Introduction

We, as a society nowadays, are in constant demand for energy. Although, even with the development of calculating, conducting and studying it, we don't really know what the energy is, we are depending on that abstract quantity. The rise of energy consumption has proven that in the future we will almost certainly require even more. The necessity of searching new possible ways to harness it through renewable sources has become a global issue. Our concern of the environment had never been that serious before. With conventional methods still being the vast majority, the new approaches that might solve most of our problems, are arising to become significant. Among all of the ideas created in a past few decades, solar energy is believed by many to be the most reliable and promising. In the last ten years the world solar PV electricity production has grown impressively, being almost three times bigger in 2016, than in 2010 (1). In theory, the Sun has the potential to fulfil earth's energy demand if it is not for the technology. Annually, nearly 10^{18} EJ of energy reaches our planet and of that $10^4 EI$ is claimed to be harvestable (2). The possibility of converting the solar energy into electric energy has been studied since discovery of the basic photovoltaic effect and the development semiconductor technologies.

Since the first crystalline silicon solar cell, the technology had undergone a fast development. The number of different methods that the cell can be created with and the quantity of possible final outcomes isn't inconsiderable. Yet, solar cells can be classified into three generations in accordance to the development

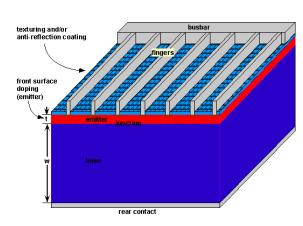


Fig. 1. Basic schematic of silicon solar cell. (103)

technology and materials used. (3) The first generation describes silicon based solar cells, which so far is the most researched type. It includes monocrystalline and polycrystalline silicon cells.

The second generation contains thin film solar cells. It refers to amorphous silicon-, copper indium gallium selenide (CIGS)-, GaAs- and CdTe- devices. (3) (4) (5). With many advantages such us direct band gaps resulting with harvesting light in very thin films they still have a small share in the market of PV devices (<10%) due to their instability and limitation in module technology (3).

Yearly sum of Global Horizontal Irradiation (GHI)

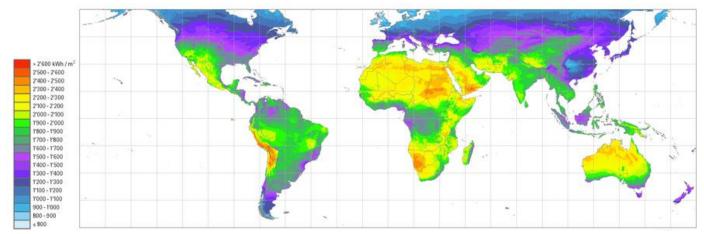


Fig. 2 Annual yearly sum of Global Horizontal Irradiation (102)

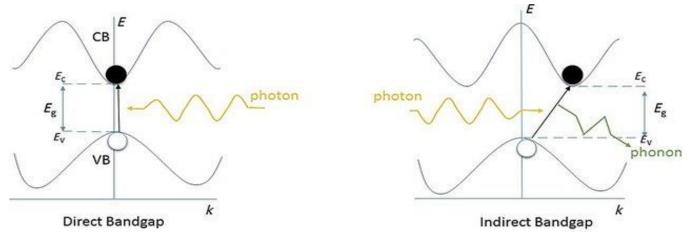


Fig. 3. Schematic illustration of direct and indirect bandgap. The indirect transition is accompanied by phonon emission. (104)

The third generation cannot be simplified as much as its' formers. It is usually defined as solar cells that are still in scientific research phase. (3) (6) Its' main motivation is to achieve a higher efficiency using newly discovered physical phenomena, structures and materials. Their aim shall vary depending on the development technique and the physical principles. Today, the third generation mainly includes perovskite solar cells(PSCs) (7) (8), organic/polymer solar cells(OSCs) (9), dye sensitized solar cells(DSCs) (10) (11) and quantum dot based solar cells(QDSC). (12) (13) While still being considerably less common, the cons of investigating the nature and crucial properties of third generation PVs is overwhelming. Thanks to the ultimate specification of third generation approach, the devices can in perspective generate greater efficiency than other generations(for example by creating multiple electron-hole pairs and multiband cells), while being significantly cheaper.

