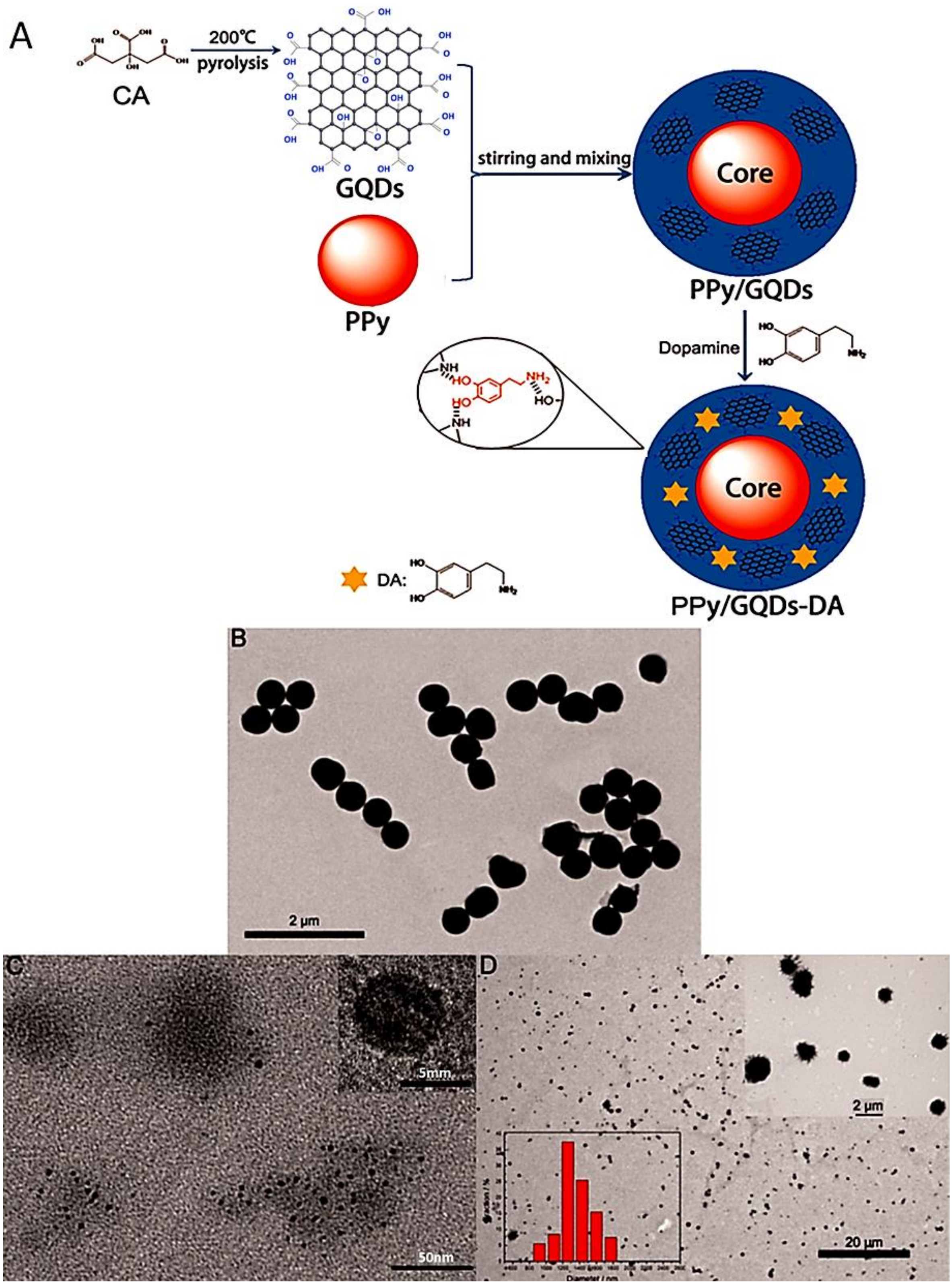

Fig. 5. (A) Synthesis scheme for PPy/GQDs and PPy/GQDs-DA. Transmission electron microscopy (TEM) images of (B) PPy; (C) GQDs (inset: HRTEM image of GQDs); and (D) PPy/GQDs (upper right inset: enlarged TEM image of the core-shell structure of PPy/GQDs, bottom left inset: the corresponding size distribution of PPy/GQDs) [67]. CA = citric acid; PPy = polypyrrole; GQD = graphene quantum dots; DA = dopamine; PPy/GQDs = polypyrrole/graphene quantum dots; PPy/GQDs-DA = polypyrrole/graphene quantum dots/dopamine. Reproduced with permission from Elsevier

Table 1. *Determinations of dopamine in human serum and urine samples [67]. Reproduced with permission from Elsevier*

Sample	Spike (nM)	Found (nM)	Recovery (%)	RSD (n = 3, %)
Serum	50	51.05	102.10	1.75
	100	101.35	101.35	0.86
	500	492.65	98.53	1.52
	1000	978.80	97.88	1.47
	2000	2024.80	101.24	2.13
Urine	50	49.20	98.40	0.89
	100	97.65	97.65	2.27
	500	505.45	101.09	1.69
	1000	990.50	99.05	0.98
	2000	2065.0	103.28	1.73

Polythiophene/graphene quantum dots

Besides polyaniline and polypyrrole, polythiophene is another imperative conjugated polymer having efficient charge transportation, redox features, optoelectronic characters, and ecofriendly properties [68, 69]. In spite of this, limited processability of polythiophene backbone in organic solvents hinders its methodological applications, therefore research has been extended towards the development of modified polythiophene, functional derivatives, and nanocomposites having optimum solubility and hydrophilicity characters [70, 71]. Consequently, polythiophene as well as derived forms have been analyzed for key solicitations in the fields of electronics (sensors, diodes, etc.) and energy (batteries, solar cells, fuel cells, etc.) devices [72]. Regarding polythiophene/graphene quantum dots, few studied have been noticed in the literature so far [73, 74]. Routh et. al. [75], for instance, used a polythiophene derivative, i.e., polythiophene-g-poly[(diethylene glycol methyl ether methacrylate)-co-poly(N,N-dimethylaminoethyl methacrylate)], as a matrix materials for graphene quantum dots. The resulting polythiophene derivative/graphene quantum dots had reasonable power conversion efficiency (around 1.8 %) for photovoltaic applications. Henceforth, according to literature reported so far, including pristine as well as modified quantum dots in different conjugated polymers depicted essential enhancements in the electrical conductivity and charge transportation features, therefore leading to imperative technical applications for supercapacitors, sensors, and photovoltaic devices.

