



## REVIEW

### Comparative Assessment between Glowing Family Elements of Metal and Graphene Quantum Dots Based on their Properties: A Theoretical and Experimental Study

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The quantum dots (QDs) have wide range of applications in the field of biology and energy due to exceptionally photo-physical properties and highly active larger surface area. Both metal quantum dots (MQDs) and graphene quantum dots (GQDs) have created a new platform for quantum chemistry not only theoretical point of view but also open various real applications. The photo-physical properties arise due to tunable band gap and active functional groups adhere to the surface. Both MQDs and GQDs have very good properties such as solubility in wide range of solvent, good stability in photo-physical and chemical environment, less poisoning in animal body, small size and tunable photoluminescence is favourable for cell dynamics and imaging, up-conversion and down conversion photoluminescence nature, good electrochemical activity. The functionalization of QDs with micro and macromolecules in all direction are possible as results we find different shape of QDs due to very small size of QDs just like few atoms. Some theoretical studies have been observed to evaluate the origin of these properties. This review focused on the comparative studies including advantage and disadvantage between GQDs and MQDs based on their properties and also give significant imminent to motivate more encouraging development. Moreover, this review study offers significant insights for enhancing the characteristics of quantum dots.

**Keywords:** Graphene quantum dots, Metal quantum dots, Optical properties, Electrochemical properties.

## INTRODUCTION

Nanomaterials are a cause for concern since they have the potential to bridge the gap between the microscopic and macroscopic states of molecules, which might lead to exciting new possibilities in fields like biomedical research and optoelectronics. Past few years, research aims to center on many important properties of nanostructured semiconductors like optoelectronic as generated from size distribution in nanoscale. Nanostructure materials, which are characterized by differences in optical and electrical properties caused by variations in particle size  $\sim 1$ -100 nm, are categorized as zero, one and two-dimensional objects, such as quantum dots, quantum wires and thin films, respectively. Quantum dots (QDs) are zero dimensional nanostructure; it has few numbers of electrons as results in isolated energies level was observed in the density of states (DOS) [1]. In recent years, significant advancements have been observed in quantum dot research since the first isolation of this astonishing

material. Quantum dots (QDs) have been altering the scenery of many fields in science and technology; particularly electronics, energy storage and conversion and biomedical research [2]. Significant challenges persist in revealing the complete potential of quantum dots and their derivatives.

Metal quantum dots (MQDs) hold excellent assets, which are in between cluster material and single molecule. MQDs have quantum confinement effect, which can help to tune the excitation and emission wavelength. Also, MQDs can be generated characteristic emitted frequency by hitting external photon on MQDs surface [3]. Additionally, separation of the electron-hole pair of MQDs increases with the particles size decrease, hence, MQDs have higher photocatalytic activity than its bulk. Zero dimensional MQDs can be applied in multidimensional fields, such as catalysts, solar cell, solid-state device, electro-optical devices and biological labeling due to the size tunable absorption and emission properties. Due to their exceptional steady photoluminescence property, MQDs can be considered

a possible replacement material [4-7]. But problems rise due to MQDs are toxic and poorly soluble in water [8]. In addition, MQDs have large crystal size than a biomolecule, which might be create negative effect on cell dynamics for target molecules and easily form imitation cluster into biomolecular studies [4]. Synthesis of graphene quantum dots (GQDs) is another powerful and attractive tool for the changing the band gap and generated the photoluminescence properties of graphene [9]. GQDs have distinct edge effect and powerful quantum confinement effect than graphene based material. This nature of GQDs has created new interesting optoelectronic phenomena that might be difficult to observe in MQDs. The GQDs have several attractive merits, such as high solubility, low cytotoxicity and excellent biocompatibility, large surface area, facile to bind with biological molecule, tunable band gap, which help to use in photovoltaic devices, outstanding strength in presence of light, insignificant dimension *i.e.* zero dimension, less poisons effect in animals body, changing optical property, excellent multi-photon excitation, electrochemical property in presence of light [9]. This review emphasizes a recent report on the comparative assessment of GQDs and MQDs and seeks to provide important recommendations for highlighting their advantages.

