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Multifunctional Hybrids of Graphene Quantum Dots with Inorganic Nanoparticles (Metal, Metal Oxide and MOF) - Topical State and Evolutions

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Abstract

Carbon based quantum dots have been discovered as unique florescent nanoentities having different types, such as carbon nanodots, graphene quantum dots, and polymer dots. Out of these, graphene quantum dots can be seen as zero dimensional derivatives of graphene (2 dimensional nanosheet). Due to recent advancement in the field of graphene quantum dots, various inorganic and organic hybrids have been reported in the literature so far. In this concern, inorganic nanoparticles like metal, metal oxide, as well as metal organic framework (MOF) have been used to design graphene quantum dots/metal nanoparticles, graphene quantum dots/metal oxide nanoparticles, and graphene quantum dot/MOF type hybrids. These nanomaterials have been designed using efficient synthesis strategies including hydrothermal, solvothermal, freeze dying, solution, in situ, and several other methods. Accordingly, this state-of-the-art review article aims to highlight advantageous physical properties, such as microstructure, electron/charge conduction, florescence, catalytic activity, etc., in addition to dispersion, compatibility, and interface formation of graphene quantum dots with inorganic nanoparticles. Consequently, high tech application areas of graphene quantum dots/inorganic nanoparticle hybrids have been identified for optical sensors, supercapacitors, photocatalysts for environmental treatment, and antimicrobials for wound healing purposes. Despite the success of graphene quantum dots/inorganic nanoparticle hybrids so far, focused future research efforts on these nanomaterials may lead to large/commercial scale applications in energy, electronics, environment, and biomedical fields by overcoming underlying design/synthesis/performance challenges.

Keywords: Graphene quantum dots, Metal/metal oxides, MOF, Hybrids, Optical sensors, Photocatalysts, Wound healing

Introduction

Graphene, since its discovery, has been analyzed for nanostructural uniqueness (1 atom thick 2 dimensional nanosheet) and remarkable physical properties (electronic, electrical, thermal, mechanical) [1]. Wide ranging top down and bottom-up synthesis methods have been used for the fabrication of graphene and its modified nanostructures. Consequently, pristine graphene and resulting hybrids exhibited promising applications in the fields of photovoltaics, light emitting diodes, fuel cells, sensing/capacitive devices, charge storage batteries, ecological materials, biomedical appliances, and countless other technological areas [2,3]. Due to synthesis and optimal property challenges of graphene, its modified forms and nanocomposites have been reported for high end advancements in the potential applied areas [4].

In this regard, graphene quantum dots appeared as inimitable graphene derivatives [5]. Graphene quantum dots are basically a type of carbon based quantum dots, besides carbon nanodots and polymers dots. Like pristine graphene, graphene quantum dots have also been investigated for their fabrication, properties, and applications as advanced hybrid nanostructures [6,7]. Similar to other quantum dots, graphene quantum dots revealed quantum confinement, edge/surface effects, florescence, and other advantageous physical properties [8]. Consequently, graphene quantum dots have been studied for valuable energy/electronics and biomedical (bioimaging, biosensing, drug delivery, etc.) applications [9,10]. In the form of hybrid nanostructures, graphene quantum dots have superior dispersion, tunable nanostructure, adjustable properties, and high tech applications [11]. In this concern, high performance inorganic hybrids of graphene quantum dots have been designed using metal nanoparticles [12], metal oxides [13], and metal organic frame (MOF) [14]. According to the literature so far, applications of graphene quantum dot/inorganic nanoparticle hybrids have been noted for electronic devices, energy devices, environmental treatments, and biomedical areas [15,16]. Despite the to date technical success of graphene quantum dots/inorganic hybrids, several existing field challenges have been observed for these nanomaterials, which can be overcome via focused future research investigations by the field scientists [17-19].

This novel review article has been designed to emphasize almost every possible structural, synthesis, physical property, and applied prospect of graphene quantum dot and metal, metal oxide, and MOF derivative hybrids. In this regard, important graphene quantum dot/inorganic designs, morphological, conducting, florescence, catalytic, and other physical features have been reported in the literature so far. Nevertheless, future progress and industrial scale uses of graphene quantum dot/inorganic hybrids can be achieved through comprehensive research attempts to overcome underlying design, compatibility, and processing challenges to enhance structure-property-performance relationships of these nanomaterials [20,21].

Graphene quantum dot hybrids with metal nanoparticles

Graphene quantum dots have been modified to form hybrid nanostructures using numerous functional nanomaterials [22,23]. In this concern, metal nanoparticles have been considered as promising nanoadditives for graphene quantum dots to design hybrid nanostructures [24,25]. As per scientific data, pristine metal nanoparticles and their nanocomposites have been researched for widespread technical applications, ranging from engineering materials and electronic/energy devices to biomedical (drug transfusion, bioimaging, etc.) sectors [26-28]. Accordingly, metal nanoparticle based nanocomposites have been designed using matrices, such as polymers, carbon, and inorganic nanomaterials (metal/metal oxide) [29,30]. Consequently, literature reports have been observed on design/fabrication, physiochemical characteristics, and applications of variety of metal nanoparticles (gold, silver, iron, zinc, copper, etc.) and related hybrids [31-33]. It seems that due to nanolevel sizes, interfacial interactions, and compatibility, metal nanoparticles can develop synergistic structural and functional relationships with graphene quantum dots [34,35]. In this regards, we found a number of scientific surveys up till now on the hybrids of graphene quantum dots with metal nanoparticles [36]. Notably, Ibarra *et. al.* [37] applied hydrothermal technique for the formation of graphene quantum dots using various nanocarbon precursors, including graphene, carbon nanotube, and amorphous carbon. Then, high performance nitrogen doped graphene quantum dots and gold (Au) nanoparticle based hybrids were also hydrothermally synthesized. **Figures 1A(a)** and **1A(b)** show transmission electron microscopy micrographs of graphene quantum dots (prepared using graphene precursor). Graphene quantum dots can be observed as hexagonal nanoparticles of 20 - 50 nm occurring as aggregates. **Figures 1A(c)** and **1A(d)** show transmission electron microscopy micrographs of nitrogen doped graphene quantum dots/gold nanoparticle hybrids. As per images, Au nanoparticles (lattice spacing 0.24 nm) seemed to be uniformly dispersed within the nitrogen doped graphene quantum dots aggregates, due to interfacial coupling effects between these nanoparticles. **Figures 1B(a)** and **1B(b)** show color differences of graphene derived nanodots in visible light (reddish brown) and UV light (bluish green), respectively. In addition, **Figure 1(C)** demonstrates photoluminescence spectra of graphene quantum dots, which were hydrothermally prepared by different nanocarbon sources. Accordingly, graphene, carbon nanotube, and amorphous carbon depicted emission peaks at 488, 490, and 496 nm, respectively. Here, photoluminescence features of the nanodots were seemed to be dependent upon their sizes (according to precursor nanocarbon used during their synthesis) as well as quantum confinement effects.

