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Review

A brief review of features of copper indium disulphide ($CuInS_2$) nanomaterials for quantum dot solar cells

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ABSTRACT

Copper indium disulphide $CuInS_2$ (CIS) is found to be an interesting nanomaterial belong to group I-III-V for quantum dot solar cell (QDSCs) application due to low toxicity, multiple exciton generation effect, high light absorption in the visible spectral range, appropriate band gap that coordinate well with the solar spectrum, unusual radiation tolerance, noticeable defect tolerance and low cost. Properties of this material that makes it important for use in quantum dot solar cell is also discussed in this paper. This paper summarizes the research going on in the field of synthesis of $CuInS_2$ nanomaterials reported by different authors across the globe. Optical and photovoltaic properties of reviewed CIS QDSCs is also highlighted in this paper.

1. Introduction

Energy demand is continuously rising due to rapid growth of population. Energy supplied fulfil by conventional sources like fossil fuels are restricted and consumed rapidly. This situation forces us to search for other source of energy that are non- exhaustible. There are many types of non- exhaustible sources of energy like wind energy, solar energy, hydro energy, geothermal energy etc. Out of all these energy sources solar energy has come out as promising energy source. Solar cell which uses photovoltaic technology is found to be one of the ways by which solar energy can be converted to electrical energy. Initially this technology is mainly based on the material silicon but due to its high cost, researchers across the globe are vigorously working to produce different types of solar cells in particular dye sensitized solar cells and quantum dot solar cells (QDSCs) that have low fabrication cost and have capacity to achieve high efficiency. Semiconductor nanomaterials (CdS & CdSe) have been extensively used as active sensitizers in quantum dot-sensitized solar cells (QDSCs) as shown in Figure 1. This type of solar cell has acquired great research attention over the last few years due to unique features such as size dependent tunable energy band gap (quantum confinement effect) and multiple exciton generation effect. Theoretically QDSCs has an efficiency of greater than 44% but it is difficult to achieve it practically [1]. Till today, practically QDSCs have attained a power conversion efficiency of 18.1% [2]. However, majority of the reported QDSCs are using cadmium or lead compounds as sensitizer which is toxic material [3]. To bring QDSCs in commercial scale an affordable, stable and nonhazardous material is needed to produce solar cell modules. Nonpoisonous quantum dots (QDs) sensitizers are generally without lead and cadmium materials called as “eco-friendly” sensitizers.

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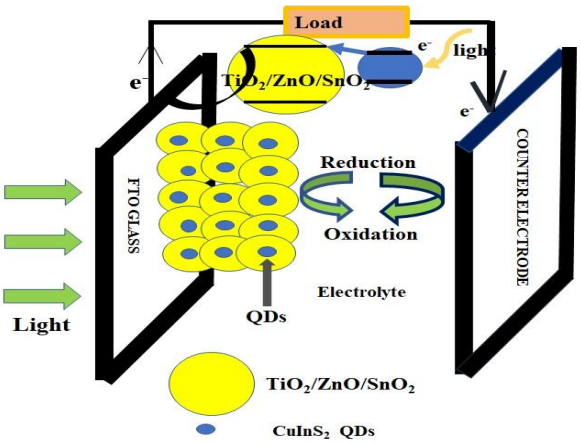

Fig 1.CIS quantum dot-sensitized solar cell

It has been established that the $CuInS_2$ (CIS) is an outstanding light absorbing materials because of its large absorption coefficient, has a bandgap of about 1.45~1.53eV [4], which matches with energy conversion range of the solar spectrum, size-tunable photoluminescence in the range 640nm to 850nm and possesses excellent defect tolerance [5]. In recent years, use of CIS QDs as sensitizers for fabrication of QDSCs has become popular because the Bohr exciton radius of this material is around 4.1 nm, and the quantum confinement can be seen until it attains 8 nm in diameter [6]. The objective of the present study is to highlight $CuInS_2$ a promising nano material for quantum dot solar cell due to its exceptional properties and to review the technologies, photovoltaic properties that has been used for application in QDSCs.

