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Quantum dots sensitized solar cells

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To improve the conversion efficiency of solar cells, quantum dots (QDs) have been widely used for the design of third generation solar cells for their interesting properties, such as multiple exciton generation, high absorption coefficient, tunable band gap, hot electron transfer, etc. Quantum dots sensitized solar cells (QDSSCs) as some of the third generation solar cells are under intense investigation. Immense efforts have been devoted to improve the photovoltaic performance of QDSSCs in the past few years. The conversion efficiency of QDSSCs was greatly improved and the best record of 7.04% has been achieved in a recent report. In this paper, the QDSSCs were reviewed from the aspects of working principle, performance, current research progress and developing trends of key parts (QDs sensitized photo-anode, counter electrodes, and electrolyte). Finally, some potential solutions for QDSSCs are proposed for further development.

Key words: Quantum dots (QDs), quantum dots sensitized solar cells (QDSSCs), conversion efficiency, counter electrodes, electrolyte.

INTRODUCTION

Due to excessive use of fossil energy, many problems have been caused, such as energy shortage, global warming, ecological deterioration, environmental pollution, etc. [1-3]. In recent years, these problems are becoming more and more serious. To figure out these problems, several renewable energy sources are being investigated to evaluate their potential to address large-scale demand. These sources include wind power [4], tidal power [5], solar energy [6-8], etc. Compared with other renewable energies, solar energy has these advantages: clean, widespread, inexhaustible, safe and reliable. So, many countries have invested a lot of research funding on the exploitation and utilization of the solar energy, and action plans have been formulated by the governments of these countries. The most direct way to utilize the solar energy is the use of photovoltaic (PV) cells, and the PV market has shown exponential growth over the last few years. Worldwide PV systems with a total peak power of about 6GW were installed in 2008. Nevertheless, PV electric power cannot compete with the price of electricity from the grid and is therefore supported by national subsidy programs with the aim of reducing the cost of PV systems by increasing the production volume. So, more research should be done in the future to increase the conversion efficiency and reduce the costs of PV systems.

So far, the PV cells have gone through two generations, which depends on the underlying technologies. The first generation device is single or multi-crystalline p-n junction silicon solar cells, which are the most common PV systems with a market share of about 85%. The second generation device is semiconductor thin film solar cells, which have currently a market share of about 15% and are mostly based on CdTe, $CuInS_2$, $CuInGaSe_2$, etc. The former has reached conversion efficiency of over 20% [9]. However, many problems have not been solved out, such as high material costs, high fabrication temperature, negative influence for environmental protection, etc. The latter has lower cost but low conversion efficiency. The highest conversion efficiency is only about 14%.

According to the limit theory of S-Qmodel [10], which was proposed by Schockley and Queisser, the limit conversion efficiency of 1st and 2nd generation PV systems is about 32.9% [11]. How to balance the costs and the conversion efficiency is becoming more and more important, this being the bottleneck in developing solar cells.

To figure out this bottleneck, several potential PV technologies were proposed, such as dye sensitized solar cells (DSSCs) [12], quantum dots sensitized solar cells (QDSSCs) [13], and organic solar cells (OSCs) [14]. Although these technologies are still at the exploratory stage, they are becoming the hottest topic in the field of solar cells, considered as third generation PV devices.

DSSCs were firstly fabricated by Grätzel and O'Regan in 1991 [15]. Since then, extensive efforts

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have been exerted to increase the conversion efficiency of DSSCs. However, the development of DSSCs has not contributed to more than 12% of the highest recorded conversion efficiency over the last 20 years [12, 16]. There are several problems that should be further studied and solved out, such as complicated fabrication process, weak absorption in the near infrared wavelength range, poor steadiness, short life of excited state. The performance of the sensitizer is the key factor for improving the conversion efficiency of DSSCs. Therefore, some new sensitizers or semiconductors are required, which can replace the organic dye.