Zhang *et al.* [38] applied hydrothermal technique to form nitrogen doped graphene quantum dots using green precursors (hydroxyethyl cellulose/L-citrulline). Afterwards, hybrids of graphene quantum dots with silver (Ag) nanoparticles were prepared using silver nitrate salt, in a solution method (**Figure 2(A)**). **Figures 2B(a)** and **2B(b)** show transmission electron microscopy micrographs of nitrogen doped graphene quantum dots and nitrogen doped graphene quantum dots/Ag hybrid, respectively. As per results, pristine nitrogen doped graphene quantum dots had uniform spherical morphology, whereas nitrogen doped graphene quantum dots/Ag hybrid revealed dispersion of silver nanoparticles attached to quantum dot surfaces. In high resolution transmission electron microscopy micrograph, a clear interface formation can be seen between the nanodots and inorganic nanoparticles (**Figure 2B(c)**). Consequently, **Figures 2(C)** and **2(D)** show florescence excitation and emission spectra of graphene quantum dots and their nanocomposites, respectively. As per results, nitrogen doped graphene quantum dots depicted narrow emission band (400 - 495 nm) due to narrow particle size distribution. The hybrid revealed excitation and emission bands at 340 and 408 nm, respectively. Accordingly, nitrogen doped graphene quantum dots/Ag hybrid was found efficient as a photocatalyst for H_2O_2 induced florescence for uric acid detection.

In this way, we analyzed effectiveness of few important designs of metal nanoparticle (Ag, Au, Pt, Cu, Fe, etc.) functional graphene quantum dots in the fields of energy devices, sensors/electronics, catalysis, and biomedical sectors [39-41].

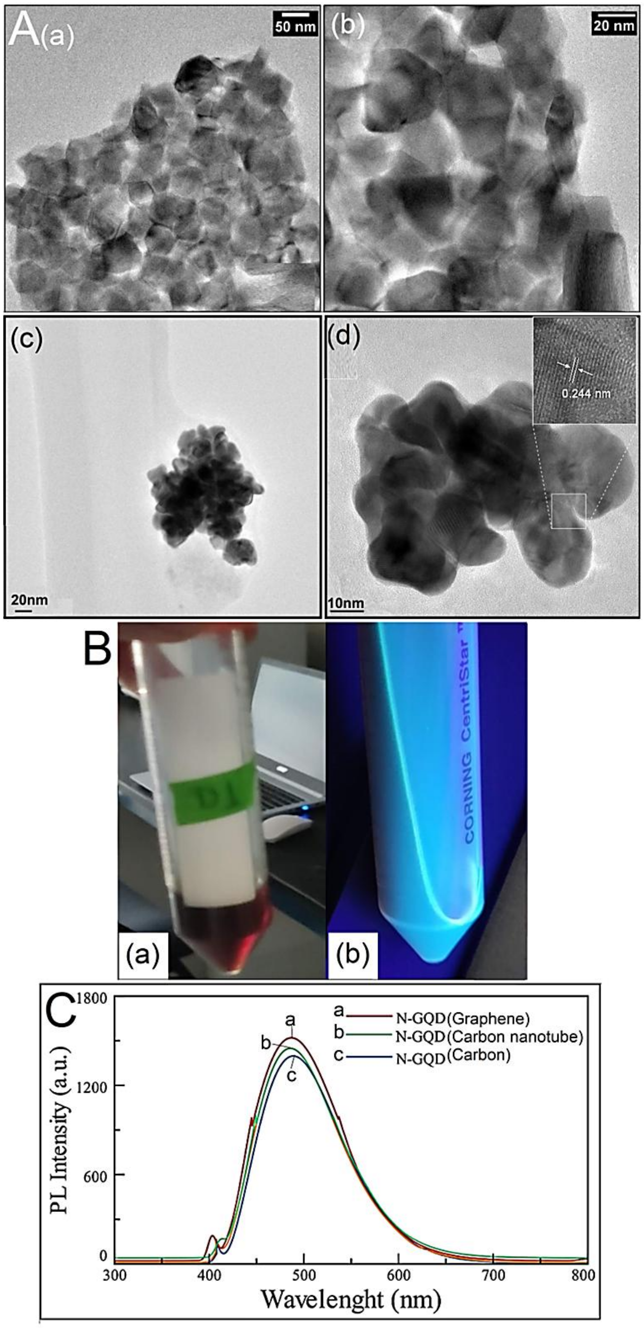

Figure 1 (A) Transmission electron microscopy images of (a,b) graphene quantum dots obtained from graphene; (c,d) nitrogen doped graphene quantum dots/gold nanoparticle hybrid, inset: Au nanoparticle with lattice spacing; (B) graphene quantum dots obtained from graphene via hydrothermal method: (a) Daylight image with brown reddish color; (b) florescence in the same; (C) photoluminescence (PL) of graphene quantum dots obtained from different nanocarbons hydrothermally [37]. Reproduced with permission from MDPI.