GRAPHENE QUANTUM DOTS BASED RUBBERY MATRIX NANOCOMPOSITES

Synthetic rubber/graphene quantum dots

Significant literature searches have been noticed to explore the graphene or functional graphene reinforcements in the natural rubber or commercial rubbery matrix [76, 77]. Lim et. al. [78], e.g., introduced graphene oxide in natural rubber matrix using melt blending technique. The resulting natural rubber/graphene oxide nanocomposite had 700 % higher mechanical strength, relative to the unfilled matrix due matrix-nanofiller associations and load transference features. Similarly, Yaragalla et. al. [79] designed natural rubber/thermally reduced graphene oxide based hybrids by melt mixing. Here, nanofiller loading remarkably upsurge the tensile modulus of natural rubber, by 282 %, probably due to mechanical interlocking of the polymer main chains. Likewise, abundant research attempts have been observed regarding the use of graphene and derivative nanostructures to enhance the mechanical and thermal profiles of the rubbery hybrids [80].

As an important graphene derivative, researchers also focused the influences of graphene quantum dots addition on the physical features of rubbery polymers [81, 82]. In this concern, quantum dots filled rubbers have been found technologically worthwhile for electronics, photonics, molecular sensors, and biomedical systems [83, 84]. In due course, adding graphene quantum dots in natural/synthetic rubbers depicted notable enhancements in the matrix-nanofiller compatibility, crystallinity, and so the structural stability of the rubber/graphene quantum dots nanomaterials [85]. Regarding synthetic rubber/graphene quantum dots nanomaterials, Xie et. al. [86] designed a system based on hydrogenated carboxylated nitrile rubber with graphene quantum dots (non-grafted nanocomposite) and amine functional graphene quantum dots (grafted nanocomposite) using a solution route. Fig. 6 A shows the formation of hydrogenated carboxylated nitrile rubber-grafted-graphene quantum dots nanocomposite and probable crosslinking in its structure. The amine functional graphene quantum dots were suggested to develop covalent interactions and grafting with the carboxylic functional rubbery matrix. Figs. 6 B a-c depict scanning electron microscopy images of pristine rubbery matrix, non-grafted, and grafted graphene quantum dots based nanocomposite, respectively. As per micrographs, the non-grafted hydrogenated carboxylated nitrile rubber/graphene quantum dots nanocomposite had visible nanofiller aggregations with sizes, of 0.5-2 μm, due to lack of covalent bonging or mutual crosslinking interactions. On the other hand, hydrogenated carboxylated nitrile rubber-grafted-graphene quantum dots nanocomposite had a visibly smooth morphology without amine modified graphene quantum dots nanoparticle aggregation. Moreover, Fig. 6 C illustrates fluorescence emission spectra of pristine rubber, hydrogenated carboxylated nitrile rubber/graphene quantum dots, and hydrogenated carboxylated nitrile rubber-grafted-graphene quantum dots nanocomposites at 420 nm. Herein, superior fluorescence intensity was perceived for the grafted nanocomposite sample, relative to non-grafted one or pristine nanoparticles, seemingly due to fine dispersion of amine functional graphene quantum dots and network formation in the rubbery matrix with any aggregation. In this way, modification and covalent interactions of graphene quantum dots with rubbers beneficially enhanced the technological features of these nanomaterials. Likewise, Sreenath et. al. [87] introduced graphene quantum dots as well as two dimensional graphene oxide nanofiller in the carboxylated acrylonitrile butadiene rubber matrix via hydrothermal, ultrasonication, and solution practices. Varying graphene oxide contents, of 0.5-4 phr, were used to form these rubbery nanocomposites. In this research attempt, hydrothermally prepared graphene quantum dots were converted to amine functional graphene quantum dots using triethylamine and 1-(3-dimethylaminopropyl)-3-ethylcarbodiimide hydrochloride (Fig. 7 A). The resulting amine functional graphene quantum dots acted as covalent crosslinkers between graphene oxide-graphene oxide nanosheets as well as between carboxylated acrylonitrile butadiene rubber matrix-graphene oxide. Figs. 7 B & C show transmission electron microscopy micrographs of carboxylated acrylonitrile butadiene rubber-grafted-graphene quantum dots-grafted-graphene oxide nanocomposite with 2 and 4 phr nanofiller contents, respectively. The functional graphene quantum dots, of 20-60 nm sizes, can be seen uniformly dispersed on the graphene oxide nanosheets owing to mutual covalent interlinking effects. Fig. 7 D illustrates tensile stress versus strain curves of pristine rubber, carboxylated acrylonitrile butadiene rubber/graphene quantum dots, caboxylated acrylonitrile butadiene rubber/graphene oxide, and carboxylated acrylonitrile butadiene rubber-grafted-graphene quantum dots-grafted-graphene oxide nanocomposite. Herein, 2 phr graphene oxide based grafted nanocomposites revealed 386% higher stress values relative to neat rubber, as well as non-grafted nanocomposites, due to covalent crosslinking and efficient load transfer through the matrix. In addition, 0.5 and 4 phr graphene oxide loaded covalently bonded nanomaterials raised the strength, by 289 % and 320 %, respectively, relative to neat rubber and non-functional quantum dots derived samples. On the other hand, owing to intrinsic elasticity, unfilled carboxylated acrylonitrile butadiene rubber matrix had 100 % strain, which was about 700-900 % higher than the carboxylated acrylonitrile butadiene rubber-grafted-graphene quantum dots-grafted-graphene oxide nanocomposites with 0.5-4 phr nanofiller loading. Thus, functional quantum dots were found to valuably upsurge the mechanical profile of the rubbery matrix via establishing covalent interlinking in matrix-nanofiller.