Our main goal will be to study the QD sensitized solar cells and their characteristics. The base shall be put on

PbS quantum dots as a light-harvesting material. We will proceed to consider their tuneable band gap and other properties such as control of composition, dipole moments, stability or collocation and fit other materials to study the effects and their attributes. In principle, the QD solar cells can be distributed into four different classes: Schottky junction solar cells, p-n junction solar cells, hybrid QD-polymer solar cells and quantum dotsensitized solar cells(QDSCs).

1. Basic principles of a Solar Cell.

The parameters that determine the design and performance of solar cell are:

- a) Concentration of doping atoms(donors-which donate free electrons, acceptors-which accept electrons)
- b) The mobility μ (a measure of how easily can charged particles be moved through a semiconductor material) and diffusion coefficient D of charge carriers (tend of particles to move into regions of low

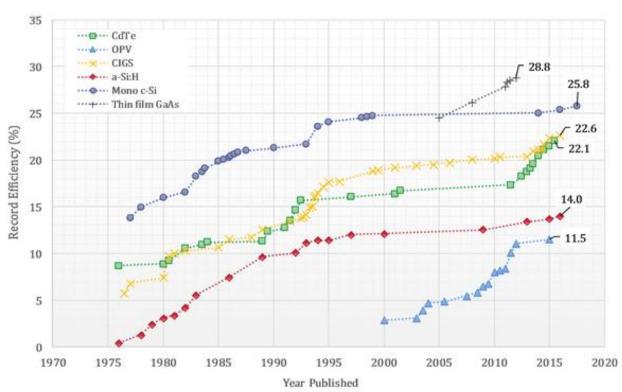


Fig. 4. Record efficiencies for second-generation thin-film cells, compared to organic PV (4)

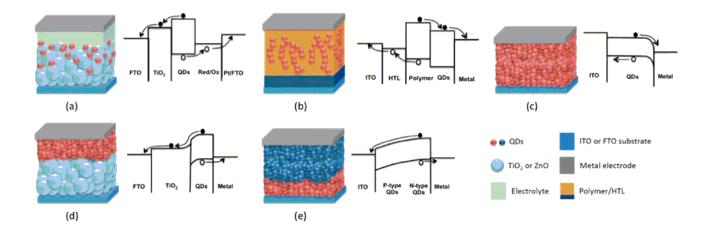


Fig. 5. Schematic illustration of configurations and band structures of QD solar cells: a) QD-sesitized b) hybrid QD-polymer c) Schottky junction d) p-n heterojunction e) p-n homojunction (3)

- concentration), which characterises the transport properties
- c) The lifetime of carriers and diffusion length which is directly connected with it and characterises the recombination processes.
- d) The band gap energy and whether it is direct or indirect
- e) The complex refractive index linked to absorption coefficient that characterise the ability to absorb electromagnetic radiation

The working principle of PV device is based on photovoltaic effect, where the emitted electrons directly enter new material upon emission, in contrast to photoelectric effect alone. The electrons can be emitted by absorbing the energy of the electromagnetic wave of energy $E = h\nu$, where ν is the frequency of the radiation and h is a Planck constant. Albert Einstein, in 1905, assumed that the light consist of energy quanta called photons. The foregoing energy is considered for such a particle. Einstein only assumed the existence of photons but the occurrence of Sub-Poisson statistics was shown in Quantum Electrodynamics, and that has proven the guess correct. The absorption of photon in a material by a basic model means that the energy is conducted to excite electron from the lower energy level called a valence band to a higher energy level called a conduction band. Between those bands, there are no energy states populated by the electrons, so the energy must exceed the difference of valence band edge and the conduction band edge. In reality the model is much more complicated but the concept stays the same. After excitation the Electron-Hole pair is created and the excess carriers are present($n_0 \cdot p_0 = n_i^2 < n \cdot p$ where n_0, p_0 are equilibrium concentrations, n_i is intrinsic concentration and n, p are excess carriers concentrations). After that, the recombination process occurs. It is described by the recombination rate, which strongly determines the performance of solar cells. The recombination and generation processes can be both direct and indirect. They are much more likely to