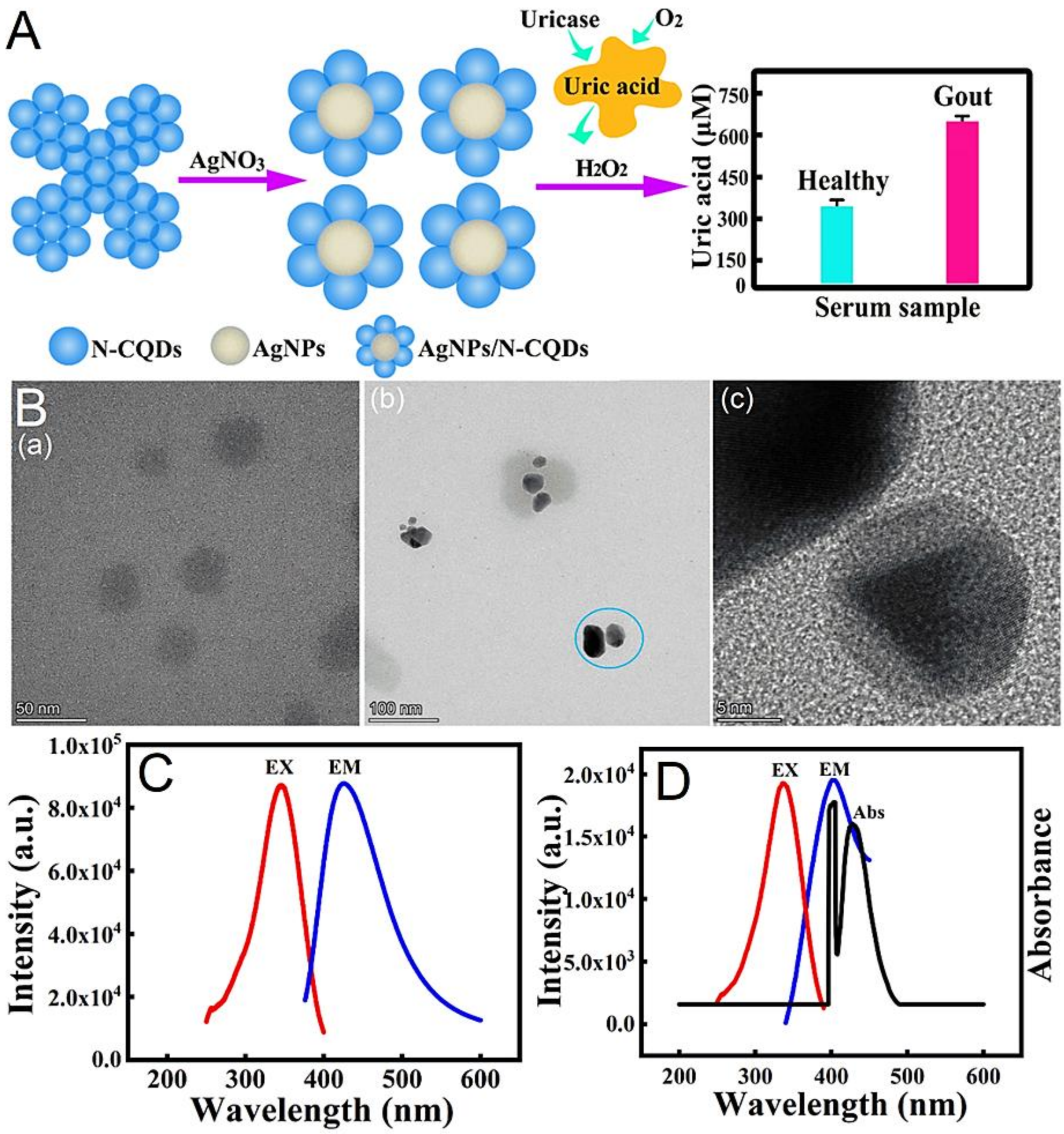

Figure 2 (A) Schematic illustration of the preparation of silver nanoparticle modified nitrogen doped graphene quantum dots (AgNPs/N-CQDs) used for uric acid detection; (B) transmission electron microscopy image of: (a) Nitrogen doped graphene quantum dots (N-CQDs); and (b) AgNPs/N-CQDs; (c) high resolution transmission electron microscopy image of AgNPs/N-CQDs; (C) florescence excitation and emission spectra of N-CQDs; (D) florescence excitation and emission spectra and UV absorption for AgNPs/N-CQDs [38]. Reproduced with permission from MDPI.

Combination of graphene quantum dot with metal oxide/inorganic nanoadditives

As mentioned above, modification of graphene quantum dots using metal nanoentities showed advantageous enhancements in electronic, electrical, thermal, catalytic, and biocompatibility characteristics of the resulting graphene quantum/inorganic nanoparticle hybrids [42,43]. Besides metal nanoparticles, hybrids of metal oxides and graphene quantum dots have been designed and investigated for their physical or covalent interactions [44-46]. Metal oxides (crystalline solids) nanoparticles usually consist of metal cations and oxide anions [47]. Pristine metal oxides as well as metal oxide hybrids have been manufactured via number of efficient synthesis techniques, including in situ, solution, sol gel, template, and hydrothermal approaches [48]. Like metal nanoparticles, metal oxides have also been frequently studied in the literature for their significant physical properties and myriad of technical applications, ranging from solar cells and sensors to drug delivery and other biomedical areas [49]. Accordingly, graphene quantum dots/metal oxide nanoparticles hybrids have been explored for photovoltaic, catalytic, water splitting, environmental, and other technological applications [50-52]. In this concern, graphene quantum dots/metal oxide hybrids seemed to develop promising heterointerfaces responsible for their advanced applications in ecological photoelectrochemical energy production devices, such as fuel cells, perovskite solar cells, etc. [53-55]. Among inorganic metal oxide nanoparticles, Sudhagar et. al. [56] designed hybrids of titania (TiO_2) with graphene quantum dots. For this, hollow nanowires of TiO_2 (100 - 200 nm) were developed and decorated with graphene quantum dots on their surfaces. The ensuing graphene quantum dots/TiO_2 nanowire hybrids had superior photocharge carrying properties and 70 % higher photocurrent, than pristine nanowires. These results were attributed to mutual structure-property associations of graphene quantum dots and TiO_2 nanowires, thereby leading to advanced photocharge transfer characteristics. Raghavan *et al.* [57] manufactured graphene quantum dots and TiO_2 based hybrids via facile sonication and hydrothermal techniques, as shown in **Figure 3(A)**.