**Structures:** Metal quantum dots (MQDs) are mostly synthesized from transition elements and also selenium, sulphur. The number of atoms in QDs in the range from 200 to 10000 and diameter in the range of 2 to 20 nm creates it neither a bulk solid structure nor a single molecule, so it might be called as artificial atoms. QDs are so called since they capability of quan-

tum confinement *i.e.* electron wavefunction is restricted with the particle diameter. Because of this, band gap energy of QDs is subject to particle diameter. Particle size of QDs has play vital role on biological activity, several experiment established that particle diameter can manipulate the poisoning level of QDs at the intracellular testing. In biological studies, the small size  $\sim 2.2$  nm of CdTe QDs has more toxic than  $\sim 5.2$  nm [10]. QDs are a mixed cluster of elements and they change in their structure and properties that depend on each component. They can modify their core and shell structure as a result morphology, diameter and surface behaviour will be changes. So, it is impossible to give common statement on their toxicity. QDs surface can have hydrophobic ligands or different hydrophilic moieties that make them soluble in the organic or aqueous solvent [11]. The most traditionally studied QDs are CdS, CdSe, CdTe, PbSe, GaAs, GaN, InP, InAs and CdSe core surrounded by a ZnS shell. The most familiar QDs are PbS, CdSe and ZnSe and also many other forms of these particles that contain other elements as well. QDs can also be fabricated by three elements like InGaAs or just single element as silicon.

GQDs are structurally distinct and particles size is below 10 nm [12] and have crystalline or amorphous structure with  $sp^2$  hybridized carbon network and it shows layer structure like graphene [13]. Although the diameter of GQDs is nanometer scale but Liu *et al.* [14] reported  $\sim 60$  nm lateral dimension of GQDs. The shape of most of the GQDs is circular but triangular, hexagonal and quadrate is also available (Fig. 1) [15]. The XRD and HRTEM measurements show that GQDs