happen in direct band gap materials in which case the transition in usually radiative (photon emission).

The recombination processes are mainly:

- a) Direct recombination
- b) Shockley-Read-Hall recombination facilitated by the impurities or defects of the lattice. The process is typically non-radiative and it dissipates the excess energy back to the lattice in form of heat. The donor and acceptor traps are possible.
- c) Auger recombination which requires three particles. The energy and momentum is conserved by transferring them to an another electron(hole), which excites it to higher levels in electronic bands. Then the particle relaxes again and the energy is transferred to the lattice by phonon emission.
- d) Shallow levels recombination
- e) Exciton recombination

2. Colloidal Quantum Dots

CQDs are semiconductor crystals of the nanometer-scale size, with the diameter of less than twice the Bohr radius size, which are restricted with surfactant molecules and deployed in a solution. They have manifested to provide a development of numerous types of optoelectronic devices including photodiodes and PV devices. The properties of CQDs are easily adjusted by changing the volumetric features of nanoparticles similarly to metallic nanoparticles in plasmonic transport. (14) (15)

a) Quantum dot size.

With changing the size of the nanoparticle we can control the band gap over significant range of spectrum. Yet, practically, the width of the nanoparticle is estimated from the energy band gap. (16)

b) Quantum dot shape.

The geometry in physics plays an important role, there is no question about that. It is obviously not different for Quantum Dots. The quantum confinement strongly depends on the shape and dimensional geometry. The ability to control the growth of certain form allows the nanoparticles to exhibit a variety of properties.

As it is the introduction part we will proceed to investigate different forms later.

3. Chemical properties controlling.

a) Ligand exchange

While long-chain surfactants enable us to have a firmly stable CQD solution they are an insulating medium as well. (17) (18) To achieve massive improvements in film properties shall we partially or fully exchange the initial ligands in solution. The first studies of reactions to exchange the ligands were based on replacing the long-chain ones with organic short molecule structures(butylamine or pyridine (19) (20)) Also, the metal chalcogenide complex for these cases were tested widely and also quite successfully deployed. (21)

b) Alloying and Doping.

The inner relationship between the band gap and fraction of nanocrystal particles in solution gives nonlinear characteristics and shall be studied to definitely provide the optimum. The distribution can determine precisely if the alloy is graded or homogeneous. (22)

Doping CQDs with other nanoparticles can also allow different strategic fabrication of PV structure than using single type of CQDs. An example of such behaviour can be PbS CQDs doped with silver, which has led to increasing the solar cell efficiency via introducing an extra charge carrier in the film (23).

c) Core/Shell Quantum Dots.

Easy desorption of the ligands from the surface can be limited by more convenient surface passivation through the introduction of extra material shell- a core/shell structure. It can provide an isolation that would allow the core atoms to last significantly longer than without it.

Again, the detailed part shall be provided in the next chapters.

d) Fully inorganic solar cells.