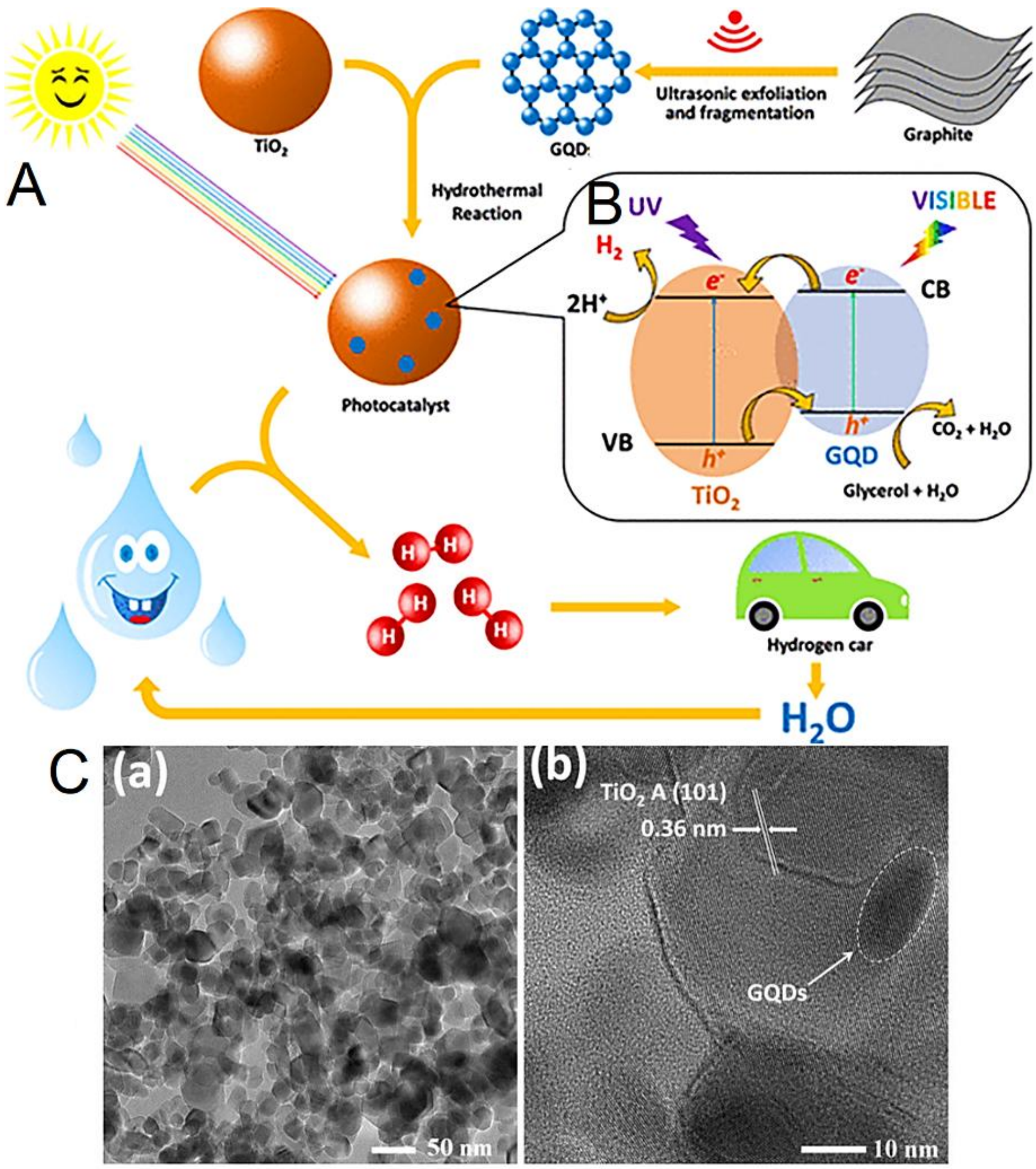

Figure 3 (A) Scheme for preparation of graphene quantum dots (GQD) and titania (TiO_2) based photocatalyst for H_2 production; (B) sensitizing effect and cocatalytic role of GQD deposited on TiO_2 for improved photocatalytic H_2 evolution under solar light, here VB = valance band; CB = conduction band; (C) (a,c) high resolution transmission electron microscopy images of graphene quantum dots/titania hybrids [57]. Reproduced with permission from ACS.

The 5 - 20 wt.% TiO_2 nanoparticles were used to form hybrid with graphene quantum dots. **Figure 3(B)** shows visible light and UV light irradiation based studies to explore photosensitization effects of these hybrids. Due to band gap of 2.26 eV and electron/charge transference, graphene quantum dots were found efficient to enhance photosensitization and photocatalytic activity of TiO_2 nanoparticles in these nanohybrids. Here, photocatalytic activity of TiO_2 seemed to be promoted by quantum dots through inducing recombination/parting of charge carriers. According to high resolution transmission electron microscopy micrographs in **Figures 3C(a)** and **3C(b)**, tiny TiO_2 nanoparticles (~ 20 nm) can be seen dispersed on graphene quantum dots surfaces. Accordingly, these inorganic nanoparticles had lattice plane and spacing of (101) and 0.36 nm, respectively. In this way, graphene quantum dots/TiO_2 hybrids were designed as environmentally benign photocatalysts for H_2 production.

Badrigilan *et al.* [58] established a hybrid combination of graphene quantum dots with iron oxide and bismuth oxide nanoparticles. They used facile solution route for the formation of resulting graphene quantum dots/iron/bismuth oxides nanocomposites. Technically, these hybrids were found effective for photothermal therapy showing > 50 % killing efficiency for malignant cells. Xi *et al.* [59] designed unique hollow nanocomposite nanospheres of nitrogen doped graphene quantum dots, carbon, and palladium (Pd) nanoparticles. For hybrid nanoparticle fabrication, initially silica nanoparticle functional graphene quantum dots were prepared and subsequently converted to polydopamine functional silica/graphene quantum dots nanostructures (**Figure 4(A)**). Next, Pd nanoparticles were in situ synthesized on silica/graphene quantum dots/polydopamine nanohybrid surfaces. Further annealing and processing led to the formation of hollow nitrogen doped graphene quantum dots/carbon/Pd nanospheres. **Figure 4(B)** shows transmission electron microscopy micrographs of hollow nanocomposite nanoparticles of nitrogen doped graphene quantum dots/carbon/Pd hybrids. Tiny Pd nanoparticles (< 3 nm in size) can be seen uniformly dispersed on hollow nitrogen doped graphene quantum dots (10 - 30 nm average sizes) surfaces. Such Pd nanoparticle functional nanodots revealed catalytic properties beneficial for H_2O_2 sensing purposes. **Figure 4(C)** shows a view on biosensing performance of nitrogen doped graphene quantum dot and Pd derived hollow nanospheres. The H_2O_2 sensing properties of finally obtained polydopamine/silica/graphene quantum dot-Pd nanoparticle nanostructures were examined for malignant cell treatment.

Hence, different combinations of graphene quantum dots with metal oxide nanoparticles (silica, titania, iron oxide, bismuth oxide, etc.) have been reported in the literature so far. Depending upon the type of metal oxide nanoparticles used and synergistic effects between graphene quantum dots-metal oxide hybrid nanoparticles, valuable applications in the fields of energy, catalysis, and biomedical sectors have been discovered by the field scientists till date.

Hybrids of graphene quantum dot and MOF

Metal organic framework or MOF constitutes an important category of porous hybrid nanostructures having metal ions connected via organic linkers and effective bonding [60]. In spite of technically desirable high surface area and porosity features, MOF nanoparticles had limited temperature stability under extreme pyrolysis conditions [61]. Therefore, to enhance practical applications of MOF, its functional nanocomposites have been designed using a number of inorganic (metal/metal oxide) and carbon nanoparticles [62-64]. Nevertheless, formation of high end MOF hybrids often faces challenges of structural compatibility and facile processing [65-67]. Noteworthy applications of pristine MOF as well as derived nanomaterials have been observed in the fields of electronics, energy maneuvers, medical, and environmental systems [68]. Accordingly, we found research attempts regarding technical designs of MOF hybrids with graphene/graphene oxide [69,70]. Subsequently, graphene quantum dots (as unique graphene derivatives) have been composited with MOF nanoparticles to investigate their structural-property synergies and related unique physical properties [71,72].