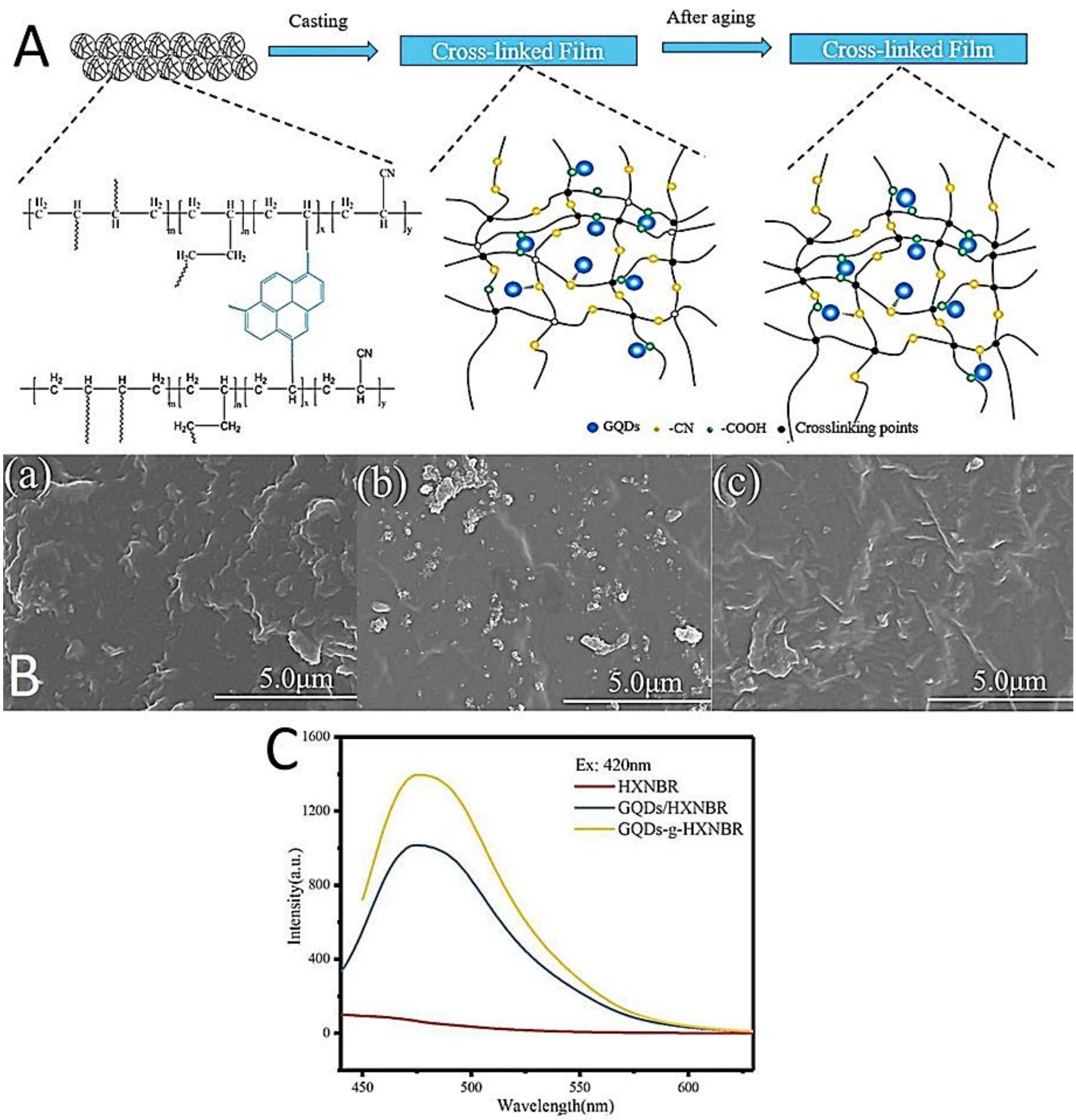

Fig. 6. (A) Preparation of graphene quantum dots modified hydrogenated carboxylated nitrile rubber (HXNBR) with interpenetrating crosslinking networks; (B) scanning electron microscopy images of (a) HXNBR; (b) non-grafted GQDs/HXNBR; (c) GQDs-g-HXNBR; (C) fluorescence emission spectra of HXNBR, GQDs/HXNBR, and GQDs-g-HXNBR films at 420 nm [86]. GQD = graphene quantum dots; GQDs/HXNBR = graphene quantum dots/hydrogenated carboxylated nitrile rubber; GQDs-g-HXNBR = graphene quantum dots-grafted-hydrogenated carboxylated nitrile rubber. Reproduced with permission from Springer