Typically, QDSCs is made up of three parts: Photoanode, a counter electrode and an electrolyte with redox couples. Photoanode is made from semiconducting nanostructured metal oxides (TiO_2 ZnO, SnO_2 etc) material painted on the surface of transparent conductive oxide (TCO) substrates mainly fluorine tin oxide (FTO) or indium tin oxide (ITO). Coated TCO is further sensitized with quantum dots (CdS, CdSe, CdTe, PbS, CIS) which forms photoanode of the QDSCs [7-8]. The counter electrode used mainly is platinum or Cu_2S coated FTO/ITO glass substrate [9] and working electrode is sulphide-polysulfide and iodide-triiodide [10].

As per the available data, reported values of efficiency of CIS QDSCs are in the range between 0.1 and 5% [11]. Differences in QDSCs efficiency occur because of the type of used electrolyte, counter electrode and the technique used to incorporate the sensitizer. In one of the studies, Photovoltaic conversion efficiency (PCE) reported utilizing CIS QDs doped with CdS is 1.42 % [12]. In Another study, reported PCE is 5.38 % using CIS QDs followed by subsequent layer of CdS doped with Mn^{2+} ions [13].

Presently, CIS QDs are not utilized in display device applications but employ as colour transform materials for manufacturing white LEDs and standard light source device due to broad full width half maximum (FWHM) and high emission intensity. This material is found to be very promising for applications in luminescent solar concentrators, QDs sensitized solar cells and solar photocatalytic applications [14]. CIS QDs are also used in biomedical applications for biological cell imaging [15] and as fluorescent probes to observe biological macromolecules in organisms & to detect heavy metal ions in aqueous solution. Recently, a particular class of solar cells that is acquiring recognition in the research group is perovskite solar cells because of large absorption coefficient in the visible region of solar spectrum and high efficiency [16-18]. Copper indium disulphide material also used in printed perovskite solar cells (PSCs) as an inorganic hole transport layer with achieved efficiency 9.9% [19]. However, with the use of copolymer-templated TiO_2, PSCs solar cell attain maximum efficiency 11.08% [20].

In quantum dot solar cell, CIS in quantum dot (QDs) form is usually preferred than polycrystalline thin films form as QDs can provide more structured charge partition by acting as sensitizers for large bandgap semiconductors such as TiO_2 in solar cell. The techniques used for nanoparticle synthesis can be divided into two class: molecular source decomposition (thermolysis, microwave irradiation, photolysis) and solution synthesis (hydrothermal and solvothermal). In general, molecular precursor decomposition build CIS quantum dots, while solution synthesis generates nanoparticles of sizes more than the quantization effect limit. The performance of QDSCs greatly depend upon the properties of used QDs which in turn depend upon synthesis technique used for their preparation. The key parameters that govern the applicability of prepared QDs is to precisely control the nucleation, the partition of photogenerated charges, followed by their move across the QD boundary. Molecular source decomposition method is direct but suffer from limitations such as sensitive to atmosphere, requirement of costly starting materials and involvement of bulky precursor synthesis steps. It has been reported CIS QDs prepared by solution method show poor optical properties, low quantum yield which can be enhance by passivating the surface with distinct ligands.

Looking to this aspect, in this review, we discuss the synthesis techniques that has been used to increase its photoluminescence quantum yield (PLQY) and photostability of CIS nanomaterial.

2. Review of methodologies used for synthesis of $CuInS_2$ nanomaterials

Many researchers across the globe are working to improve the performance of QDSCs using different QD sensitizing layer such as CdS, CdSe, PbS, CdTe etc. However, large toxicity associated with the use of Cd, Pd elements restricted the development of QDSC for large-scale application. Many Cd or Pb free quantum dots includes binary semiconductor (InP, In_2S_3) and ternary semiconductor ($CuInS_2$, $CuInSe_2$, $CuInTe_2$). Review of literature suggested that little work has been done to increase the performance of solar cell using $CuInS_2$ or core shell $CuInS_2$ QDs as sensitizing layer which is economical and non-toxic. QD layer is laid down on semiconductor coated electrode by in-situ or ex-situ techniques. Chemical bath deposition (CBD) and successive ionic layer adsorption and reaction (SILAR) are in-situ techniques [21]. In ex-situ methods, QDs are synthesized individually and further placed into semiconductor coated plate by molecular linker attachment, direct absorption and electrophoresis techniques [22]. Investigation of literature shows that many papers are published on synthesis of thin-film $CuInS_2$ but relatively less work has been done on the synthesis of colloidal $CuInS_2$ QDs as controlling defects and increasing crystallinity is difficult during QDs synthesis which is important for luminescence and photovoltaic applications.