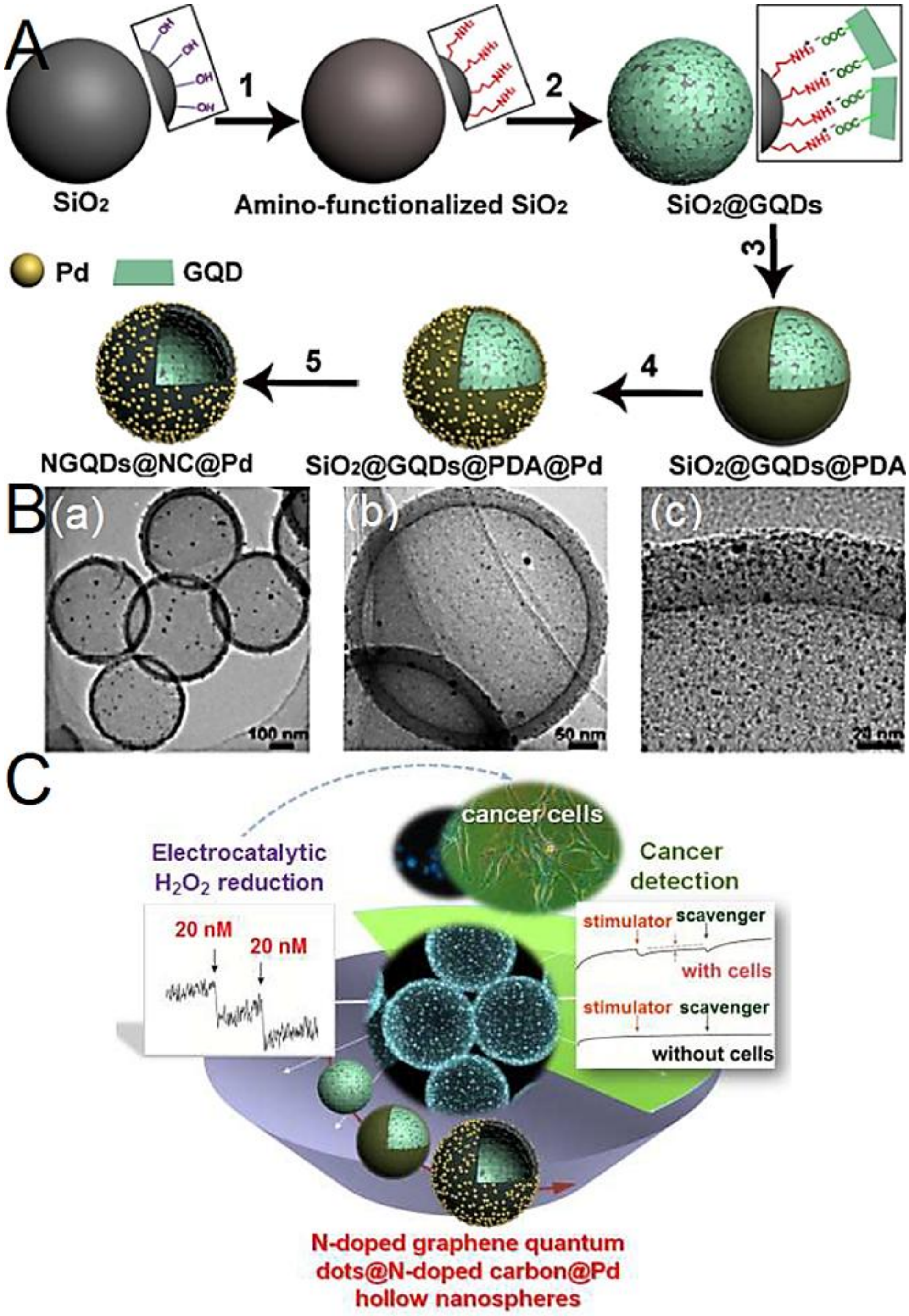

Figure 4 (A) Formation of NGQDs@NC@Pd HNSs; (1) amino-functionalization of SiO_2 nanospheres; (2) SiO_2 nanospheres wrapping with GQD nanosheets; (3) PDA coating on SiO2@GQD surface; (4) Pd NPs loading on SiO_2@GQD@PDA surface; (5) NGQD@NC@Pd HNSs formation by carbonization and hydrogen fluoride (HF) etching; (B) (a-c) transmission electron microscopy images of NGQDs@NC@Pd HNSs at different magnifications; (C) biosensing prospects of Pd nanoparticles decorated N doped graphene quantum dots [59]. Pd = Palladium; NPs = nanoparticles; GQD = graphene quantum dots; NGQD@NC@Pd HNSs = nitrogen doped graphene quantum dots@nitrogen doped carbon@Palladium/hollow structured nanospheres; PDA = polydopamine; SiO_2@GQD = silica@graphene quantum dots; SiO_2@GQD@PDA = silica@graphene quantum dots@polydopamine; SiO_2@GQD@PDA@Pd = silica@graphene quantum dots@polydopamine@Palladium. Reproduced with permission from ACS.

Xu *et al.* [73] adopted a unique method for the formation of nitrogen doped graphene quantum dots using zinc (Zn) based MOF or zeolitic imidazolate framework. For this purpose, they first converted zeolitic imidazolate framework into carbonized zeolitic imidazolate framework as a carbon source/precursor for the formation of nanodots. Later, nitrogen doped graphene quantum dots were prepared via solution, autoclave, and sonication techniques (**Figure 5(A)**). **Figure 5(B)** shows transmission electron microscopy and high resolution transmission electron microscopy images for the resulting graphene quantum dots. The tiny nanoparticles had small diameter of ~ 2 nm showing the success of synthesis method used. In addition, nitrogen doped graphene quantum dots own lattice spacing and plane of about 0.214 nm and (100), respectively. **Figure 5(C)** exhibits UV-VIS spectral studies on nitrogen doped graphene quantum dots with adsorption peak at 227 nm and blue shift at 200 nm due to π-π* transitions. In addition, weak/broad adsorption peaks (290 - 350 nm) were observed due to electron withdrawing properties of surface functionalities of nanodots.