Between all of the examples shown above, a new idea of using all-inorganic CQD Photovoltaics has marked its' extraordinary intro. The approach to brand new CQDs films production based on the replacement of organic ligands with halide anions

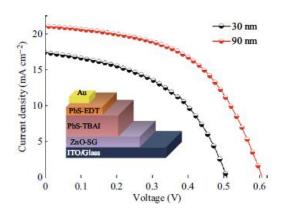


Fig. 6 The structure of all-inorganic solar cell based on PbS CQDs with optimized ZnO film and its I-V characteristics for two different ZnO film thickness. (107)

has become the new track for scientist. There are some advantages of using such a method of omitting redox reactions with reducing the inner density of trap states within the bandgap among them. The so-called ligand passivation can be a strategy to ultimately enhance device performance. (24) Shall we ponder only the working principles, by many means the scheme isn't much different from other approaches but it could remind us more about the original, first generation PV devices rather than organic ligand-based devices.

Within this rather modern idea, the PbS and PbSe CQDs are commonly used. The widely explored architecture is shown in (Fig. 6). A numerous researches and their record results has shown that architecture to be very promising. (107) The passivation of ligands has been achieved by different fabrication methods resulting in various models and their performances, yet based on the same idea. (107) (26) (27) In case of this paper we shall also investigate whether we can improve the scheme of those structures and create even more stately device prototype.

4. QDSC in principle.

The great achievement of these types of solar cells is the decoupling the charge generation and its' transport to different materials- the hole and electron transport layers. The effect of such a procedure is the decrease in recombination process and reduction of production costs. Without that technique, the architecture is adequate to the standard PV solar cell device. The generation of electron-hole pairs proceeds the injection of electrons from Conduction band of light harvesting material to electron acceptor layer and holes from valence band to hole acceptor layer. We call this process a charge separation. Nevertheless, even though we do regenerate QDs in the light harvesting area after charge

separation, we still have to struggle with recombination processes and consider them as the significant performance wasters. (24)

5. Materials and performance development.

a) Electron transporting materials (ETMs)

They are used to support charge separation as briefly described above. The function proceeds as a support for QD in the charge generation layer as it transports charged electrons to the conductive substrate(usually FTO) (25) (26) (27) The properties shall be as follows:

- Matching CB edge which will determine exciton generation and partial transfer efficiency.
- High electron mobility.
- Fine surface to provide suitable loading of particles.
- Suitable technological properties, such as stability, low cost etc.

The most widely studied materials nowadays are TiO₂ and ZnO. Materials based on first compound are of such importance due to their nontoxicity, chemical stability and low cost (28) (29). Among them (TiO₂-NP) based mesoporous layers are studied as photoanodes (28). The highest efficiency ever recorder were based on that material (30) (31) (32). Unfortunately, the probability for charge recombination in that kind of films is rather unsatisfactory. Therefore, one provides onedimensional structures such as nanorods and nanowires to improve smoother electron transport channel and decrease loses provided with recombination (33) (34) (35). In 2007 it was shown that TiO₂ nanotubes win over nanoparticles of the same material in case of transport capacity (36) (37). Thus, the research of such structures was accelerated and CdSe sensitisation was proposed (38). After that, many different sensitisations were provided, in which also PbS QDs had their small part (39). Although promising, the 1D structured TiO₂ based anodes were unsatisfactory in case of PCEs comparing to standard nanoparticles (maximum of around 6% (35)). The hypothesis was put on insufficient loading amount on 1D structured TiO2, therefore the focus

should be put on improvement of that area. A group of researchers has also used graphene frameworks incorporated into TiO₂ photoanode achieving 4.2% PCE for QDSSCs. (40) By using double-layered films with particles of bigger volume, there has been achieved a high PCE of 4.92%. A lot of dopants and different solutions, hybrid photoanode films with metal and nonmetal ions and carbonaceous layers has been proposed to improve performance of PV devices. Also with the usage of hollow structure techniques, by creating nanotube arrays, the possibility of achieving a PCE of 6% was shown. (41) Improvement light absorbance was achieved by using microporous TiO₂ photoanode for PbS quantum dot sensitised solar cells achieving up to 3.5% PCE performance. (42)