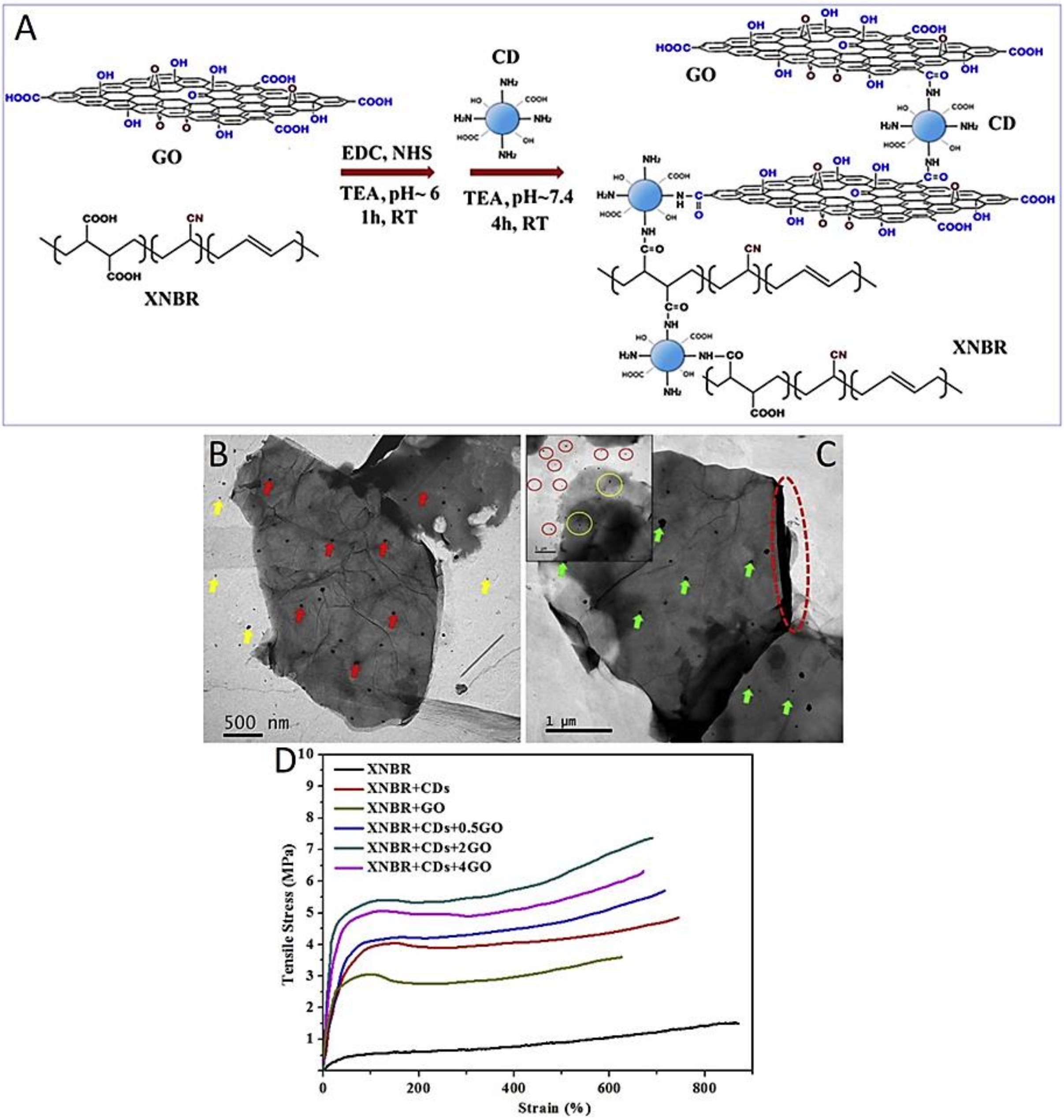

Fig. 7. (A) Schematic for the formation of carboxylated acrylonitrile butadiene rubber/graphene quantum dots/graphene oxide (XNBR/CDs/GO) nanocomposite; (B) transmission electron microscopy micrograph of XNBR/CD/GO 2 phr; (C) transmission electron microscopy micrograph of XNBR/CD/GO 4 phr; (D) tensile stress versus strain curves of XNBR, XNBR/CDs, XNBR/GO and XNBR/CD/GO with various ratios of GO [87]. CD = graphene quantum dot; GO = graphene oxide; NHS = N-hydroxysuccinimide; EDC = 1-(3-dimethylaminopropyl)-3-ethylcarbodiimide hydrochloride; TEA = triethylamine; XNBR = carboxylated acrylonitrile butadiene rubber; XNBR/CDs = carboxylated acrylonitrile butadiene rubber/graphene quantum dots; XNBR/GO = carboxylated acrylonitrile butadiene rubber/graphene oxide. Reproduced with permission from Elsevier

Additionally, few literature attempts have been noticed on silicone rubber/graphene quantum dots nanomaterials [88]. For instance, Li et. al. [89] developed γ-methacryloxy propyl trimethoxyl silane modified graphene quantum dots and incorporated in the silicone rubber matrix using solution and ultrasonic methods. The modified quantum dots, having sizes of about 2.6 nm, were grafted to silicone rubber through hydrosilylation. The ensuing silicon rubber-grafted-silane modified graphene quantum dots had bright blue fluorescence properties and so suggested for application in light emitting diodes. Recently, Koken et. al. [90] reported on the fluorescent silicon rubber/graphene quantum dots nanocomposites obtained using hydrothermal and solution methods. The resulting nanomaterials seemed to be valuable for sensing biodiesel contents in fuels via photoluminescence properties and solvatochromic/colorimetric shifts. Hence, unique fluorescent silicon rubber/graphene quantum dots were suggested for industrial level detection of biodiesel in fuel mixtures in UV light conditions.

Natural rubber/graphene quantum dots

Alongside synthetic rubbers, natural rubber/carbon dot nanocomposites have been reported in the literature [91, 92]. Edayadiyil et. al. [93], e.g., applied two roll milling and hydraulic pressing approaches for the formation of natural rubber/graphene quantum dot nanocomposites, as shown in Fig. 8 A.

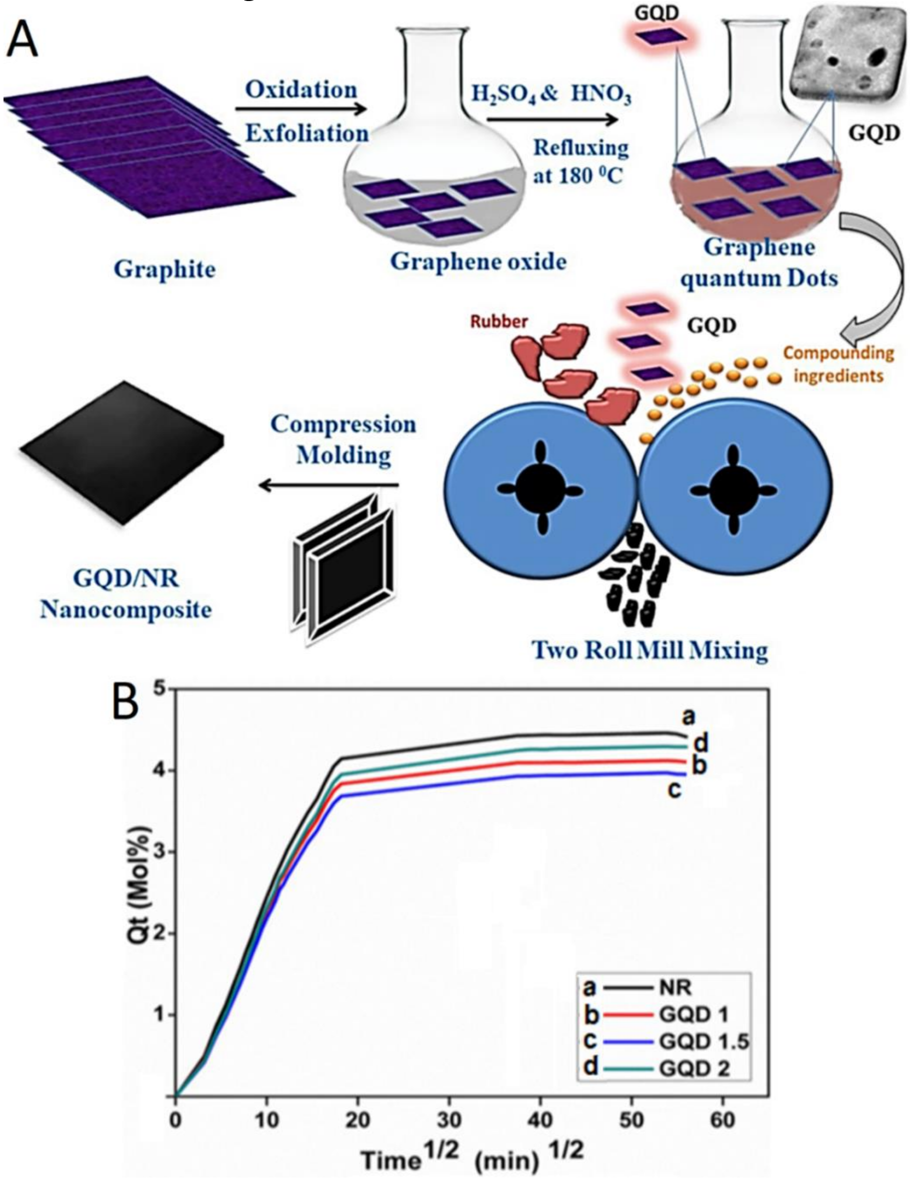

Fig. 8. (A) Schematic for the synthesis of natural rubber (NR) and graphene quantum dot (GQD) derived nanocomposites; and (B) effect of nanofiller loadings, of 1-1.5 wt.%, on the water uptake of natural rubber [93]. Reproduced with permission from Springer

Moreover, 1-2 wt.% loading effects of quantum dots were explored upon the water uptake properties of these nanocomposites (Fig. 8 B). According to the results, including quantum dots reduced the intrinsic water absorption properties of pristine natural rubber due to matrix-nanofiller anchoring effects, therefore, enhancing its main chain structural integrity and stable crosslinking tendencies. We notice research efforts on carbon dot filled rubbery nanocomposites, however, few specific reports seen up till now regarding natural rubber/graphene quantum dots therefore demanding future research on these nanomaterials to unveil their true application possibilities [94, 95].