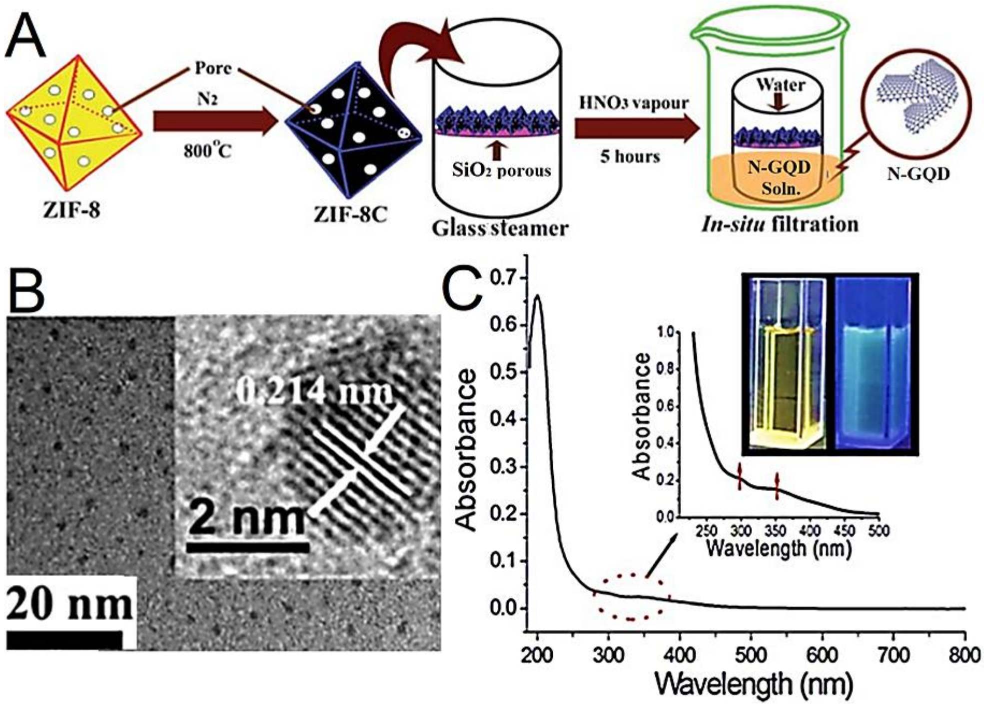

Figure 5 (A) Synthesis of nitrogen doped graphene quantum dots (N-GQDs) from zeolitic imidazolate framework (ZIF-8) and carbonized zeolitic imidazolate framework (ZIF-8C) as precursor; (B) transmission electron microscopy image of N-GQDs, inset: High resolution transmission electron microscopy image of graphitic layers of a single nanodot; and (C) UV-VIS absorption spectrum of N-GQDs dispersed in water, inset: Adsorption peaks at 290 and 350 nm and photographs of N-GQD solution IN: (Left) ambient and (right) UV light (365 nm) [73]. Reproduced with permission from RSC.

Tian *et al.* [74] formed a Zn based MOF, i.e., zeolitic imidazolate framework-8, zeolitic imidazolate framework-8/graphene quantum dots, and doxorubicin/ zeolitic imidazolate framework-8/graphene quantum dots nanocomposites using simple solution, sonication, and centrifugation techniques. **Figures 6(A) - 6(F)** show scanning electron microscopy and transmission electron microscopy studies on these nanoparticles. Here, zeolitic imidazolate framework-8 and its nanocomposites with graphene quantum dots had sizes ~ 50-100 nm. Conversion of zeolitic imidazolate framework-8 to zeolitic imidazolate framework-8/graphene quantum dots caused slight increase in nanoparticle sizes, however surface uniformity remained same both for pristine metal organic frame work and nanodot nanocomposites. **Figure 6(G)** illustrated thermogravimetry analysis thermograms showing slightly lower maximum degradation temperature for doxorubicin/zeolitic imidazolate framework-8/graphene quantum dots hybrids, relative to zeolitic imidazolate framework-8/graphene quantum dots, due to degradation of doxorubicin functionalities in structure. The ensuing doxorubicin/zeolitic imidazolate framework-8/graphene quantum dots were suggested useful for photothermal therapies in biomedical field.

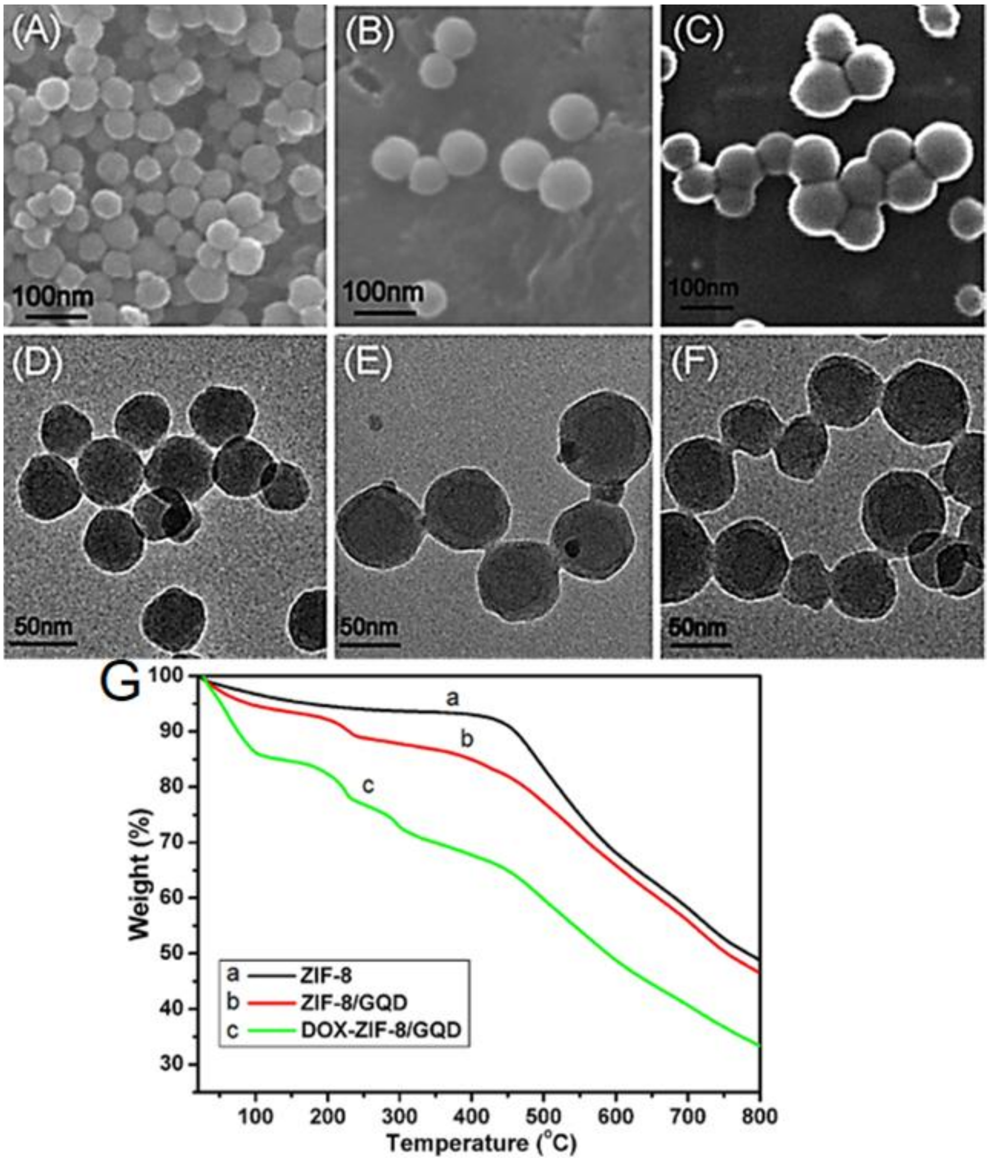

Figure 6 (A) Scanning electron microscopy image and transmission electron microscopy micrographs of: (A,D) ZIF-8; (B,E) ZIF-8/GQD; and (C,F) DOX-ZIF-8/GQD nanoparticles, respectively; (G) thermogravimetric analysis of ZIF-8, ZIF-8/GQD, and DOX-ZIF-8/GQD nanoparticles [74]. ZIF-8 = zeolitic imidazolate framework; ZIF-8/GQD = zeolitic imidazolate framework-8/graphene quantum dots; DOX-ZIF-8/GQD = doxorubicin/zeolitic imidazolate framework-8/graphene quantum dots. Reproduced with permission from ACS.