Review of literature suggested that various approaches (organic and aqueous) have been used for $CuInS_2$ QDs formation. Methods in organic channel are thermal injection methods, one-pot methods, and template methods [14]. It has been reported that in organic method, solvents which lack affinity for water like 1-dodecanethiol, oleylamine (OLA) etc. were utilized to limit the cations reactivity (Cu and In) [23-24]. In thermal injection technique, nucleation occurs rapidly when cold solution was introduced into the hot solution of anionic and cationic precursors. This technique provides well command of the nucleation and ensure formation of monodisperse QDs [25]. In one-pot non-injection technique, precursor solution was amalgamated with organic solvent in presence of 1-Dodecanethiol (DDT function as both ligand and sulfur source) and heated above 220°C [26]. QDs having good crystallinity of narrower size distribution and having strong PL characteristics in the near-infrared region were obtained through template technique. In this method first binary sulfide nanocrystals is synthesized and then CIS nanomaterial procure through the method of cationic exchange reaction [27]. Organic methods relatively require high reaction temperature and produce hydrophobic CIS QDs dissolved in non-polar solvent only. Several approaches have been reported for converting hydrophobic QDs into the water loving phase which are respectively, ligand exchange, coating in lipids, encapsulation with amphiphilic polymers (amphipols), and silanization. These approaches are usually complex and produced material having poor emission characteristics, low water solubility and bad colloidal stability [28]. So, single step preparation of water-soluble QDs is preferred in the past several years. Synthesis using aqueous solution require the use of water-soluble capping agents like L-glutathione (GSH), 3-mercaptopropionic acid (MPA), L-cysteine, thiomalic acid and thioglycerol to control the reactivity of copper ion and indium ion. For the core synthesis of CIS QDs, more reactive sodium sulfide (Na_2S) and for the core shell synthesis of CIS QDs, less reactive thiourea was employed as a sulfur source in aqueous solution method.

The influence of stabilizing ligand on the structure and characteristics of $CuInS_2$ nanocrystal was investigated through a method in which copper acetate (CuAc), indium acetate $In(Ac)_3$ and dodecanethiol (DDT) was heated in presence of non-coordinating solvent octadecene (ODE) [29]. In another development, CuI in place of CuAc, DDT as a sulfur source and oleic acid were used to lower the reactivity of thioligand (RSH) and indium ions. This method fabricates pyramidal shape nanomaterials having PL emission peak between 550nm and 820nm [30]. Further, method of cationic exchange was used to synthesize several CIS QDs from $Cu_{2-x}S$ seeds having PL emission position at 870 nm but suffer from low photoluminescence quantum yield [31]. In place of DDT, amine surfactants hexadecylamine (HDA) and OLA were also employed to stabilize the CIS QDs [32]. Review highlight that, in the technique of hot injection, use of $CuCl_2$, $InCl_3$ and NaS_2CNEt_2 as precursors and OLA as capping agent produces zinc blende and wurtzite CIS [33]. It has also been suggested that the solvothermal technique is used to synthesized CIS nanomaterial having chalcopyrite structure by establishing a copper thiourea complex in the presence of octadecylamine [34]. The review of literature demonstrated the synthesis of narrow size distribution of luminescent CIS/ZnS core/shell nanocrystals from copper iodide, indium acetate, zinc stearate, and dodecanethiol in octadecene solvent having photoluminescence peak in the span of 550 nm-815 nm and

fluorescence quantum yield of 60% [35].