Thus, according to the literature to date, pristine/functional quantum dots nanoreinforcements in natural/synthetic rubbery matrices enhanced the conductivity, florescence, and mechanical resilience of the resulting nanocomposites for high tech engineering and energy device applications.

TECHNICAL ASPECTS OF CONDUCTING MATRIX/GRAPHENE QUANTUM DOTS AND RUBBERY MATRIX/GRAPHENE QUANTUM DOTS NANOCOMPOSITES

One of the important scientific applications of carbon dots has been reported in the field of sensors, i.e., florescent, molecular, and gas sensors [96, 97]. Similarly, conjugated polymers also own valuable sensing properties due to inherent reception sites towards desired analyte molecules [98]. Consequently, conjugated polymers and graphene quantum dots have been beneficially combined to see their mutual effects on molecular/gas sensing properties [99]. Gavgani et. al. [100], for instance, developed polyaniline and graphene quantum dots derived nanocomposites through in situ chemical oxidative polymerization method. The resulting polyaniline/graphene quantum dots hybrids revealed superior sensitivity and selectivity towards ammonia gas sensing, compared with pristine quantum dots as well as unfilled polyaniline, owing to synergistic matrix-nanofiller effects. Further, Hong et. al. [101] proposed a nanocomposite based on polyaniline, nitrogen doped graphene quantum dots, and indium trioxide. Initially, nitrogen doped graphene quantum dots were synthesized by hydrothermal technique. Later, polyaniline/nitrogen doped graphene quantum dots/indium trioxide nanocomposite nanofibers were designed using electrospinning and high temperature calcination techniques. Accordingly, Fig. 9 A shows ammonia sensing mechanism of the as prepared nanocomposite, relying upon the formation/interactions of n type nitrogen doped graphene quantum dots and p type polyaniline matrix. As per analysis, upon gas exposure, a Fermi level equilibrium was generated in the nanocomposite due to the movements of holes (polyaniline) and electrons (In_2O_3) in opposite directions leading to efficient sensing properties. Fig. 9 B demonstrates selectivity of the polyaniline/nitrogen doped graphene quantum dots/hollow indium trioxide nanocomposite sensor towards ammonia, methanol, ethanol, and acetone. The resulting nanocomposite nanofibrous sensor had greater resistance, of 34 k ohm, for ammonia, compared with neat polyaniline (15 k ohm), seemingly due to aromatic stacking interactions between polyaniline-nitrogen doped graphene quantum dots. Accordingly, the polyaniline/nitrogen doped graphene quantum dots/indium trioxide nanofibrous sensors had superior response towards ammonia molecules, relative to other gases, due to efficient p and n type transportation mechanisms involved towards ammonia molecules. Accordingly, conjugated polymer/graphene quantum dots based sensors were also suggested worthwhile for sensing volatile compounds and other molecules [102, 103].

Besides, few literature reports have been found regarding the sensing application of the synthetic or rubbery polymers [104, 105]. Especially, rubbery polymers have been beneficially used in piezoresistive sensing devices for structural examinations of the automobile body parts [106]. In this concern, few literatures so far reported the natural rubbers/quantum dots based piezoresistive sensors for monitoring the mechanical damages of the transport vehicles [107-109].

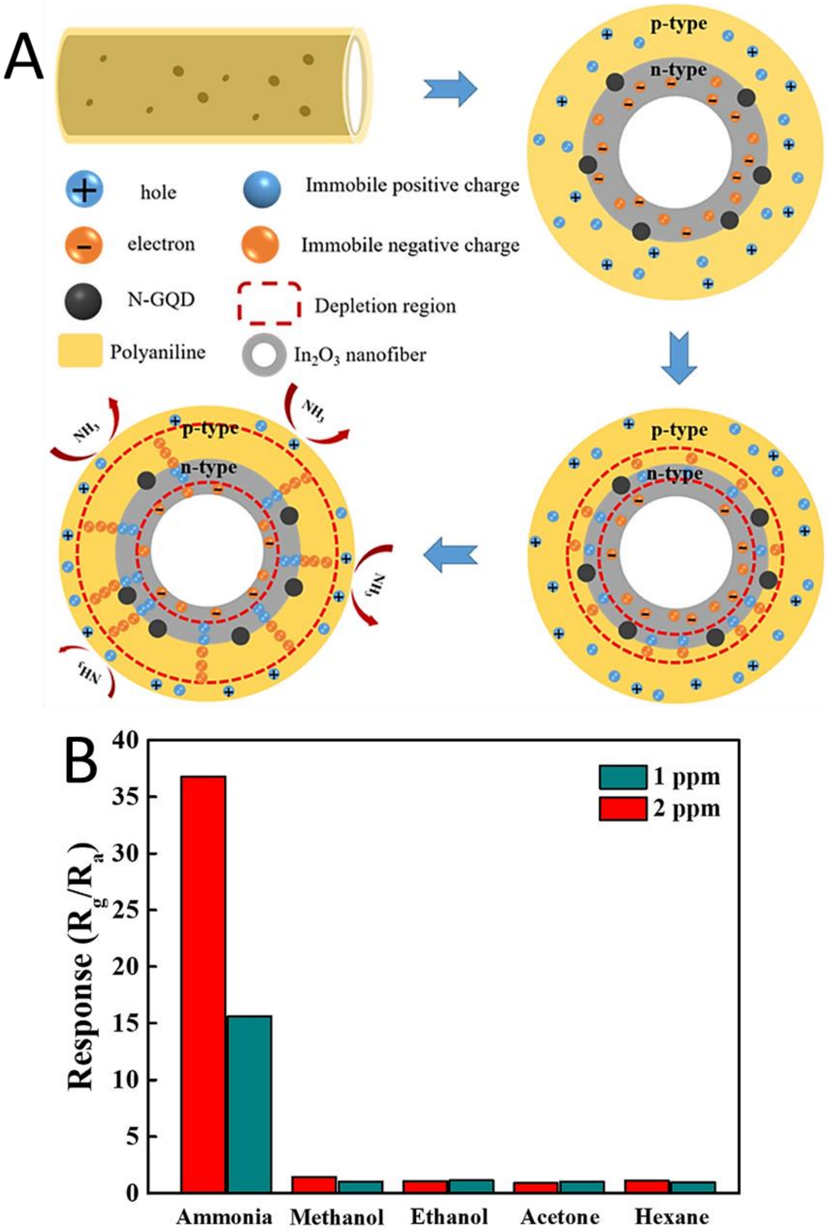

Fig. 9. (A) Schematics of the sensing mechanism and energy band gap structure diagram of PANI/N-GQD/In_2O_3 nanocomposite nanofiber sensors; (B) selectivity of the PANI/N-GQD/In_2O_3 nanofiber sensor tested at 1 and 2 ppm of NH_3, methanol, ethanol, and acetone [101]. PANI/N-GQD/In_2O_3 = polyaniline/nitrogen doped graphene quantum dots/indium trioxide; N-GQD = nitrogen doped graphene quantum dots. Reproduced with permission from MDPI