Tang *et al.* [75] prepared graphene quantum dots/Zn-MOF hybrids using solvothermal and solution techniques. These hybrids showed high specific surface area, of about 1,841 $m^2 g^{-1}$ and facilitated electron and charge transfer characteristics due to well interconnected 3 dimensional nanostructures. Accordingly, graphene quantum dots/Zn-MOF hybrids were further studied for application as high performance supercapacitor electrode showing superior specific capacitance and capacitance retention of around 200 F g^{-1} and 53 %, correspondingly. Yu *et al.* [76] designed graphene quantum dots and Zn based metal organic framework (zeolitic imidazolate framework-8) based nanomaterials. In this concern, zeolitic imidazolate framework-8/graphene quantum dots, carbon/zeolitic imidazolate framework-8, and carbon/zeolitic imidazolate framework-8/graphene quantum dots nanocomposites were synthesized by facile solution and carbonization techniques. The resultant nanoporous nanomaterials depicted superior surface area of ~ 668 $m^2 g^{-1}$. **Figure 7(A)** shows uniform surface morphology with hexagonal shaped (~100 nm) zeolitic imidazolate framework-8/graphene quantum dots hybrid nanoparticles. Development of unique microstructures pointed towards the effectiveness of synthesis techniques for self-assembly growth processes during hybrid nanoparticle fabrication. **Figures 7(B)** and **7(C)** depict nanostructures of carbon/zeolitic imidazolate framework-8, and carbon/zeolitic imidazolate framework-8/graphene quantum dots nanoparticles, respectively, formed after calcination. The annealing and carbonization seemed to develop rough surface morphologies of these nanoparticles. According to cycling stability performance measured by specific capacity and coulombic efficiency (**Figure 7(D)**), carbon/zeolitic imidazolate framework-8/graphene quantum dots had superior charge-discharge capacity of 400 - 800 mAh g^{-1} and columbic efficiency up to 100 %, as lithium battery electrodes. In addition, these hybrid electrodes showed prolonged cyclic stability in ~ 10,000 charge-discharge cycles. These resulted can be attributed to synergic effects of graphene quantum dots with porous zeolitic imidazolate framework-8; thereby enhancing overall electrochemical performance for battery electrode application.

Incidentally, high end graphene quantum dots/MOF hybrids have been detected competent for energy devices [77,78], catalysis/electronics [79,80], and biomedical [81,82], etc.

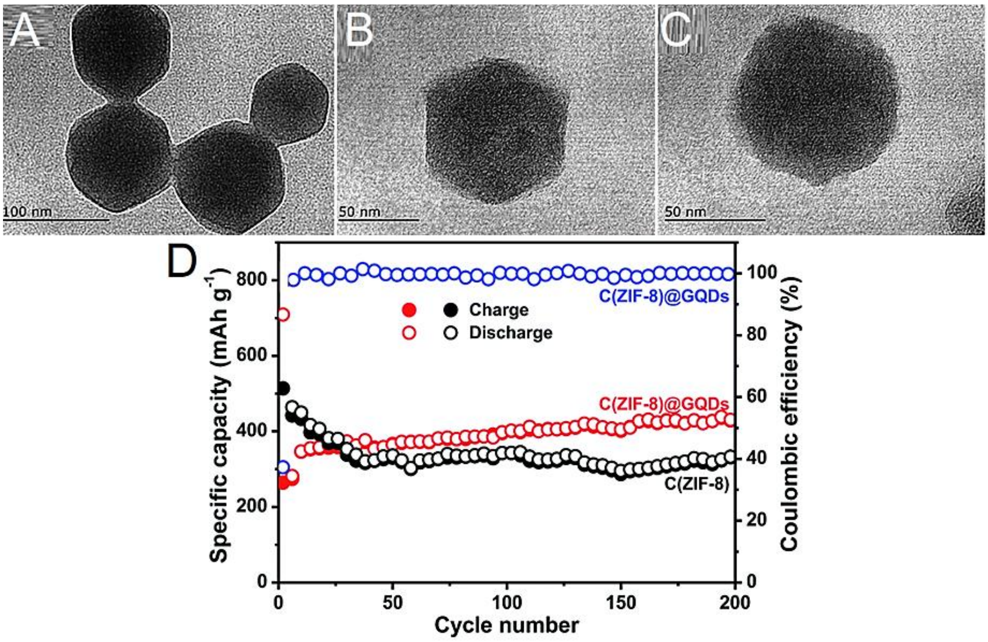

Figure 7 Transmission electron microscopy images of (A) ZIF-8@GQDs; (B) C(ZIF-8); (C) C(ZIF-8)@GQDs; and (D) cycling stability performance at 100 mA g^{-1} of C(ZIF-8)@GQDs based anode [76]. ZIF-8@GQDs = zeolitic imidazolate framework-8@graphene quantum dots; C(ZIF-8) = carbon/zeolitic imidazolate framework-8; C(ZIF-8)@GQDs = carbon/zeolitic imidazolate framework-8@graphene quantum dots. Reproduced with permission from RSC (Open access).

Prospects of graphene quantum dots/inorganic nanoparticle hybrids

Carbon based quantum dots and related nanomaterials have attracted increasing scientific interests due to quantum/surface/edge effects and physico-chemical properties; thereby leading to their high end uses in industrial level applications [83,84]. Particularly, graphene quantum dots (an important type of carbon nanodots), have been studied for myriad of applications in the fields of energy/electronics, engineering, and biomedical systems [84-86]. Among efficient techniques used for graphene quantum dots synthesis, hydrothermal method, chemical vapor deposition, microwave/plasma strategies, and chemical/ electrochemical approaches have been observed in the literature till date [87]. As discussed in above sections of this review, research progresses in the field of graphene quantum dots resulted in a number of advanced organic-inorganic hybrids of graphene quantum dots with metal, metal oxide, and MOF nanoparticles. Consequently, we noticed technical applications of graphene quantum dots hybrids as luminescent sensors for environmental and biomedical related uses [88,89]. Accordingly, **Figure 8** shows few important application areas of graphene quantum dot hybrids as luminescent devices [90]. For example, graphene quantum dots/metal nanoparticle hybrids have been designed as optical sensors and other optoelectronics [91]. Such sensors have been found efficient for detection and eradication of hazardous metal ion from environment (water or air media) [92,93]. Consequently, graphene quantum dots/metal nanoparticle hybrids based optical sensors depicted superior efficiency, selectivity, and sensitivity for toxic ion removal from water [94-96]. In addition, effectiveness of these sensors for trace metal ion detection has been analyzed through surface enhanced Raman spectroscopy (SERS) technique [97]. Henceforward, graphene quantum dots/inorganic nanoparticle hybrids derived SERS florescent sensors have been found competent for important environmental applications [98].