In case of ZnO based photoanodes the differencing property is a higher electron mobility and better conduction band edge. With them, the achievement of higher V_{oc} is more probable (43) (44). Unfortunately, the chemical stability of those films is significantly lower. Similarly to the former, the nanoparticles were widely studied. (45) (46) (47) (48) As an example, the CdS sensitised ZnO nanoparticles were used to construct a photoanode, which allowed the achievement of 4,46% PCE. They used TiO₂ passivation of ZnO surface to improve the PCE of same sensitized CdS/ZnO QDSCs by more than 2%, achieving 4.68%. (47) Similarly, the 1D structures for ZnO can be crystallised. The distinction from the former is the ease of its' development (45) (47). The ZnO nanowires sensitised with CdSe constructed in 2007 were the launch of the technology development and allowed Young et al. achieve PCE of 4.15% (49) (50) (51) (52). Many more one dimensional structures of ZnO based photoanodes were used to research maximal performance but it was shown that the problem again concerns the surface area (53). For example, it was shown that modification of surface of ZnO NRs with TiO₂ improves PCE from 1.54% up to 3.14% (54). The second strategy to improve the performance of such material based structures is double layer replacement with two different 1D structures such as NR on bottom and TP on top. However, the PCEs of ZnO based films

ETM	QDs	CE	Electrolyte	$J_{\rm sc}$ (mA cm ⁻²)	V _{oc} (V)	FF	PCE (%)	Ref.
TiO ₂ -NP	ZCISe-CdSe	MC/Ti	Polysulfide	27.39	0.752	0.619	12.75	112
TiO2-NW	CdS/CdSe	Cu ₂ S/FTO	Polysulfide	19.32	0.531	0.586	6.01	145
TiO2-NT	PbS	Pt	I-/I ₃ -	8.48	0.64	0.63	3.41	127
TiO2-NR	PbS	_	Spiro-OMeTAD	13.56	0.52	0.579	4.10	133
TiO2-NS	CdSe	Cu ₂ S/brass	Polysulfide	16.95	0.591	0.50	5.01	148
ZnO-NP	CdS/CdSe	Cu ₂ S/brass	Polysulfide	10.48	0.683	0.623	4.46	149
ZnO-NW	$Zn_xCd_{1-x}Se$	Cu ₂ S/brass	Polysulfide	18.05	0.65	0.40	4.74	105
ZnO-NW	ZnSe/CdSe	Cu ₂ S/brass	Polysulfide	11.96	0.836	0.45	4.54	150
ZnO-NR	CdS/CdSe	Cu ₂ S/brass	Polysulfide	13.28	0.642	0.567	4.83	151
ZnO-TP	ZnSe/CdSe/ZnSe	Cu ₂ S-RGO	Polysulfide	17.3	0.761	0.471	6.2	152
SnO ₂	CdS/CdSe	Cu ₂ S	Polysulfide	10.13	0.700	0.616	4.37	153
NiO	PbS	Pt	$[Co(dtb-bpy)_3]^{2+/3+}$	5.27	0.227	0.33	0.40	154

Table 1. Photovoltaic performance summary for QDSCs based on different Electron transport materials. (3)

still stay behind TiO₂ ones. (55) (56) The Al/Cl hybrid doping allowed to use ZnO in IR spectrum. (57)

There are other kinds of ETMs researched as well. Using big tandem structures has manifested superb PCE through simulations. (58) The doping of synergistic fullerene electro transport layer has proven to increase Solar Cell performance (59) A series of materials such as SnO₂, NiO, BiVO₄, Zn₂SnO₄,BaTiO₃, CoO has been incorporated to QDSC manifesting future potential. (41) High performance has been achieved using CdS thin films as single-source precursors to ETM layer providing over 8% PCE. (60)