As per literature surveys, another important use of the conjugated polymers and derived materials has been investigated for supercapacitors [110, 111]. Nevertheless, limited research attempts hitherto have been noticed on conjugated polymer/graphene quantum dots based supercapacitor electrodes [112, 113]. Recently, Arthisree, [114] et. al. explored the green synthesized nanocomposites of conjugated polymers, like polyaniline, polypyrrole, and poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate), with graphene quantum dots for supercapacitor application. The resulting nanocomposites had superior performance as supercapacitor electrode having remarkably high specific capacitance, of nearly 5000 $Fg^{-1}cm^{-2}$, due to the formation of electron conducting network between matrix-nanofiller. They depicted significantly higher specific capacitance in the range of 300-1001 Fg^{-1} with increasing nanofiller contents of 0.5-1.5 wt.%. As per literature, traditional polyaniline/graphene based supercapacitor electrodes, for example, as proposed by Wu et. al. [115], had specific capacitance of around 200 F g^{-1}. Similarly, Sahoo et. al. [116] developed supercapacitor electrodes based on polypyrrole/graphene with specific capacitance of about 466 F g^{-1}. On the other hand, using polyaniline/graphene quantum dot nanocomposites, e.g., in a recent report by Zhao et. al. [117] documented notably higher specific capacitance, of 998-1120 Fg^{-1}, for adding reduced graphene quantum dots to supercapacitor electrodes with ruthenium oxide nanoparticles. Here, reduced graphene quantum dots based nanostructures revealed 89 % capacitance retention, i.e., significantly higher than conventional ruthenium oxide nanoparticles based electrode (38 %). According to results, it appears that graphene quantum dots can beneficially enhance the electrical conduction and charge transfer through the nanomaterials, and so the resulting electrodes were recommended for high tech supercapacitor applications

In addition, energy conversion potential of zero dimensional graphene quantum dots and related nanomaterials has been noticed due to their superior surface area, functionalities, and electrical conductivity properties, especially for solar cell devices [118]. In this concern, few literature reports have been observed for the use of graphene quantum dots for dye sensitized solar cells, organic polymer solar cells, bulk heterojunction solar cells, etc. [119-121]. Here, Dinari et. al. [122] developed polyaniline/graphene quantum dot nanocomposites for solar cells. The resulting hybrid systems yielded power conversion efficiency of 1.6 % in dye sensitized solar cells. Chen et. al. [123] formed polypyrrole/graphene quantum dots nanocomposites for bulk heterojunction solar cells. The as designed nanocomposite had superior power conversion efficiency of > 5 %, relative to traditional platinum electrodes. Recently, Hoang et. al. [124] filled poly(3-hexylthiophene):[6,6]-phenylC61-butyricacid methylester and poly(3,4-ethylenedioxy-thiophene):polystyrene sulfonate based solar cell designs with graphene quantum dots. The resulting system had superior solar cell efficiency of 1.43 %, i.e., 44 % higher than unfilled system. Nevertheless, future developments of graphene quantum dots for high performance solar cells seem to be connected to comprehensive focused endeavors by the field researchers.

Self healing polymeric materials have been traditionally used in various technical industries using mechanically robust and anticorrosion structural designs [125, 126]. Here, development and use of smart and self healing coatings/nanomaterials have been practiced to protect/replace conventional metal based industrial architectures [127, 128]. In addition to pristine polymers [129], graphene based self healing coatings have been used to enhance the structural strength and anticorrosion performance of the nanocomposites [130]. Recently, polymer/graphene quantum dot nanocomposites have been found efficient for mechanical and anticorrosion related self healing effects [82, 131]. In this regard, thermoplastic, thermosets, as well as conductive polymers filled with graphene quantum dots attained a recent research focus [132]. Self healing efficiencies of graphene quantum dots based nanocomposites have been reported > 97 % at room temperature conditions [133]. Although, more concentrated efforts seem desirable to unfold the true potential of graphene quantum dots based self healing nanomaterials.

In biomedical applications, like tissue engineering, rubber based nanocomposites have been applied for tissue regeneration and fabrication of artificial scaffolds [134-136]. In bone tissue engineering, natural rubber/hydroxyapatite nanomaterials have been applied for constructing strengthened tissue scaffolds [137, 138]. Following this, quite a few researches so far mentioned the synthesis and mechanical worth of natural rubber/hydroxyapatite/graphene quantum dots based bone tissue scaffolds [139, 140]. Nevertheless, research on biomedical uses of nanocarbons, like graphene quantum dots, revealed challenges of toxicity [141], neurotoxicity [142], microbial infections [143], organ damage [144], and other biological problems. Usually, penetration of carbon nanoparticles in living cells/tissues may cause serious cytotoxicity effects [145]. However, toxicity effects of graphene quantum dots can be controlled by identifiying the minimum threshold level for safe use of these tiny nanoparticles in biological systems. Therefore, using graphene quantum dots can be valuable for biological systems due to intrinsically tunable surface chemistry, non-toxicity, purity, and fluorescence effects [146]. In this concern, recent research trends have mentioned the use of non-toxic stimuli responsive polymer/quantum dots materials for drug delivery and tissue engineering applications [147].

It is also important to compare the performance of graphene quantum dots with other carbon based quantum dots, like carbon dots or polymer dots. Both graphene quantum dots and carbon dots own high surface area, low toxicity, fine solubility, chemical inertness, fine electronic/electrical properties, charge transference, and facile synthesis to be employed for high tech industrial applications [148]. Among similar application grounds, graphene quantum dots and carbon dots have been technically applied in the field of photoluminescence sensors, electrochemical sensors, bioimaging, drug/gene delivery, and other biological uses. However, when we compare properties of graphene quantum dots with carbon dots or polymer dots, graphene quantum dots possess superior crystallinity due to the presence of layered nanostructure [149]. In sensors or biosensors application, graphene quantum dots own several times higher sensitivity relative to carbon dots or inorganic quantum dots [150]. For supercapacitors, graphene quantum dots have remarkedly rapid electron transference properties, relative to carbon dot nanoparticles [151]. Similarly, electrocatalytic activity of graphene quantum dots has been found superior relative to other known quantum dots so far, which can be beneficial for important applications like H_2 production [152].