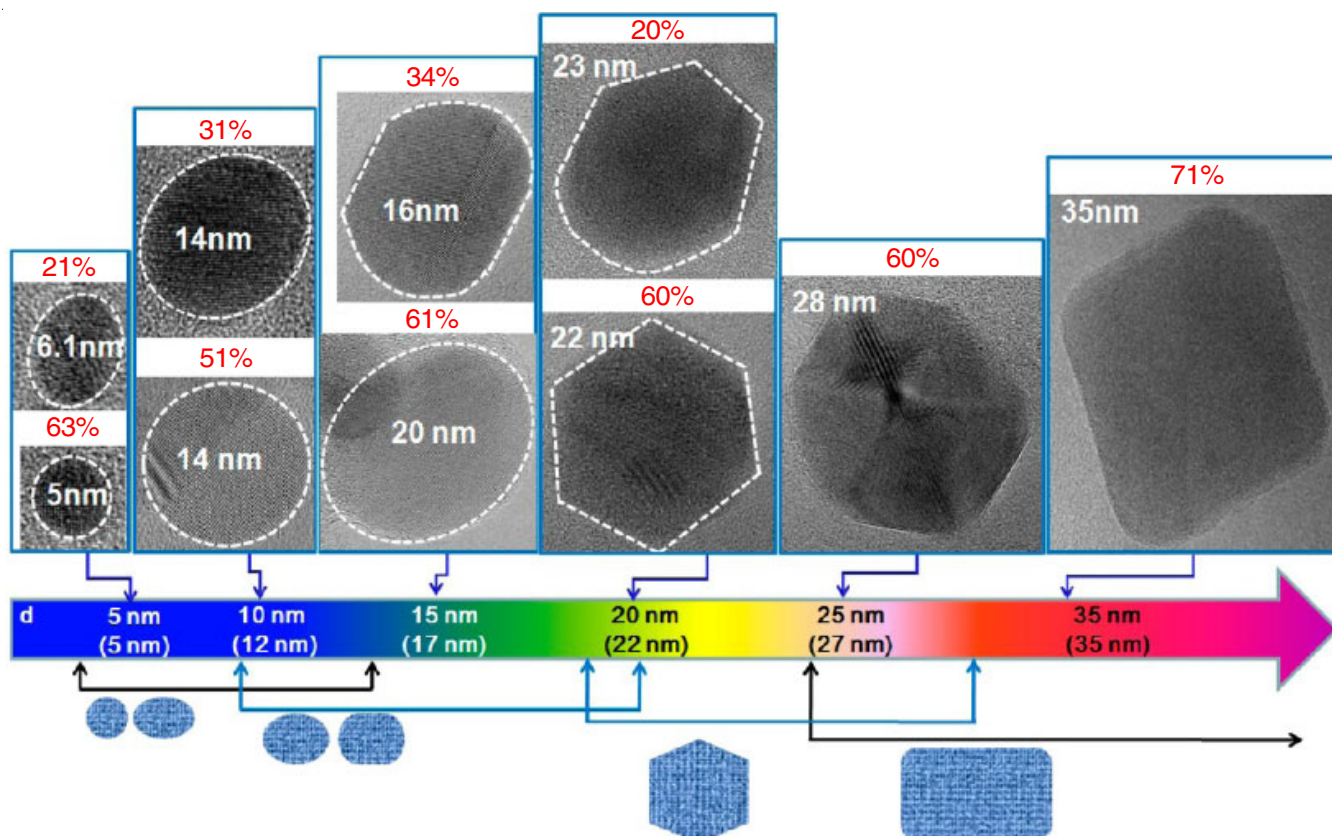


Fig. 1. HRTEM images show different shapes of GQDs. The arrows shows the average size in which GQDs has major morphology are observed. Reprinted with permission from Ref. [15]

has graphitic structure and inplane lattice spacing of 0.18-0.24 nm is lesser compare to graphene interlayer spacing of 0.334 nm due to the presence of different functional groups.

### Optical properties

**Absorbance:** The UV-visible absorbance spectrum (UVAS) of inorganic QDs can predict about the size, geometry, intensity and peak position changes with size of the QDs (Fig. 2a) [16]. The peak area of UV-vis spectra can provide an evidence for dispersity of the QDs sample. The absorbance spectrum varies by the aggregation of the QDs due to change the particle size and show red shift with decrease the intensity in the UV-vis spectrum. The GQDs possess inhomogeneity size distribution as a result existence of distinct energy levels and corresponding changes the optical properties of the semiconductor QDs. One of the most valuable properties of noble metal QDs is localized surface plasmon resonance (LSPR) and observe when certain frequency of photons can induce the group oscillation of conduction electrons on the QDs surface and this brings discriminatory photon capture. Silver quantum dots provide more details regarding the properties [17].

In general, GQDs has UVAS peak at low wavelength region due to  $\pi$ - $\pi^*$  transition of C=C bonds. GQDs characteristically appearance strong UV-visible absorbance peak in the wavelength range of 260-320 nm. GQDs shows a small peak in the wavelength range of 270-390 nm due to non-bonding to  $\pi^*$ -transition of C=O bonds. The chemical bonding with the different moieties or physical absorption can tune the UV-visible absorbance (Fig. 2b) [4,18].

**Photoluminescence:** Inorganic QDs has strong bright photoluminescence property than other molecules such as dyes, biological proteins and carbon quantum dots due to their large surface area, excellent photo-stability and possess symmetric emission with less widen. QDs are characterized by size dependent characteristics [19,20]. Recently core-shell quantum dots [21] have superior properties and applications than single spherical QDs due to shell shields the core structure from degradation as a result good photo stability and wide bandgap. Also quantum rods [22], doped QDs [23], mixed QDs [24] have been developed for getting a prominent photoluminescence properties over pure spherical QDs due to shape, doping environments and synergistic effect. Composition inside the QDs can play a vital role for tuning the optical properties like quantum confinement effect [25]. By controlling the size of the QDs from 2 to 8 nm, CdSe is able to emit at wavelengths ranging from 500 nm to 650 nm, making it a prominent QD for a variety of applications. Other QDs such as CdS, ZnSe, ZnS have activity in ultraviolet region and CdTe, CdSe<sub>x</sub>Te<sub>1-x</sub>, PbS emit in near-infrared and also PbSe has mid-infrared emission (Fig. 2c-d) [1,25].