All of the above materials are n-type semiconductors. Furthermore shall we proceed to introduce the p-type metal oxides and p-type Quantum dot sensitised cells. Typically, the NiO semiconductor is widely used. The promising and comprehensive material that can be implemented is CuSCN, which is an inorganic of ptype. Of course, the extraction of a hole from light harvesting layer to a redox couple will be rather slower than in case of electron. (39,70,192) Therefore, being the potential solution to this problem, the p-type semiconductor based layers are beginning to leave their mark in PV devices.(193-195) As expected, the most accurate application of that type of films would be the tandem configuration construction, which contains both n-type and p-type QDSCs to overcome so called a Shockley-Queisser limit(196). However appealing, the efficiencies of p-type QDSCs are yet to be enhanced. (3)

b) Hole transporting material (HTM) layers

The electrolyte or HTM is crucial to QDSCs as well. The properties of such film shall be:

- Low corrosivity.
- Redox potential appropriate to regenerate QDs and maintain fine $V_{\rm oc}$.
- High conductivity through ions.
- Stability and transparency in visible spectrum.
- Possibility to fully regenerate. (3)

The common electrolytes:

- Liquid electrolytes
- Quasi-solid state electrolytes
- All-solid state ODSCs

were described comprehensively in the following research papers (3) (61) (62)

Although using Sb_2Se_3 as a thin light harvesting film, a group of researchers has also used PbS colloidal quantum dots as a hole transport layer achieving 6.5% certified PCE in 2017. (63) In 2016 the usage of graphdiyne(a novel large π -conjugated carbon hole

transporting material) was used for an efficient hole transport layer for solar cells based on PbS-EDT colloidal quantum dots. (64) Before that, in 2015, colloidal CuInS₂ QDs were layered to hole transporting solution and even though the scientist did their research on Perovskite solar cell, they accomplished to get almost 8.5% PCE. (65) The extended device stability and a rise to 10.6% of PCE was certified using CQD solar cells using P3HT as a hole transport material. (66)

c) QD sensitizers

The main part of our PV device, the QDSC are QDs. The ability of harvesting interfering light is a crucial component of such device. (67) (68)Therefore, the ideal nanostructures should exhibit certain properties:

- A higher CBE than the one in the electron transport material and lower than in hole transport material to provide effective charge separation.
- Obviously, as in all semiconductor PV devices, we shall provide a material with accurate band gap, ideal for our purpose. The reason is clear, we are interested in superb absorption in wide range of solar spectrum.
- The stability is the crucial property as well.
- From the technological point of view- the simple preparation and of course low toxicity would be rather expected.

Therefore, we will now examine methods to deposit them and the differences between certain QDs materials.

Because of QDs being inorganic and possessing larger size than simple molecular dyes, they are much more difficult to tether onto metal oxide to form a high quality monolayer. Therefore high QD-loading amount is rather high to achieve. (69) We can difference the deposition methods by putting them into two categories: in situ and ex situ. In the first one, the QDs are grown directly on the surface of the metal oxide substrate, being created using an ionic precursor. We can include chemical bath deposition(CBD) and successive ionic layer adsorption and reaction(SILAR) into this category. The second approach bases on pre synthesising of QDs and then depositing them onto the metal oxide. Easily processable and finely reproductible, the in situ method has been used more widely than its' counter. In that kind of deposition we can control not only the OD-loading amount but also size distribution. Unfortunately, the achieved density of trap states is rather high, therefore the obtained maximal PCE is about 7% (70) (71). In this case, excluding the QDs growth process, we have to accurately deploy them onto the surface. The most common methods are: direct absorption, linkermolecule-assisted self-assembly, electrophoretic deposition. (3) (72)The ligand part in QDs is, as mentioned before, the important part of obtaining high efficiency of ODSCs. (73) The comparation of halide ligands in PbS CQDs for field effect transistors has been made by researchers in 2018. (74) The capping-ligandinduced self-assembly method was the clue for TiO₂ photoanodes. (3) Nevertheless, researchers haven't discovered a satisfying approach yet. In 2012, the deposition method were optimised through using ligand-exchange techniques. (75) (76) Thanks to that, the PCE record of 13% was achieved (77) Not only CLIS deposition has been used. The aqueous solution provided the simplification of QDs fabrication with shorter ligands. (78)Some relatively high PCEs have been achieved with that method. (3) Then, the organic molecules usage allowed scientist in 2015 to achieve 300% times PCE compared to common creation of PbS quantum dots. (79) The certified PCE of 11.21% has been achieved via so called "solvent curving", which is the simplified method of PbS QDs fabrication processing. (80) The passivation of PbS QD surface with L-glutathione was used to produce QDSCs with promising results. (81) 10.6% PCE was achieved thanks to solvent-polarity-engineered halide passivation. (82)