CHALLENGES, FUTURE AND CONCLUSIONS

In preceding sections of this in-depth manuscript on unique classes of conjugated polymer/graphene quantum dots and rubber/graphene quantum dots, we have surveyed almost all possibly reported aspects of these nanomaterials, covering synthesis—to—applications (Fig. 10 & Table 2). As per literature explorations so far, future of conjugated and rubbery matrices filled with graphene quantum dots rely upon extensive focused field investigations to unveil the commercial scale potential of these nanomaterials for electronics, energy, and biomedical related fields. Additionally, we suggest imperative future possibilities of graphene quantum dots based nanomaterials in fluorescent devices. Henceforth, major challenging factor, in enhancing the applied potential of conjugated or rubbery polymer and graphene quantum dots nanomaterials, is the lack of comprehensive recent surveys on controlled processing and underlying mechanisms responsible for electron/charge transportation and fluorescence features. Consequently, researchers must strive to define seamless synthesis methods and optimized conditions to attain high performance conjugated polymer/graphene quantum dots or rubbery matrix/graphene quantum dots having desirable physical contours for next generation technical applications. Fleetingly, factual property improvement mechanisms/phenomenon as well as synergistic effects operative in polymer-graphene quantum dots need to be sightseen to gain future insights of these technically promising nanomaterials.

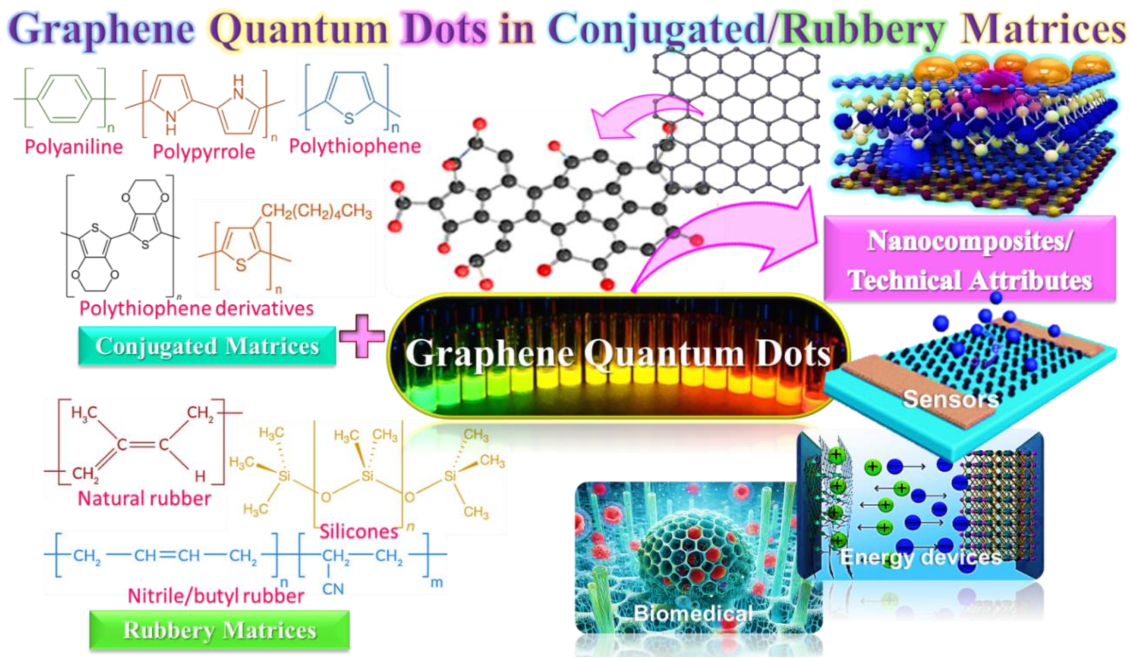

Fig. 10. Prospects of graphene quantum dots in conjugated and rubbery polymers

Table 2. *Important aspects and technical implication of polymer/graphene quantum dots nanomaterials*

Polymer	Nanoadditive	Fabrication technique	Physical properties/significance	Ref
Polyaniline	Graphene quantum dots	Chemical oxidation polymerization	Charge and electron transference	[53]
Polyaniline	Graphene quantum dots	*In situ* chemical oxidation	Supercapacitors; specific capacitance > 1000 Fg^{-1}	[55]
Polyaniline	Graphene quantum dots	Screen printing method	Cr^{6+} ion detection > 106%; Water purification membrane	[56]
Sulfonated polyaniline	Graphene quantum dots	*In situ* chemical oxidation	Supercapacitors; specific capacitance 245 F g^{-1}	[59]
Polyaniline	Nitrogen doped graphene quantum dots	Hydrothermal method; in situ technique	Arrhenius law; electrical conductivity changes vs. temperature; mechanisms of electron conduction by Arrhenius transport and thermally induced hopping	[61]
Polypyrrole	Graphene quantum dots	Hydrothermal technique; in situ method	Supercapacitors; specific capacitance retention ~ 50-60 %; 1000 cyclic recital; electrical conductivity 7.4×10^{-3} $2.1\text{-}\times10^{-2}$ S cm^{-1}	[66]

Polypyrrole	Graphene quantum dots	In situ; core-shell	Dopamine sensing efficiency > 100 %; detection limit 10 pM; linearity up to 8000 nM	[67]
Polythiophene derivative, i.e., polythiophene-g-poly[(diethylene glycol methyl ether methacrylate)-co-poly(N,N-dimethylaminoethyl methacrylate)]	Graphene quantum dots	Solution technique	Photovoltaics; power conversion efficiency 1.8 %	[75]
Hydrogenated carboxylated nitrile rubber	Graphene quantum dots; amine functional graphene quantum dots	Solution route	Covalent interactions/grafting in matrix-nanofiller; Homogeneous dispersion; fluorescence emission studies	[86]
Carboxylated acrylonitrile butadiene rubber	Graphene quantum dots; two dimensional graphene oxide 0.5-4 phr	Hydrothermal; ultrasonication; solution practices	Tensile stress 300-400 % higher than neat rubber; 700-900 % higher strain of unfilled rubber, relative to covalently linked nanocomposites	[87]
Silicone rubber	Graphene quantum dots	Hydrothermal approach; solution method	Fluorescent sensors; detection of biodiesel in fuel mixtures under UV light	[90]
Silicone rubber	γ-methacryloxy propyl trimethoxyl silane modified graphene quantum dots	Solution tactic; ultrasonic method	Bright blue fluorescence; light emitting diode	[89]
Natural rubber	Graphene quantum dots	Two roll milling/ hydraulic pressing	Quantum dots reduced the intrinsic water absorption of pristine natural rubber	[93]
Polyaniline	Graphene quantum dots	In situ chemical oxidative polymerization	Ammonia gas sensing; high sensitivity/ Selectivity	[100]
Polyaniline nanofibers	Graphene quantum dots; indium trioxide	Electrospinning; high temperature calcination	Ammonia sensing; resistance 34 k ohm; p-type and n-type transportation	[101]
Polyaniline; polypyrrole; poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate),	Graphene quantum dots	Green in situ synthesis routes	Supercapacitor electrodes; specific capacitance 5000 $Fg^{-1}cm^{-2}$	[114]
Natural rubber	Graphene quantum dots	Solution route	Piezoresistive sensors; damages sensing of transport vehicles	[107-109]