The wider particle size distribution and incompatible experimental findings make it extremely difficult to have a clear understanding of the tunable photoluminescence capabilities of GQDs. Quantum confinement effects of GQDs can help to create most attractive feature of tunable photoluminescence properties which favour for many applications like biomedical and energy devices. The photoluminescence quantum yield (QY) of bare GQDs is lower compare to inorganic QDs due to the presence of different

functional group and defecting sides so; surface modification is required to enhance their bright emission. Different emission coloured of GQDs have been reported, ranging from blue to red and the maximum GQDs exhibit wide-range of emission spectra due to the large dispersity in diameter and the presence of different elements in GQDs skeleton (Fig. 2e) [18]. Chen group have reported a theoretical calculations for originating the emission of a GQDs by using density-functional theory (DFT) and time-dependent DFT. According to their report, the  $\pi$ -electrons in  $sp^2$  carbon network are responsible for originating the photoluminescence properties of GQDs and altered by its size, edge pattern, morphology, bonded chemical groups, doping environment and defects configuration (Fig. 2f) [26]. Jin *et al.* [27] reported that the functionalized -NH<sub>2</sub> groups can decrease the bandgap of GQDs for increasing negative charge density in GQDs. Moreover, Pan *et al.* [28] demonstrated that the photoluminescence property arises due to carbene like triplet state of GQDs. Thus, the GQDs have excellent photostability compare to organic fluorophores and can be modified by extent of oxidation. It is also found that the microwave synthesized GQDs show green photoluminescence but after reduction by NaBH<sub>4</sub>, GQDs changes its photoluminescence to blue with doubled quantum yield [29]. Polymer passivation on the surface of GQDs can also improve the photoluminescence property. Shen *et al.* [30] showed that synthesized PEG passivated GQDs has doubled quantum yield compare to bare GQDs. The chemical functionalized by different amine and thiol can enhanced photoluminescence property because the electron pushing groups generally increase quantum yield through avoiding of non-radiative recombination and wavelength shifting [31]. Beyond the enhanced quantum yield of controlling reduction, surface passivation and functionaliation GQDs but has some limitation for real life applications due to increase particle sizes. Doped and co-doped of heteroatom in GQDs surface is important for generation and significantly tune the photoluminescence properties of GQDs. Heteroatom such as nitrogen, oxygen, sulphur, phosphorous, boron, *etc.* are commonly doping in GQDs surface. Among the different heteroatom nitrogen is very easily doped in GQDs surface account for similar size with carbon atom and it is important for optical and catalytic properties due to larger charge separation. It is exhibited that N-doping on GQDs surface are pyridinic, pyrrolic and graphitic configuration. Pyridinic and graphitic N-doped GQDs has quantum yield 0.34 is higher than bare GQDs of 0.102. Generally, the quantum yield of N-doped GQDs are varies from 0.15 to 0.94, which is extremely sensitive to the precursor materials and experimental method [32]. Anh *et al.* [33] reported that N,S co-doped GQDs has quantum yield of 0.419.

**Up-conversion photoluminescence:** Up-conversion (UC) photoluminescence means emit shorter wavelength light from converted two or multiple low-energy photons, has great application in energy conversion and biomedical fields. The UC generally observed in rare earth metal containing metal nanoparticles or dye induced triplet-triplet annihilation (TTA) but first one, intensity of light during excitation is much higher than the solar flux and later TTA can work below the solar flux but main problem is low photostability of organic chromophore.



