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Quantum Dot-sensitized Solar Cell Based on nano-TiO₂ Electrodes

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Abstract - Quantum dots-sensitized solar cell (QDSSC) is one of the third generation solar cell that is the most promising low cost, easy to manufacture and highly efficient solar cell. Compared to Dyesensitized solar cell (DSSC), quantum dots (QDs) of QDSSC has a narrow bandgap and possess excellent properties such as tunable band gaps, strong light absorption and high multiple electron generation. Titanium dioxide or titania (TiO₂) is an oxides semiconductor material that is frequently used as a photoanode in this photovoltaic system due to high stability under visible light illumination. TiO₂ is also known as a good photocatalyst and an excellent choice in environmental purification. The efficiencies of electron injection and light harvesting in QDSSC are affected by the nature, size morphology, and quantity of this nanomaterial. In this review, the concept and principles of the QDSSCs are reviewed. The preparation and fabrication method ofTiO₂ electrode in QDSSC are also discussed. It is worthwhile to know the architecture of TiO₂ in order to enhance the efficiency of QDSSC.

Keywords: Quantum dots-sensitized solar cell, titanium dioxide, quantum dots

Introduction

Sunlight continued to be harvested by technologies up to the early years of the industrial revolution. Since then, the temperature has risen by 0.6°C because of the global activities which cause the greenhouse effect whereby the quantity of carbon dioxide increases and eventually causing global warming (Du, Li, Brown, Peng, & Shuai, 2014; El Chaar, Lamont, & El Zein, 2011; Gong, Liang, & Sumathy, 2012). In recent years, renewable energy has attracted high interest due to these factors. As an alternative source of energy, the sun sends high quantities of light energy to the surface of the earth (Selinsky, Ding, Faber, Wright, & Jin, 2013). It is also completely renewable and definitely an abundant resource with rapidly declining conversion cost (Jun, Careem, & Arof, 2014). The energy provided by the sun for our planet is 10,000 times more than world demand whereby 10 % of the efficiency of the solar cell would fulfil global needs (Kouhnavard et al., 2014). A broad range of solar cell research is currently underway and they include dye-sensitized solar cell (Abdullah & Rusop, 2014), organic solar cell (Halim, 2012), silicon solar cell (Halim, 2012) and heterojunction solar cell (Church, Muthuswamy, Zhai, Kauzlarich, & Carter, 2013; Guo, Shen, Wu, & Ma, 2012).

The photovoltaic technology (PV) is a highly potential candidate for an alternative or renewable source of energy in the current market. PV can be classified into first, second and third generation solar cell. Solar cells based on silicon wafer, so-called first generation technology solar cell, make up

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the most number of solar cells present in the market and can reach as high a 27% solar cell efficiency (Green, 2002).Meanwhile solar cell utilized with inorganic film is the second generation solar cell which is cheaper to produce but has less than 14% solar cell efficiency (Jun, Careem, & Arof, 2013). Chronologically, the invention of the third-generation solar cell is to decrease cost by significantly increasing efficiencies as high as above 30% and at the same time maintain the economic and environmental cost advantage (Conibeer, 2007). Figure 1 shows the PV production per square meter against the efficiency of solar cell and the cost unit power.



Figure 1: Efficiency and cost projection for first (I), second (II) and third generation (III) (waferbased, thin films, and advanced thin film, respectively)

Dye-sensitized solar cell (DSSCs) is the first third generation which has attracted much attention due to low fabrication cost and high efficiency, flexibility in colour, shape and transparency (Golobostanfard & Abdizadeh, 2014). However QDSSCs is the further improvement of dye-sensitized solar cell (DSSCs) in boosting the overall efficiency by coupling synthesized inorganic quantum dots (QDs) semiconducting materials as sensitizers (Prabakar, Minkyu, Inyoung, & Heeje, 2010).QDSSC based on semiconductor nanocrystal has attracted attention as an alternative to DSSCs owing to their great stability, good absorption over wider wavelength range and multiple exciton generation leading to the production of power efficiencies (Z) that are much higher than DSSC (Xu, Zou, Yu, & Zhi, 2013; Yang, Chen, Roy, & Chang, 2011). All these unique characteristics of the QDSSC have raised high interest among researchers in renewable energy research field.

Despite all of these good characteristics of QDSSCs, the power conversion efficiency is still not as impressive as DSSCs mainly due to several reasons such as bad charges separation, less efficient photo excited electrons and unsuitable sensitizers (Li, Yu, Liu, & Sun, 2015). The electrode also plays a critically important role in contributing to the high efficiency of the QDSSC performance. Photo anode with high strong light scattering, efficient electron transport, high QD loading and quick electrolyte is of great importance to the QDSSC system (Zhou et al., 2014). The unique textural and structural characteristic of nanostructured material such as TiO₂, SnO₂ and ZnO has attracted much interest in the past decade(Malekshahi Byranvanda, 2013). The unique textural and structural characteristics are particle size distribution, specific surface area, morphology, crystallinity and crystal structure (Hu et al., 2014). This review paper is focused on TiO₂ as an electrode in QDSSC.