Over the last few years, quantum dots (QDs) have attracted widespread attention due to their outstanding opto-electronic properties. QDs are nano-crystals made of quasi-zero-dimension semiconductor materials. The size of QDs is usually about 1-10 nm, which is small enough to exhibit quantum mechanical properties. Specifically, their excitons are confined in all three spatial dimensions, which are smaller than or comparable to their deBroglie wavelength or to the Bohr radius of the excitons in the semiconductor bulk. The frequency range of emitted light is inversely related to its size. Since the size of QDs can be set during their production, their conductive properties may be carefully controlled. This allows the excitation and emission of QDs to be highly tunable. Moreover, they show quantum confinement effect (QC) and multiple excitons generation effect (MEG) [17-19]. QD-based solar cells devices were proposed to realize 3rd PV systems and to achieve conversion efficiency beyond the S-Q limit, where the QDSSCs are one of these architectures.

The QDSSCs were evolved from DSCs, and they have similar structures. The big difference between them is that QDs were used in QDSSCs to replace the organic dye in DSCs to excite more electrons. Compared with DSCs, QDSSCs have many advantages, such as broad absorption spectrum range, good chemical stability, high extinction coefficient, multiple excitons generation effect, etc.

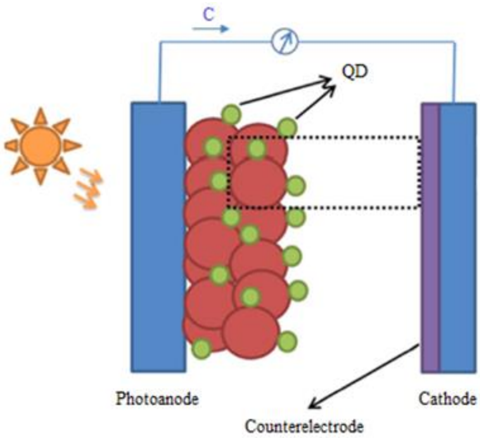
However, the results of recent research have shown that the conversion efficiency of QDSCCs is lower than that of DSCs. The main reason is that there is serious electron loss because of charge recombination at the electrolyte-electrode or electrolyte-counter electrode interfaces [20-21]. In order to overcome this problem, many studies have been done, and some important results have been achieved. It is beneficial for the researchers to do some further study after the current research results are reviewed.

Here, I will review the QDSSCs from the aspects of working principle, performance, current status and developing trend. Finally, some potential solutions for QDSSCs are proposed to reduce the costs and improve the conversion efficiency.

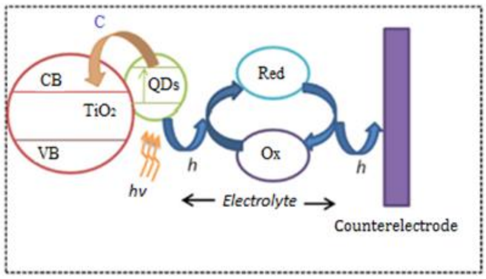
WORKING PRINCIPLE OF QDSSCs

Fig.1 shows the working principle of typical QDSSCs, where Fig.1 (b) is the amplified picture of the dotted line rectangle of Fig.1 (a).

In general, QDSSCs consist of quantum dots sensitized photo-anode, a counter electrode, and an electrolyte. The photo-anode is composed of an oxide semiconductor material, mostly TiO_2, which can be prepared and sintered easily as the TiO_2 colloidal solution is coated on transparent conductive oxide (TCO) substrate. The QDs are used to create electron-hole pairs as they transit from the lower state to the excited state. Electrons from the conductive band (CB) of QDs are injected into that of TiO_2. The counter electrons are Pt- and carbon-based materials coated on the TCO substrate. The electrolyte I^-/I_3^- is frequently used to transfer electrons between TiO_2 and the counter electrodes [23-24].



(a)



(b)

Fig.1. Working principle of typical QDSSCs [22].