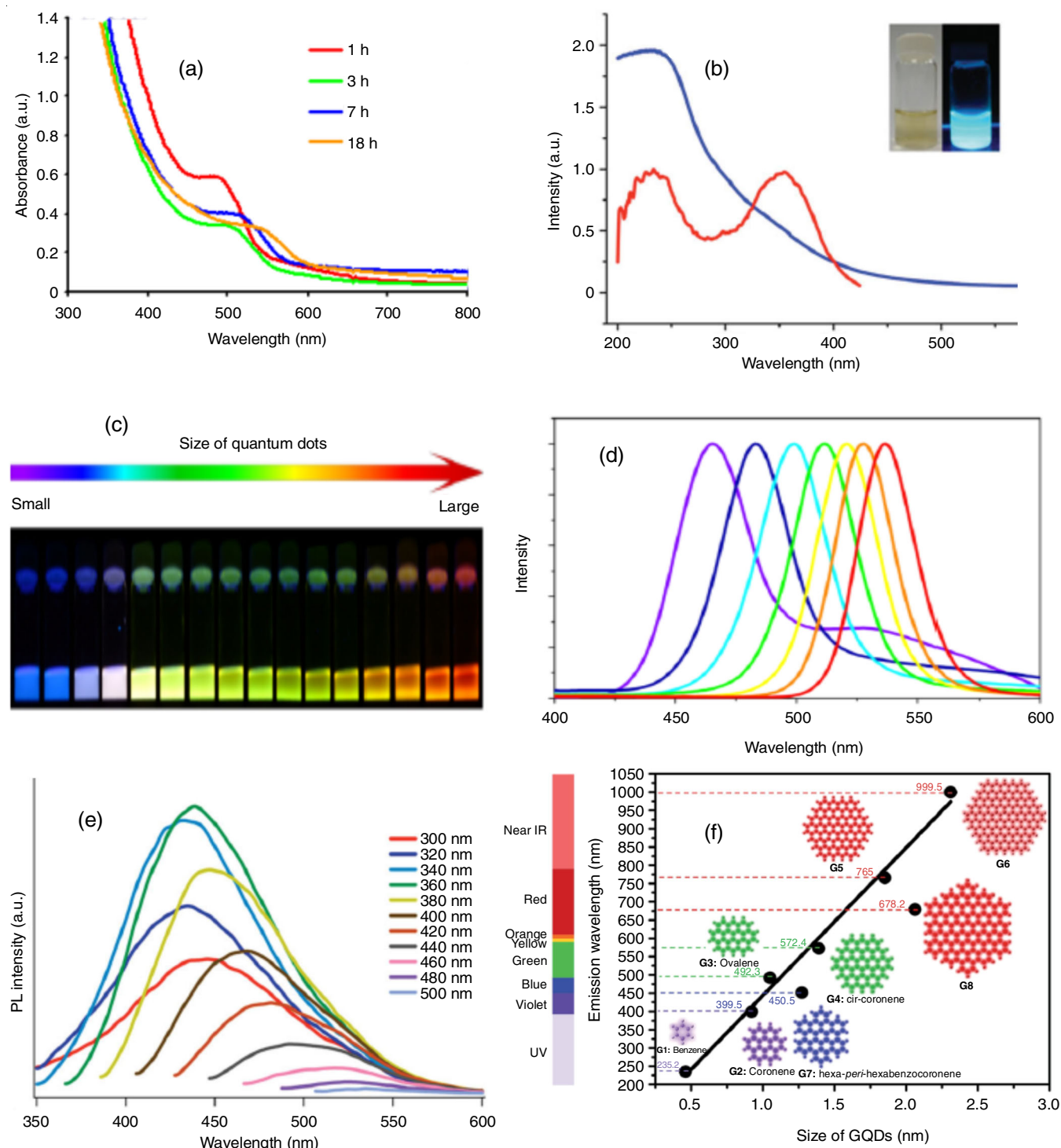


Fig. 2. (a) UVAS of thin films of the CdTe QDs synthesized at various times [16], (b) blue means UVAS and red for excitation spectra of GQDs and the inset shows the photographs of GQDs [18], (c) sixteen emission colours of CdSe QDs in the presence of near-ultraviolet [1], (d) photoluminescence spectra of CdSe QDs [1]. (e) Emission spectra of GQD at various excitation wavelengths [18], (f) Measured emission wavelength by TDDFT method in vacuum as a function of the size of GQDs [26]. Reprinted with permission from Ref. [1,16,18,26]