In another work, a direct hydrothermal technique has been suggested for the preparation of CIS QDs utilizing mercaptopropionic acid (MPA) as a ligand at temperature of 150°C. However, ligands used in aqueous synthesis had low complex stability towards In^{3+} leads to phase separation [36]. Few studies also suggested use of secondary ligand carrying multicarboxylate group (e.g., sodium citrate) to command the reactivity of the In^{3+}. Preparation of CIS QDs by employing binary capping agent, GSH and sodium citrate was shown at temperature of 95°C. It has been reported that use of highly-reactive Na_2S source at low temperature sometimes result in rapid nucleation which lead to introduction of more defects in the core and hence decrease the quantum yield (QY) [37]. As reported, this problem can be minimized by effectively passivating the core with a ZnS shell [38]. Methodologies for aqueous synthesis of CIS/ZnS core/shell QDs are facile but result in low QY (2–5%) [39]. In another study, a green route using autoclave has been found to be effective for the preparation of CIS QDs having 14% QY at temperature of 100°C and reaction time of 8h. This technique was found be unpleasant due to the application of short-chain thiols as it hydrolyze/pyrolyze to a certain degree and the rate of release of sulfur was effectively small when temperature is very high [40]. In another work, a simple technique that combine hydrothermal and successive ion layer adsorption and reaction has been suggested for the preparation of tetragonal chalcopyrite structure of CIS QDs utilizing MPA as a ligand at atmospheric pressure. It has been reported that growth of ZnS shell over CIS QDs increases the PLQY from 6.95% to 17.19% [41]. In one of the reports, work has been done on the synthesis of $CuInS_2$–ZnO nanocomposites by two integrated methods, solvothermal and chemical precipitation. The photocatalytic activity was studied due to different ZnO content on $CuInS_2$. This study suggests that $CuInS_2$–ZnO nanocomposites was found to be satisfactory for application in water depollution by photocatalysis as photodegradation rate for RhB discharge by $CuInS_2$–ZnO nanoparticles become greater with the incorporation of ZnO nanoparticles [42].

At national level, very little work has been reported on CIS nanomaterials. The review of literature suggested that the hydrothermal synthesis of tetragonal chalcopyrite CIS nano-cubes using copper chloride, indium, sulfur and deionized water at 180 °C for 20 h [43]. In another study, the deposition of CIS thin films has been demonstrated using chemical bath deposition technique employing cupric chloride, indium chloride as an anionic precursor, thiourea as a cationic precursor and triethanolamine (TEA) as a complexing agent in the reaction. The influence of deposition temperature and deposition time on the physical characteristics of CIS thin films were studied. The films deposited at 40°C was noted to be a single-phase and well adherent to the substrate [44]. Few work outline deposition of n-type $CuInS_2$ (CIS) thin film on indium–tin-oxide glass substrates by alkaline solution growth technique. In this technique, thin film was deposited from a solution containing copper sulphate, ammonia solution, indium chloride, citric acid solution and thiourea $(NH_2)_2CS$ in deionized water at pH value 11. Addition of triethanolamine in the solution gives more uniformity of film. It was reported film prepared at 40°C contains nanorods of diameter of the order of ∼20–38 nm and length of the order of ∼ 200–250 nm which display blue shift in optical spectra [45].

3. Optical & Photovoltaic properties of CIS QDSCs

In one of studies, work has been done to study the photovoltaic performance and optical properties of different sizes of pyramidal shaped $CuInS_2$ QDs capped with 1-dodecanethiol (diameter between 2.9 nm and 5.3 nm). Fig. 2(a) & 2(b) shows absorption and emission spectra for different sizes of $CuInS_2$ QDs in toluene. Absorption spectra display absence of well-defined excitonic peaks but shows presence of broad characteristic shoulder because of the dominance of internal and surface defects. The photoluminescence (PL) spectra exhibit broad emission in the visible and near IR. The PL emission peak broadens and shifted to the red wavelength side with increasing $CuInS_2$ QDs size. The large stoke shift shows absence of band edge emission and existence of defect induced emission. Fig. 2(c) shows J-V characteristics of the QDSCs for the various sizes of $CuInS_2$ QDs under the irradiation condition of AM 1.5 on an area of 0.20 cm^2. The highest photovoltaic characteristics was reported for 3.9 nm and 4.3 nm $CuInS_2$ with power conversion efficiency, PCE of 2.16 and 2.51 %, respectively. The drop in PCE is described for larger QDs having diameter 5.3 nm credited to reduction in charge separation following bandgap excitation [46].