As shown in Fig.1, the ground state of QDs is regenerated through electron donation from the electrolyte, which is commonly a redox system, such as polysulfide (S^{2-}/S_x^{2-}) redox couples. Another oxidation occurs in the photo-anode electrolyte interface in the electrolyte [25-27].

$$S^{2-} + 2h^{+} \rightarrow S \qquad (1)$$

$$S + S_{x-1}^{2-} \rightarrow S_x^{2-} (x = 2 - 5) \qquad (2)$$

On the counter electrode, the oxidized groups $S_x^{2-}$are re-reduced toS^{2-}. So, electrons migrate via the external load to complete the circuit [26].

$$S_x^{2-} + 2e^{-} \rightarrow S_{x-1}^{2-} + S^{2-} \qquad (3)$$

Subsequently, the voltage is generated by variation in Fermi levels between the electron in the photo-electrode and the redox potential of I^{-}/I_3^{-} in the electrolyte.

QDSSCs PERFORMANCE

To evaluate the performance of QDSSCs, the conversion efficiency (ŋ) and the incident photon-to-electron conversion efficiency (IPCE) will be introduced in this section. They are two important parameters.

The conversion efficiency (ŋ) can be expressed by [28]:

$$\eta = \frac{P_0(\max)}{P_{in}} = \frac{I_{sc} \times V_{oc} \times FF}{P_{in}} \quad (4)$$

Where, $P_0(\max)$ is the maximum of the output power; P_{in} is the input power; I_{sc} is the short current; V_{oc} is the open circuit voltage; FF is the fill factor.

The ideal value of FF is equal to 1, but the actual value is always below the ideal value.

The incident photon-to-electron conversion efficiency (IPCE) can be expressed by [29]:

$$IPCE(\lambda) = LHE(\lambda) \times \varphi_{inj} \times \varphi_{coll} \quad (5)$$

Where, $LHE(\lambda)$ is the light harvesting efficiency at a certain wavelength; φ_{inj} is the quantum yield of electron injection; φ_{coll} is the efficiency of collecting injected electrons at the back contact.

In order to compare the performances of different QDSSCs, the above data should be measured under uniform conditions. A standard solar spectrum of AM (air mass), 1.5G (global) is used. This spectrum is derived from the path length the light needs to travel through the atmosphere and reach the surface [29].

CURRENT RESEARCH OF QDSSCs

Many theoretical and experimental results of the QDSSCs proved that the conversion efficiency of QDSSCs is related to the QDs sensitized photo-anode, the counter electrode, the electrolyte, etc.

In this section, some current research results about the above three factors will be proposed.

QDs sensitized photo-anode

The major advantage of using QDs as sensitizers in QDSSCs is the size-dependent band gap, which is a characteristic of the optical properties of QDs. By varying the size of QDs, the light harvesting energy in the solar spectrum can be controlled [30].

The QDs employed in QDSSCs as sensitizers include $CuInS_2$ [31], PbS [32], CdS [33], CdSe [34], CdTe [35], ZnS [36], etc. Some research results have shown that CdS and CdSe QDs have been considered as an adequate sensitizer of QDSSCs.

Besides using different materials, changing the size of QDs, and improving the fabrication process are also important for improving the conversion efficiency of QDSSCs.

Chen et al.[37] reported that higher conversion efficiency was obtained when two different sizes of QDs were used in QDSSCs. Jung et al.[20] used a passivation layer (ZnS) in their research to improve the performance of QDSSCs located between TiO_2 and the electrolyte in CdS QDs. Hu et al.[38] fabricated multi-layer QDs using the chemical bath deposition (CBD) method to improve the conversion efficiency from 0.34% to 1.47%. Yu et al.[39] found the influence rules between the annealing temperature and the QDSSCs performance. A remarkable efficiency of 4.21% was obtained based on a TiO_2-CdSe/CdS-ZnS photo-anode (400°C, 300s calcination), polysulfide electrolyte and Cu_2S counter electrode achieving a power conversion efficiency of 4.21% under AM 1.5 G one-sun illumination. Fig. 2 shows the structure of QDSSCs.

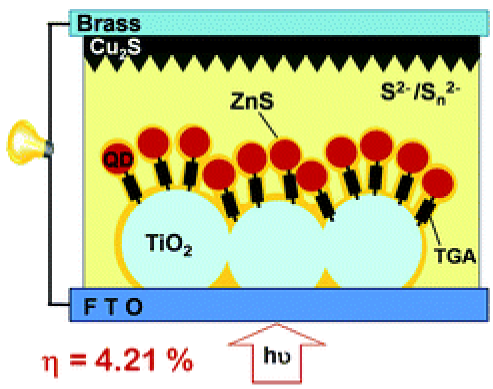


Fig.2. Sructure of QDSSCs (ŋ=4.21%, under AM 1.5 G one-sun illumination) [39].