Semiconductor QDs can solve these limitations successfully. Okumura *et al.* [34] reported a TTA-UC system by surface modification of 4-(10-phenylanthracene-9-yl)pyridine CdSe/ZnS core-shell QD (csQD) used as sensitized and presence of excess emitter molecules of 9,10-diphenylanthracene. The TTA-UC

system shows up-converted photoluminescence at 433 nm under excitation wavelength at 532 nm is shown in Fig. 3a. A UC system was fabricated by phosphoric acid substituted anthracene ligand binding CdSe QDs with annihilator of 1,10-diphenylanthracene. The UC was observed by excitation of QDs at 488

nm and energy move to the outer side attached ligands to ligand centered at triplets state that transfer their overall energy to the annihilator and generating 430 nm fluorescence, as result quantum efficiencies of 17% [35]. Song *et al.* [36] revealed that the hydrothermally synthesized MoS<sub>2</sub> QDs showed up-conversion photoluminescence property at 480 nm emission under excitation wavelength of 780 nm (Fig. 3b). Generally, thermalization can suffers the efficiency of solar cells of low bandgap materials like silicon and transmission can reduce the efficiency for high bandgap materials such as CdTe. The triplet energy transfer (TET) from lead sulphide QDs to rubrene *via* UC process, observed that introduced of ZnS and CdS as shell in defect sides as a result of improvements of UC quantum yield till 700 and 325-folds. This enhancement is due to the decrease

the radiative and non-radiative rates compare to the core and widened absorption and photoluminescence linewidths recommend TET may occur from thermally accessible mid-gap defect states and dark excitonic states at the band-edge [37]. Tang *et al.* [38] proved the size factor of QD is very important criteria for modulate the UC quantum yield in PbS-tetracene-rubrene derivatives by reducing the QDs size from 3.5 to 2.9 nm of PbS and 3.2 to 2.5 nm of PbSe, where PbS, tetracene and rubrene acted as photo-sensitizer, mediator and emitter, respectively the comparative UC quantum yield is improved ~ 700 and 250 times. This result explained through reducing the QDs diameter, the entrap sides may create important factor due to rise in the dangling bonds and unpassivated surfaces, while the mid-gap states may become more available energetically.