Up to date, binary QDs have also been used as sensitizers. There are many examples but the most common are: InP, InAs, CdS, CdSe, CdTe, Ag₂S and with them, the most interesting one considered in our case-PbS. (83) (84) (85) (86) (87) (3) The main problem with them is to control and balance the narrower band gap and relatively high conduction band edge. For example, for PbS nanocrystals the band gap is narrow, but the conduction band edge is rather low, which causes the problem and has to be dealt with. In case of binary QDs we have to balance between the light harvesting efficiency and efficiency of charge injection. Mixing binary structures (for example PbS with PbSe) has been also proven to enhance the interesting efficiency. (88) (89)Treatment of PbS QDs with metal salts provided some advantages in CQD PV devices resulting in the increase of short circuit current and fillfactor. (90) The crucial part of success in getting high efficiency would also be suitable engineering of solvent. (91)

The other methods to use QDs as sensitizers in PV devices is creating a Core/Shell QDs. Their unique properties have been mentioned before. The alignment in these provides ability to tune the light-absorption range, recombination processes and charge separation. Usage of them in QDSCs is rather modern. The first noticed implementation was created by Lee et al. in 2009. Through SILAR method, he achieved PCE of 4.22%. (92) Yet the difficulties in creating specific materials may occur, because of inability to prepare a

stable precursor for deposition methods. Core shell QDs can be classified into three categories: type I, reverse type I, and type II structures. (93) (94) (95)A great deal of detailed information can be possessed from that review (3).

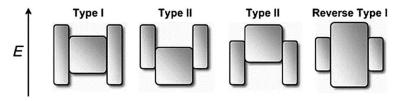


Figure 6.. Schematic illustration of energy levels in different core/shell QDs types.

The massive perspective in PV devices has also been established by using Alloyed QDs. These allow us to create the non-linear band gap (96) (97) (98). Stability in such materials is again much higher than in their constituents. A lot of scientific research was done in that area. The summary is satisfactorily described in publication below. (3) The review finely describes recent achievements and potential usage of alloyed QDs in the near future.

We can also include dopants to QDs. There were a handful of propositions in the former review as well. Yet, the promising idea has been introduced with the usage of metallic nanoparticles dopants by considering plasmonic physical phenomena. (99) (100) (62)

d) Counter electrode

As we may already expected, they are also playing an important role in achieving high performance of PV device. The electrodes have to catalyse reduction reaction. (7,42,51) These shall poses properties as follows:

- Good conductivity
- High catalytic activity
- Fine stability, either chemical and physical

We can also put them into four categories:

- Noble metals
- Metal chalcogenides
- Carbon materials
- Composite CEs

Recent progress has been comprehensively described in those publications (3) (101) (61)

6. Recombination control

Non-radiative charge recombination can seriously disable any performance of PV device. (33,38) Because of defects states in QDs, the recombination is more serious than in normal DSCs. The recombination processes maintain standard paths, but of course they have their own specificity. (3) Controlling of

recombination processes can be either through material engineering and interface engineering. (3) More specific information shall be provided later.

7. Stability

Controlling of stability can be achieved through material design and of course temperature adjustment. More specific information is also provided with the (3) review.

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