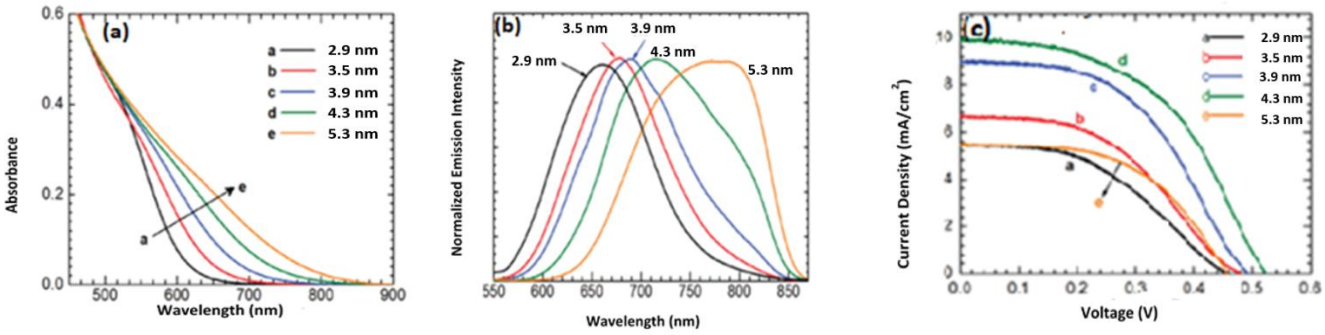

Fig 2. (a) & 2(b) Absorption spectra and emission spectra of various sizes of $CuInS_2$ QDs in toluene under 450nm excitation
(c) J-V characteristics of QDSCs manufactured with $CuInS_2$ QDs of different sizes under the illumination condition of AM 1.5 [46]

In another study, work has been done to increase the photovoltaic conversion efficiency from 4.04% to 4.68% in $TiO_2/CuInS_2$ QDSCs by modulating the interface connection of the solar cells with different heating temperature and heating time. Fig. 3(a) & (b) depicts J-V curves of $TiO_2/CuInS_2$ QDSCs with different heating temperature & heating time. Excellent charge separation and charge transfer properties has been investigated under heating treatment at 300°C for 5 min due to good quality of interface connection between $CuInS_2$ QDs and TiO_2 films [47].

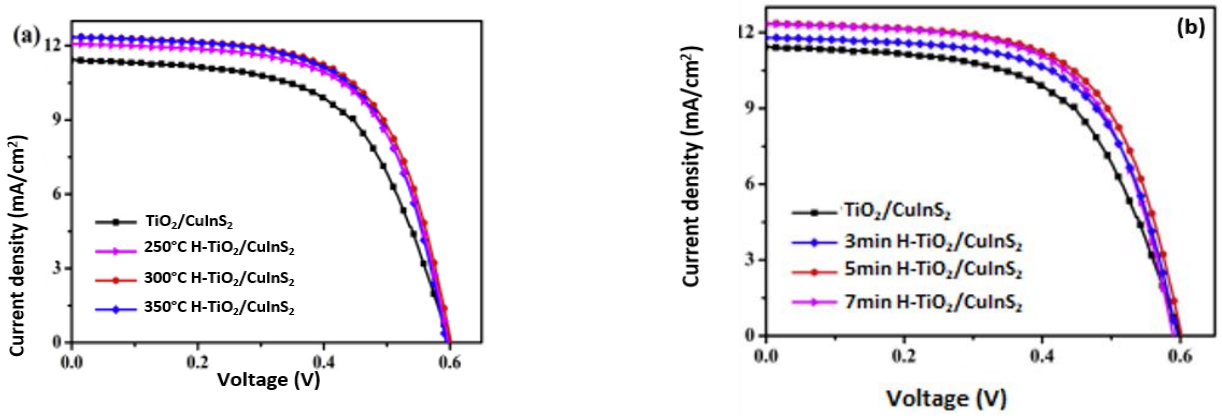

Fig 3. J-V curves $TiO_2/CuInS_2$ QDSCs with different (a) heating temperature (b) heating time [47]

In one of the works, reported efficiency of solar cells sensitized with CIS nanoparticles (NPs) is 0.1203%. In this work, CIS NPs of average size 38.9 nm were synthesized by biomimetic technique using copper sulphate and indium chloride as precursor salts and biological molecule glutathione (40 mM GSH) as sulfur donor at low temperatures and at atmospheric pressure [48].