As per literature, most of research on graphene quantum dots is still in lab scale in small laboratories and research institutions [153]. Here, lack of sufficient focused lab scale researches and prototyping can be seen as major factors hindering large scale manufacturing and commercialization of graphene quantum dots. Therefore, focused future efforts seem desirable for industrial scale evolution of graphene quantum dots based nanomaterials [154]. The basic research and development (R&D) programs must be supported by relevant industrial and government agencies for large scale productions [155]. For this purpose, economic likelihoods for large scale production of graphene quantum dots keeping in view of sustainability aspects must be considered [156]. Along with repeated R & D scientific attempts, rapid patent reports and licensing procedures definitely seem desirable for commercialization of graphene quantum dots [157, 158]. Afterwards, coordination between researchers/engineers and mass manufacturing teams can lead to industrial scale designs [159]. Ultimately, large scale designed prototypes need involvement of marketing units/companies for commercialization [160, 161].

By means of non-hazardous green nanomaterials like graphene quantum dots can definitely play important role to decrease the environmental burden for eco-safe sustainability solutions [162-164]. Traditional metal or carbon nanoparticles have been found to affect the environment, although used in minor amounts [165]. In this concern, greenhouse emissions and ecological impacts of using quantum dots and traditional carbon nanoparticles have been comparatively analyzed [166]. Consequently, estimated life-cycle and impact of graphene quantum dots on our ecosystem have been explored [167]. Particularly, cradle-to-grave life cycle analysis of graphene quantum dots revealed need of replacing traditional nanoparticles with these sustainable green nanodots [168]. Several environmentally green and sustainable routes have been discovered for safe degradation of graphene quantum dots by using natural heat/light, microorganisms, or environmental effects [169]. Therefore, graphene quantum dots derived green nanomaterials have attained research curiosities for future aerospace, defense, devices, and biomedicals industries [170]. For example, Research on geopolymers and graphene quantum dots can expand this field towards sustainable construction material with superior durability, mechanical and thermal resistance properties [171]. To expand the field of rubber/graphene quantum dots towards sustainable green nanocomposites, waste crumbs rubber can be modified with quantum dots nanoparticles to beneficially upsurge mechanical properties and structural resilience for construction industry [172].

According to our analysis, future investigations must focus on novel designs and structure-property relationships of polymer/graphene quantum dots nanocomposites for high temperature stability, electrical conductivity, mechanical robustness, permeability, wear, anticorrosion, etc., to unveil promising applications for aerospace, engineering structures, defense, energy, electronics, membranes, biomedical, and so on [173]. For this purpose, adding graphene quantum dots in conjugated/rubbery polymers by using an appropriate method may develop structure-property relationships in the nanocomposites via matrix-nanofiller interactions and nano-reinforcement effects [174]. Herein, key functional parameters governing overall property enhancements of polymer/graphene quantum dots nanocomposites seem to rely on physical/covalent interactions and interfacial effects between polymer-nanodots. Nevertheless, hardly any literature seen so far on design-property-performance aspects of graphene quantum dots based nanocomposites, which may be helpful for attaining next generation technological materials and applications. We conclude that future progress of graphene quantum dots filled conjugated/rubbery polymers relies on comprehensive research efforts for design optimization, physical property enhancements, interfacial design/interactions, and actual mechanisms involved in structure-property relationships of these nanomaterials [175].

For important industrial applications of graphene quantum dots in energy, space, automotive, or defense related sectors, superior heat stability of these nanomaterials seems indispensable, in addition to mechanical resilience. For designing thermally stable polymer/graphene quantum dots nanomaterials, nanofiller contents, dispersion, surface groups, and compatibility with the matrices have been found accountable for superior performance of these nanomaterials. In this concern, Schneider et al. [176] observed reasonably high glass transition temperature of 120 °C for 5 wt.% graphene quantum dots loaded nanomaterials, relative to neat polymer (70 °C). George et al. [177] also documented superior thermal stability of polymer/graphene quantum dot nanocomposite having no considerable degradation by heating up to 800 °C and had high char yield of about 62 %. It seems that quantum dots nanoparticles may act as efficient thermal barriers to hinder macromolecular distortion, therefore, enhancing the heat stability of polymer/graphene quantum dots nanocomposites.

To see actual future commercial level graphene quantum dots derived nanomaterials, high tech design and technical features/performance must be focused by the field researchers. For future commercial markets, graphene quantum dots derived systems seemed beneficial for next generation solar panels [178], luminescent diodes [179], sensing [180], bioimaging [181], batteries [182], super/microcapacitors [183], labs on chips [184], and fluorescent nanoelectrochemical (NEMS) [185].

Conclusively, this state-of-the-art and purposeful review article argues two noteworthy categories of polymeric materials (conjugated polymers and rubbers) reinforced with graphene quantum dots. For these nanocomposites, facile synthesis approaches have been employed, including in situ polymerization, solution mixing, and melt blending methods. Including graphene quantum dots in conjugated/rubbery polymers seems to beneficially alter the microstructure, intermolecular macromolecular interactions, and resultant physical attributes (electrical/charge transport, mechanical/thermal stability, etc.) of these nanocomposites. In this regard, we systematically probed the structure-property-potential features of graphene quantum dots derived nanomaterials under important categories of conducting/elastomeric polymers. Last but not the least, all the above stated design/characteristics/performance attributes of the conjugated polymer/graphene quantum dots and rubbery matrix/graphene quantum dots point towards a crucial necessity of focused scientific efforts to improve their promising possibilities for sensors, supercapacitors, photovoltaics, biomedical, and limitless unexplored technical areas up to now.

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