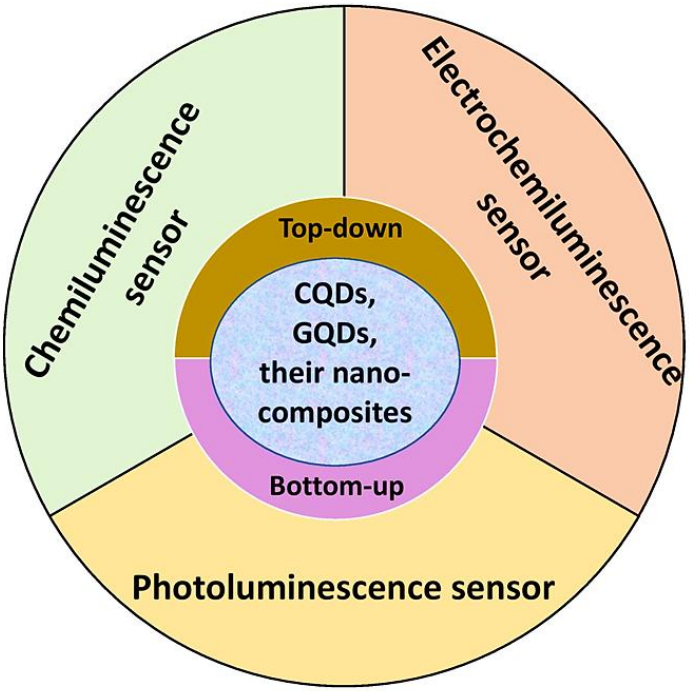

Figure 8 Graphene quantum dot and related nanocomposites in sensing technology [90]. Reproduced with permission from MDPI.

Another important application of graphene quantum dots/inorganic nanoparticle hybrids has been found in the field of capacitive devices [99]. For instance, Xiaoshan *et al.* [100] designed boron doped graphene quantum dot and inorganic Nb_9VO_{25} nanoparticles based supercapacitor electrodes. Here, Nb_9VO_{25} (an inorganic nanomaterial) was formed by coordination of Nb(V) and V(V). Combining boron doped graphene quantum dot with Nb(IV)/V(IV) developed efficient electron transfer paths and narrowed bandgap for rapid charge/electron flow through the hybrid nanostructure. In addition, graphene quantum dot/Nb_9VO_{25} hybrid had tunable nanostructure, doping, conductivity, and capacitance properties. The resulting supercapacitor showed high energy density of ~ 77 – 146 W h kg^{-1}, specific capacitance of ~ 138 - 263 F g^{-1}, and > 95 % capacitance retention over 10,000 cyclic recitals.

In biomedical sectors, graphene quantum dots/metal nanoparticle hybrids have been used for wound healing purposes [101-103]. In this concern, Norouzi *et al.* [104] fabricated graphene quantum dot-TiO_2-graphene oxide hybrid mats for wound dressing application though an electrospinning technique. These nanomaterials had optimum water resistance and biodegradability properties for safe wound healing applications. Moreover, water contact angle studies on graphene quantum dot-TiO_2-graphene oxide hybrid mats depicted hydrophilicity nature for better wound treatment. In addition, mechanical stability studies for effective prolonged use of graphene quantum dot-TiO_2-graphene oxide hybrids demonstrated superior strength and flexibility properties due to their uniform and interracially compatible interlinked porous nanostructures. Hence, graphene quantum dot/TiO_2 hybrids were applied as efficient wound healers due to antibacterial activity against Gram-negative and Gram-positive bacterial strains.

Similar to metal and metal oxide nanoparticles, outstanding applications of graphene quantum dot/MOF hybrids have been reported for lithium ion batteries [105], supercapacitors [106], radiation shielding [107], environmental purposes [108], and biomedical sectors, such as drug delivery [109]. For example, Wei *et al.* [110] designed nanocomposite of graphene quantum dots and zinc-MOF for photocatalytic applications. The resulting graphene quantum dot/Zn-MOF hybrid was investigated as a photocatalyst for carbon dioxide CO_2 conversion. These hybrids exhibited superior photocatalytic CO_2 conversion efficiency due to rapid electron-hole pair generation. Hence, environmental remediation application of graphene quantum dot/MOF hybrid has been well established in the literature.

Conclusions and views

Conclusively, this revolutionary review highlights designs, fabrication, properties, and applied aspects of graphene quantum dots and their hybrids with inorganic nanoparticles, like metal, metal oxide, and MOF. Combination of graphene quantum dot with inorganic nanoparticles developed compatible interfaces and mutual interactions; thereby enhancing microstructural, electrical/charge conductivity, florescence, catalytic/ photocatalytic, ion recognition, and antimicrobial effects. Accordingly, we observed applications of graphene quantum dots/inorganic nanoparticles hybrids in the fields of sensors, energy devices, and biomedical fields. Nevertheless, as per literature availability so far, we can say that research and progress on these multifunctional hybrid materials are still in primary stages. Hence, we suggest comprehensive and focused scientific investigations to form high performance graphene quantum dot/inorganic nanomaterials for future industrial scale applications for optoelectronics (sensors), energy devices (solar cells, batteries and supercapacitors), environmental remediation, and biomedical (antimicrobials and drug transfusion) fields.

Henceforth, fundamentals and technological potential of graphene quantum dots/inorganic nanomaterials have been argued in this manuscript. Nevertheless, literature up till now include random research reports on these technically viable hybrids and no systematic studies conducted so far towards commercial level deployments. As per our analysis, systematic fabrication of these hybrids using predefined and optimum synthesis conditions seems indispensable for their commercial scale applications. For that reason, concerned field researchers must perform in-depth research investigations on graphene quantum dots based hybrids to overcome challenges of possible toxicity, impurities, uncontrolled nanoparticle sizes, design reproducibility, low quantum yields, low photoluminescence, aggregation, low electron/charge conductivity, and nanodot-inorganic nanoparticle synergies; thereby enhancing their commercial scalability for future devices and environment related industries. In this regard, we suggest use of green synthesis approaches and ecological or biodegradable precursors, and nontoxic solvents and reagents to reduce greenhouse emissions and related environmental burden. Furthermore, graphene nanodots and derived hybrids need to be comprehensively investigated for their cradle to grave life cycle and influence on our ecosystem.

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