In one of the reports, QDSCs sensitized with core–shell CIS–CdS QDs shown an efficiency of 4.69% under the illumination condition of AM 1.5 and 100 mW cm^{-2}. However, on doping CdS with Mn, CIS–MnCdS QDSCs shown an increase efficiency of 5.38% as illustrated in Fig. 4(b) depicting J–V characteristics of QDSCs solar cells sensitized with different QDs. Fig. 4(a) shows absorption spectra of TiO_2 photoanodes sensitized with different QDs. From the Fig. 4(a), it can be clearly seen that absorption edge of CIS QDs is blue-shifted due to significant quantum confinement effects. With Mn-CdS coating on the CIS QDs, absorption of light was greatly enhanced in the visible light region and absorption edge shows a significant red shift indicating a promising sensitizer for QDSCs [49].

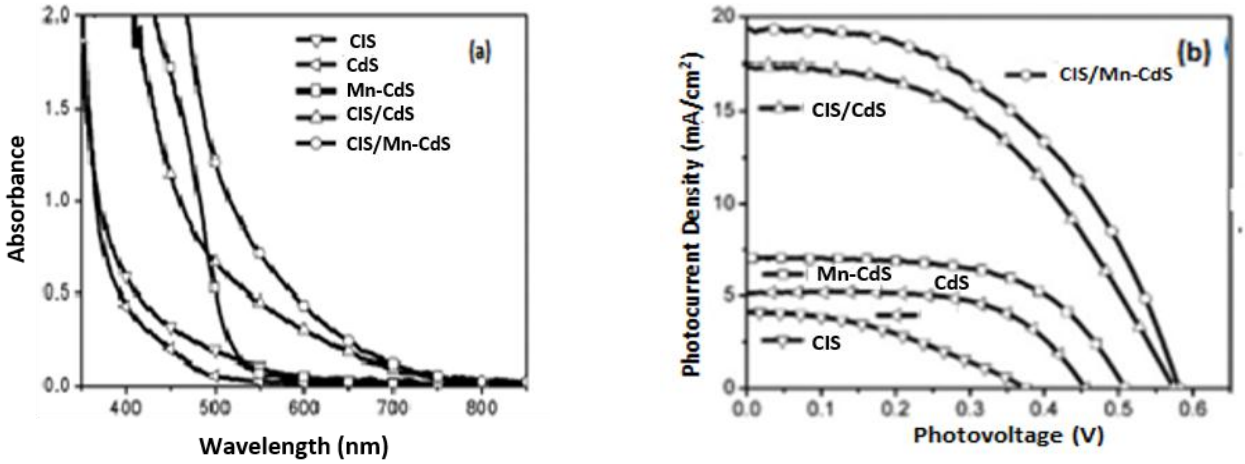

Fig 4. (a) Absorption spectra of TiO_2 electrodes sensitized with different QDs (b) J–V characteristics of QDSCs solar cells sensitized with different QDs [49]

Early studies suggest that $CuInS_2$ QDSCs containing quasi-spherical particles of diameter about 2.5 ± 0.5 nm were synthesized by the combined ex-situ/in-situ technique possess a PCE of 1.84% under the illumination conditions of AM 1.5 and 100 $mWcm^{-2}$ which is more than the PCE of QDSCs containing water-soluble QDs prepared by conventional one-pot protocol. In this case QDSCs consists of TiO_2 photoanode containing $CuInS_2$ QDs synthesized by ex-situ method through a two-step ligand-exchange method. Prepared $CuInS_2$-QDs based TiO_2 electrodes was further deposited with $CuInS_2$ QDs synthesized by in-situ method through the repeated SILAR cycles followed by ZnS passivation layer. Prepared ex-situ $CuInS_2$ QDs were encapsulated by oleic acid (OA) ligands form OA($CuInS_2$) QDs. MPA($CuInS_2$) QDs formed when OA($CuInS_2$) QDs were mixed with bifunctional molecular linker, mercaptopropionic acid MPA. Water-soluble QDs synthesized by conventional one-pot method were denoted as CIS@MPA QDs.