During the past decade, TiO_2 has become one of the most popular electrode materials and different methods apply to photovoltaic application and QDSSC specifically. In addition TiO_2 is an eco-friendly commercial product and has been known to be effective and is of great value(Liao et al.,

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2012). TiO₂ is a semiconductor with wide band gap known to be n-type. It has three crystalline phases which are anatase (tetragonal), rutile (tetragonal) and brookite (orthorhombic). The most stable phase is rutile TiO₂ whereby anatase and brookite TiO₂ are metastable and they can be converted into rutile phase at high temperature that is, around 750°C (Wang, He, Lai, & Fan, 2014). Single crystal anatase is reported to be more effective than rutile phase in photovoltaic application (Bet-moushoul, Mansourpanah, Farhadi, & Tabatabaei, 2016).

Basic principle of QDSSC

QDSSCs have similar configuration with DSSCs and the only difference is that QDSSCs uses inorganic semiconductor quantum dots (QDs) as light absorbing material instead of molecular dyes, onto the surface of a thin film of nano-TiO₂ electrode that acts as a working electrode (Song et al., 2014). Similar to DSSCs, in QDSSCs, excitons are formed in quantum dots whereby the charge separation occurs in the QD molecule layer upon the photoexcitation as electrons are injected from the QD excited state into the conduction band of the nano-TiO₂ and that eventually produces a photovoltaic effect as shown in figure 2.



Figure 2: Operating principle of QDSSC

QDs is restored through electron donation from the frequently used polysulfide electrolyte, which consist of (S^{2-}/S_x^{2-}) the redox system. The oxidized QD is then restored (hole is filled with electron) when it is reduced by S^{2-} from the electrolyte and in turn it is oxidized into S_x^{2-} that diffuses to the counter electrode.

$$S^{2-} + 2^{h+} \to S \tag{1}$$

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

The oxidized group S_x^{2-} are reduced to S^{2-} occur on the counter electrode.

$$S_x^{2-} + 2e^- \to S_{x-1}^{2-} + S^{2-} \tag{3}$$

In the electrolytes, voltage is generated in the Fermi levels between the electron in the photo electrode and the redox potential of I^-/I_3^- . I^- ion reduced to I_3^- at the counter electrode whereby platinum and carbon based materials coated on the substrate (Guo, Shen, Wu, Wang, et al., 2012; Lee & Chang,

2008; Yu, Lia, Qiu, Kuang, & Su, 2011). The efficiency of the solar cell can be determined by the equation below:

$$\eta = \frac{(J_{SC} \times V_{OC} \times FF)}{P_{in}},\tag{4}$$

where J_{SC} is the short circuit photocurrent density, V_{OC} is the open circuit voltage, FF is the fill factor and P_{in} is the power intensity of the incident light. The J_{SC} , V_{OC} and FF values can be calculated from the direct current density-voltage (J-V) curves.

TiO₂ as an electrode

The excellent properties of TiO₂such as good chemical stability, low cost production, high corrosion resistance, non-toxicity, high photocatalytic activities and good charge transport properties play an important role in the performance of QDSSC (Barbe et al., 1997; Kong, Chang, & Jang, 2014; Ou & Lo, 2007). TiO₂ nanostructure such as nanoparticles(Balis, Dracopoulos, Bourikas, & Lianos, 2013; Chen, Chappel, Diamant, & Zaban, 2001; Ito et al., 2007; Jung, Kim, Kim, Choi, & Ahn, 2012; Kongkan, Tvrdy, Takechi, Kuno, & Kamat, 2008; Zaban, Mic'ic, Gregg, & Nozik, 1998; Zhang et al., 2009), nanotubes (Chen et al., 2009), nanorods (Gonfa et al., 2014), nanowires (Nikhil, Thomas, Amulya, Mohan Raj, & Kumaresan, 2014; Sun et al., 2012) and nanoflower (Yu, Li, Liu, Cheng, & Sun, 2014b) have been widely recognised as excellent photo anodes in QDSSC. The size of the TiO₂ building units, apparently in nanometer scale, highly influences the performance of QDSSC (Kavitha, Gopinathan, & Pandi, 2013). Table 1 shows the example of QDSSC and the solar cell efficiency performance based on nano-TiO₂ as an electrode.

Table 1: Example of QDSSC and the solar cell efficiency performance based on nano-TiO₂ as an electrode.