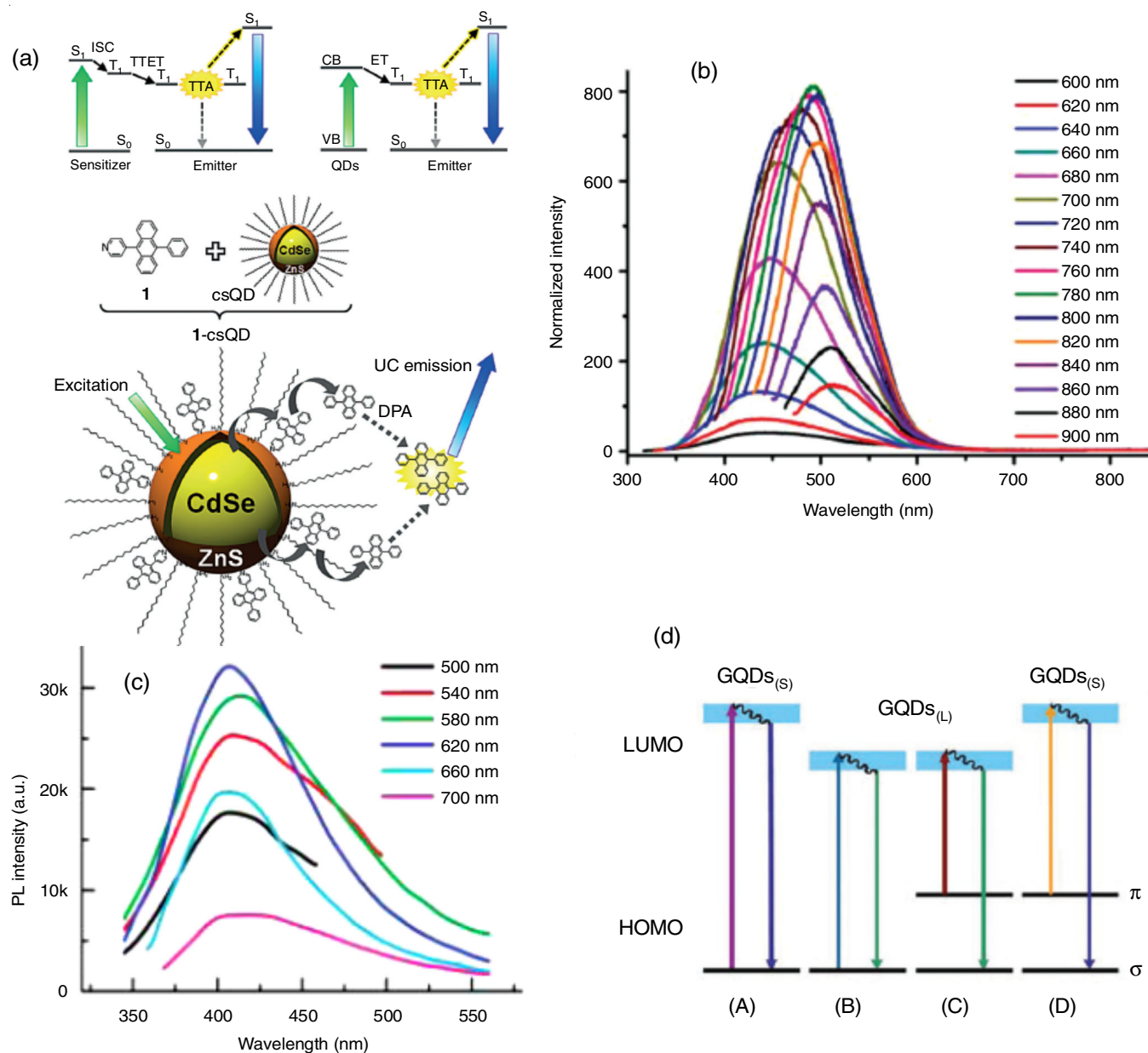


Fig. 3. (a) Schematic presentation of TTA-UC based on molecular sensitizers (top left) and QD sensitizers (top right), showing the involved energy levels [34]. (b) UC PL spectra of MoS<sub>2</sub> QDs and (c) UC emission of the GQDs with various excitation wavelengths [36,39]. (d) presentation of electron transitions diagram of GQDs. (A) regular fluorescence for small size GQDs and large size (B); UC PL for large size (C) and small size (D) [41]. Reprinted with permission from Ref. [34,36,39,41]

GQDs possess up-conversion photoluminescence properties which is favourable for cell imaging ability inside the body due to higher excitation wavelength. It is observed that GQDs have excitation-independent upconversion photoluminescence behaviour. The emission peak of GQDs at ~407 nm almost intact with the excitation wavelength varies from 500 to 700 nm (Fig. 3c) [39]. The upconverted emission of GQDs may be confirmed by multiphoton absorption process. Gong *et al.* [40] observed solvothermally prepared biocompatible nitrogen doped GQDs as capable for two-photon fluorescent. The two-photon absorption cross-section of nitrogen doped GQDs reaches 48 000 Goppert-Mayer units, which is more than dyes molecule and is comparable MQDs. Nitrogen doped GQDs can attain a depth in tissue penetration of 1800  $\mu\text{m}$  at excitation 800 nm. Shen *et al.* [41] recommended about the UC mechanism with constant energy difference of GQDs. It is familiar, for UC process the orbitals of carbon elements in GQDs must be carbene like ground-state multiplicity and corresponding energy separation between the  $\pi$ - and  $\sigma$ -orbital might be below 1.5 eV. According to their view, the energy separation between the excitation and emission light in the UC process was ~1.1 eV. They predicted that the GQDs exist more similar to the anti-stokes photoluminescence and it happen when a bunch of high wavelength light agitate the  $\pi$ -orbital electrons, the  $\pi$ -electrons jump to a higher energy excited state and then the electrons go back to a low-energy state after release extra energy in from of emission and it is called UC photoluminescence [41]. The experimental method must be carefully considered in order to determine the upconversion photoluminescence property of the GQDs sample. Wen *et al.* [42] reported the UC emission could originate from the usual photoluminescence excited by the leaking elements from the second diffraction in the monochromator of the photoluminescence spectrophotometer.