Fig. 5(a) depicts absorbance and emission spectra of OA($CuInS_2$) QDs, MPA($CuInS_2$) QDs & CIS@MPA QDs. From the figure, it can be clearly seen, wavelength of the highest photoluminescence (PL) emission location for the CIS@MPA QDs is around 602 nm. Shape of PL peak of OA($CuInS_2$) and MPA($CuInS_2$) QDs did not change, but shows a red shift (10 nm) in the PL peak position. Fig. 5(b) shows the photovoltaic characteristics of the $CuInS_2$-based QDSCs under the irradiation condition of AM 1.5G and 100 $mWcm^{-2}$. Fig. 5(b) shows MPA($CuInS_2$)-based QDSCs lead to a PCE of 0.64% which is more than OA($CuInS_2$) and CIS@MPA based QDSCs. After the in-situ growth of $CuInS_2$ QDs on the TiO_2 surface, MPA($CuInS_2$)/$CuInS_2$-based QDSCs obtained after five SILAR cycles shows a maximum PCE of 1.84% higher than the PCE manifested by $CuInS_2$ QDSCs synthesized by either ex-situ for and in-situ technique respectively [50].

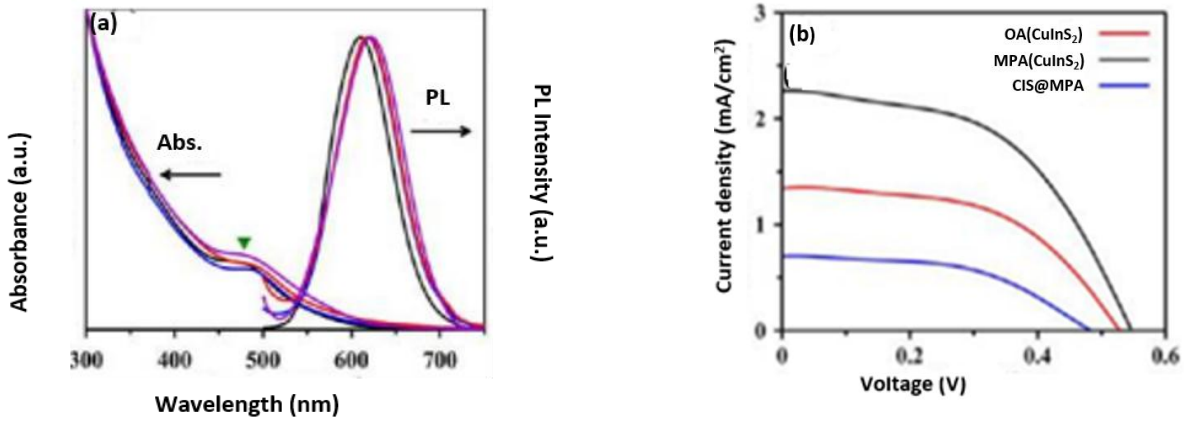

Fig 5. (a) Absorption and Photoluminescece spectra under excitation of wavelength 430nm of OA($CuInS_2$) (blue lines), MPA($CuInS_2$) (red lines) & CIS@MPA (purple lines) QDs (b) J−V characteristic curves of OA($CuInS_2$) QDSCs, MPA($CuInS_2$) QDSCs & CIS@MPA QDSCs [50].

4. Conclusion

The present review highlights that Copper indium disulphide $CuInS_2$ (CIS) is an attractive nanomaterial for quantum dot solar cell. Properties of CIS nanomaterials are found to be interesting for solar energy conversion. It has been reported that in quantum dot solar cell, CIS in particle form having diameter in the range of 1nm-10nm is usually preferred than in polycrystalline thin films form. Synthesis of CIS quantum dots has been reported by two methods organic method and water-soluble method. However, organic method does produces quantum dot that has better crystallinity, better control of shape and size, better photoluminescence properties but suffer from challenges of synthesizing at room temperature, complex synthesis steps and not found to be useful in biological applications. Water-soluble method produces nanomaterials suffer from poor optical properties, difficult to control size but found to be useful in biological applications. Photovoltaic properties of CIS QDSCs depend on the size of used QDs, heating temperature and heating time during synthesis of CIS QDs, type of doping, and the technique used for the synthesis of CIS QDs. Combination of ex-situ & in-situ method shows better PCE than either ex-situ or in-situ method.

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Conflict of Interest

The author declare that they have no known competing financial interests or personal relationships that could have appeared to influence the review reported in this paper.

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