These results show that higher conversion efficiency will be obtained using multi-layered QDs incorporated with a passivation layer.

Counter electrodes

Platinum (Pt) was commonly used as counter electrode in QDSCs and initial QDSSCs systems. It has been confirmed that Pt counter electrodes have low resistance and superior electro-catalytic activity for the iodide/triiodide redox couple in DSSCs [40]. Nevertheless, Pt counter electrodes have short lifetime and are expensive when iodide/triiodide is used as the electrolyte in QDSSCs. It was also reported that Pt counter electrodes have problems when a polysulfide redox couple is used as the electrolyte in QDSSCs. The conductivity and surface activity of the Pt electrodes decreased and their lifetime was short [33].

To figure out this problem, various non-Pt materials such as PbS, CuS and other materials have been investigated [41-44]. Kim et al.[41] have proved that doping of a PbS counter electrode with Mn ions in a QDSSC greatly improves the electro-catalytic activity compared with Pt and PbS counter electrodes, achieving an efficiency of 3.61%. Chandu et al.[44] used Mn-doped CuS as counter electrodes, which were in situ grown on FTO glass substrate by a facile CBD method and then tested for use in the QDSSCs system as counter electrodes catalyst without any post-treatment. Their research results proved that Mn-doped CuS counter electrodes showed better catalytic activity for polysulfide reduction than bare-CuS and Pt counter electrodes. Thus, the obtained Mn-doped CuS counter electrode delivered an efficiency of 5.46%, which was comparable to that of the bare-CuS electrode (4.29%) and Pt electrode (1.37%) tested under similar conditions.

Many research results also show that higher performances will be obtained using as counter electrodes compound materials containing metal sulfides and carbon.

Fig.3 shows the current density-voltage (J-V) characteristics of QDSSCs based on CuS, Mn-CuS and Pt counter electrodes.

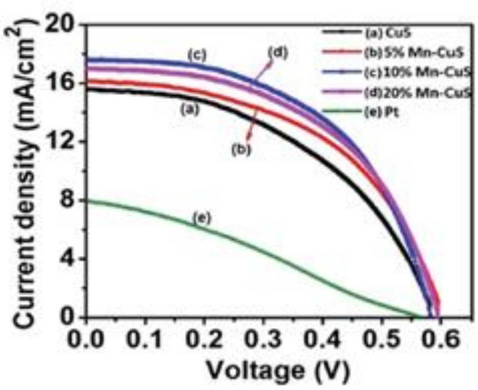


Fig.3. J-V characteristics of QDSSCs based on CuS, Mn-CuS and Pt counter electrodes [44].

Electrolyte

For QDSCs, Na_2S and S aqueous solution was commonly used as electrolyte, which has high surface tension. So it is difficult to permeate to the mesopores of TiO_2 film, which means that the photo-anode and electrolyte cannot be in close contact, and the performance of QDSCs is poor. To solve out this problem, alcohol was used to replace part of the aqueous solution, but its electrolytic dissociation is worse than that of the aqueous solution. Considering the permeability and the dissociation, Lee et al.[26] proposed an aqueous solution mixed with methanol (7:3). Sixto et al.[45], reported that electrolytes with a low concentration of S can inhibit the reaction between the QDs and the electrolyte, and the concentration was set as 1 mol/L. Li et al.[46] achieved a 3.2% conversion efficiency and 0.89 fill factor (FF) using methyl ammonium sulfide electrolyte, which was synthetized by themselves. Their research provided a novel method for electrolyte preparation.