**Electrochemical property:** MQDs have strong electrochemical properties due to possess clear oxidation and reduction centre. The bandgap of MQDs is intensely affected on the surface defects and impurities, which can give some important information about quality of MQDs. The defect site can generate a new state and can play with electrons and holes, therefore changing the oxidizing and reducing centre. Like optical band gap, electrochemical band gap is important to find easily HOMO and LUMO position of MQDs and essential for devices applications. Generally, the electron transfer in a MQDs are various ways such as (i) electron injection into a MQDs, (ii) electron removal from a MQDs, (iii) concurrent addition of an electron and a hole in two non-interacting MQDs, and (iv) formation of an negative and positive charge cloud into the MQDs. The potential required for generating a non-interacting electron-hole pair of MQDs and gap between electron and hole is stated as the electrochemical band gap, this means potential gap of first reduction and oxidation processes in MQDs. The CdS QDs shows oxidation peaks at -2.15 V and corresponding reduction peaks 0.80 V in DMF electrolyte and CdSe has bandgap of 2.36 V with cathodic -1.56 V and anodic peaks at 0.80 V. It is also observed that like optical band gap, the electrochemical band gap of CdS and CdSe QDs decrease with enlarge the particle diameter. Not only particle size but also ligands may

control on the cathodic and anodic processes of the CdSe QDs [43-45]. Liu *et al.* [46] reported the similarity in the onset potentials at oxidation process with capping ligands coated in order of SA < PY < TDPA < OLA CdSe QDs (where SA = stearic acid, PY = pyridine, TDPA = tetradecylphosphonic acid, OLA = oleylamine). The ligand-dependent oxidation and reduction process of QDs can be ascribed due to the variation of the chemical environment and the steric hindrance effect generated by the ligand.

Pure GQDs has poor electrochemical activity due to no distinct redox centre but functionalized GQDs have electrochemical property because functional groups possess redox activity. Chen *et al.* [47] established functional group dependent band gap reduction of GQDs from 2.4 to 1.88 eV due to different extent of conjugation of functional group. GQDs size has important factor for tuning the bandgap, Ghamdi & Ghamdi [48] reported size dependent GQDs bandgap *i.e.* 1.9 nm, 3 nm and 5 nm GQDs shows bandgap 1.51 eV, 2.3 eV and 3.4 eV, respectively.

## Conclusion

Quantum dots (QDs) is an encouraging materials due to their various attractive properties including solubility in polar and non-polar solvent, wide rang bandgap, high surface area with good capability for surface functionality, excellent photoluminescence behaviour, electrochemical activity, *etc.* This review mainly focused on the comparative study between metal quantum dots (MQDs) and graphene quantum dots (GQDs) based on their properties. Some mechanisms in terms of photoluminescence enhancement, tuning bandgap, structure optimization, surface passivation, *etc.* are discussed. Both materials have excellent long-term photostability, which needs to be addressed soon for practical applications and GQDs have superior environmental stability than MQDs, but their chemical stability is opposite. GQDs have poor quantum yield with wide emission compared to the conventional fluorophores is a vital problem for future applications.

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## CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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