Fig.4 shows the J-V characteristics of the TiO_2/TGA/CdS-3-based cell measured under one-sun illumination (AM 1.5G, 100 mW cm^{-2})

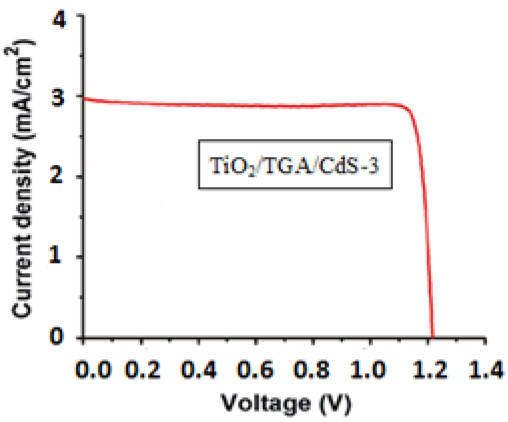


Fig.4. J-V characteristics of TiO_2/TGA/CdS-3-based cell measured under one-sun illumination (AM 1.5G, 100 mW cm^{-2}) [46]

Besides the above single electrolyte material used in QDSSCs, Karageorgopoulos et al.[47] employed two different electrolytes, namely, a quasi-solid electrolyte that contains a polysulfide (S^{2-}/S_x^{2-}) and a hybrid organic-inorganic (ICS-PPG230) material in ZnO/CdS/CdSe QDSSCs. The conversion efficiency of solar cells was enhanced from 1.2% to 4.5%. Their results showed that the quasi-solid electrolyte was more appropriate than the aqueous electrolyte.

ANALYSIS AND OUTLOOK

As a representative of the third generation solar cells, the QDSSCs are under intense investigation. Immense efforts have been devoted to improve the photovoltaic performance in the past few years. The conversion efficiency of QDSSCs was greatly enhanced and the best record of 7.04% has been recently reported [48].

However, the conversion efficiency of QDSSCs is still much lower than that of DSSCs (about 12%). And most of these current results are obtained only in the lab, and are not suitable for industrial production. In order to obtain higher conversion efficiency of QDSSCs, the future work should be focused on these following aspects:

1)develop a new material of semiconductor QDs with a large wavelength range of optical absorption, and improve the QDs loading onto TiO_2 to increase the light harvesting efficiency of QDs;

2)develop a new material and optimize the fabrication process of counter electrodes for improving the catalytic activity;

3)develop a novel organic polysulfide electrolyte and a solid-state hole conductor as electrolyte;

4)improve the long-term stability of the counter electrode for polysulfide electrolyte;

5)reduce the fabrication costs of QDSSCs.

CONCLUSIONS

In this paper, QDSSCs are reviewed from the aspects of working principle, performance, current research progress and developing trends. Immense efforts have been devoted to improve the photovoltaic performance of QDSSCs in the past few years, and some theoretical and experimental results of the QDSSCs have been achieved, but most of them were obtained only in the lab, and are not suitable for industrial production. Studies of many key issues for practical applications are currently under way. The conversion efficiency of QDSSCs is still much lower than that of 1st and 2nd generation PV systems, but they offer a potential novel developing direction of solar cells.

With the development of the material science, nanotechnology and the fabrication process, the conversion efficiency of QDSSCs will be highly improved in the near future. It is a promising 3rd generation PV systems.

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СОЛАРНИ КЛЕТКИ, СЕНСИБИЛИЗИРАНИ С КВАНТОВИ ТОЧКИ

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(Резюме)

Квантовите точки (QDs) се използват широко за подобряването на ефективността на превръщане при соларните клетки и за проектирането на трето поколение соларни клетки (QDSSC). Това се дължи на интересните им свойства (генерирането на множествени екситони, висок абсорбционен коефициент,подходяща забранена зона и горещ електронен пренос).Огромни усилия са положени през последните години за подобряването на фотоволтаичните характеристики на QDSSCs.Ефективността на конверсия на QDSSC е била значително подобрена, като по последни сведения най-добрия резултат е 7.04%. В настоящата работа тези соларни клетки са разгледани от гледна точка на работния принцип, работата им, съвременния напредък и тенденциите на развитие (QD-сенсибилизарни фото-аноди, противо-електродите и електролитите). Накрая са предложени някои решения за бъдеще развитие на QDSSC.