TiO2	Sensitizer	Counter electrode	Efficiency	Reference
TiO ₂ nanoparticle	CuInS ₂	Cu ₂ S	1.05%	(Gong et al., 2012)
TiO ₂ sol	CdSe	Cu ₂ S/CNT	1.05%	(Golobostanfard & Abdizadeh, 2014)
TiO ₂ nanoparticle	CdSe	Pt	3.65%	(Prabakar et al., 2010)
TiO ₂ nanotube	$CdSe_{x}Te_{1-x}$	Pt	0.588%	(Xu et al., 2013)
TiO_2 beads	CdS/CdSe	Cu_2S	4.33%	(Zhou et al., 2014)
TiO ₂ nanoparticle	CdSe	Pt	2.23%	(Song et al., 2014)
TiO ₂ nanoparticle	CdS	Pt	1.15%	(Lee & Chang, 2008)
Mesoporous spherical TiO ₂ powder	CdS/CdSe	Pt	0.29%/0.34 %	(Kong et al., 2014)
TiO_2 sol gel	InP	Pt	-	(Zaba, Mic´ic´, Gregg, & Nozik, 1998)
TiO ₂ nanoparticle TNT/TNP	ZnS, CdS, CdSe CdSe	Pt, CoS, CuS	2.7%	(Balis et al., 2013) (Kongkanand, Tvrdy, Takechi, Kuno, & Kamat, 2007)
TNP	CdS	Pt	-	(Jung et al., 2012)
TiO ₂ nanoparticle	CISe	Cu ₂ S	4.3%	(Yang et al., 2013)
TiO ₂ nanoparticle	CdS/CdSe/ZnS	NiS	2.97%	(Kim et al., 2014)

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Table 1: Example of QDSSC and the solar cel	l efficiency perforn	nance based on n	ano-TiO ₂ as an
electrode (continued).			

TiO2	Sensitizer	Counter electrode	Efficiency	Reference
TiO ₂ nanoparticle	CdS/CdSe	Pt/Cu ₂ S/Graphite/	1.2%	(Jun, Careem, &
		Carbon soot/		Arof, 2014)
		Orida (PCO)		
TiO, nanoparticle	SnSea	Oxide (KOO)	0.12%	(Yu et al. 2012)
TiO ₂ nanoparticle	SnSc ₂	Pt	<0.12%	(Miyauchi 2011)
TiO_2 nanoparticle	CdS	Pt/CuS	<0.1% 1.95%	(Inigation, 2011)
TiO ₂ nanonarticle	AgaSe	Pt	3.6%	(Tubtimtae Lee &
110 ₂ nulloparticle	116200	Ĩt	5.070	Wang, 2011)
TiO ₂ nanoparticle	CdS	NiS	3.6%	(Li, Yang, Zhang,
				Zhang, & Li, 2014)
TiO_2 nanorod	CdS/PbS	Pt	2.0%	(Jiao, Zhou, Zhou,
T'O meneratiale	CI-C	C C	1.050/	& Wu, 2013)
110_2 nanoparticle	$CumS_2$	Cu_2s	1.85%	(Peng, Liu, Shu, Chan & Chan
				2013
TiO ₂ nanodendrite	CuInS	CuaS	1 26%	(Peng Liu Zhao et
arrav			1.2070	al., 2013)
TiO ₂ nanoparticle	CdS/CdSe	Brass plate	0.45%	(Shen et al., 2015)
TiO_2 hollow sphere	CdS/N719	Pt	4.66%	(Cui et al., 2015)
TiO ₂ nanowire	PbSe	-	-	(Győri, Kónya, &
				Kukovecz, 2015)
TiO ₂ nanoparticle	CdS	CoS2/Pt	2.27%	(Punnoose, Kim,
				Srinivasa Rao, &
				Pavan Kumar,
T:O manatuha	C4C C.	D4		2015) (Caliban Smith
110_2 nanotube	Cas _{0.54} se _{0.46}	Pl	-	(Gaknar, Simin,
				Chidambaram
				2015)
TiO ₂ nanoparticle	$CdS_xSe_{1-x}/$	Cu _{1 8} /CuS	3.26%	(Li et al., 2015)
- 1	Mn-CdS	1.0		
TiO ₂ nanoparticle	CdS/CdSe/ZnS	NiS	3.03%	(Gopi, Srinivasa
				Rao, Kim,
				Punnoose, & Kim,
			2 400/	2015)
$11O_2$ nanorods arrays	CdSe/Mn-CdS	Cu _{1.8} S/CuS	2.40%	(Yu, L1, L1u,
				Cheng, & Sun, 2014_{2}
				2014a)

Preparation of TiO_2 as an electrode in QDSSC In QDSSC, TiO_2 nanoparticles (example like commercial P25 nanoparticles) have been extensively studied as a photoanode due to their special characteristics as mentioned before (Zhou et al.,

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2014).Anatase, rutile and brookite are the three crystalline form of TiO_2 whereby anatase is the most preferable in solar energy conversion. This is due to the ability to avoid charge recombination and efficient electron transport in photoanode (Byranvana, Bazarganb, & Kharat, 2012). In recent years, a lot of research have gone into preparing TiO_2 among them are in achieving low cost production and making them easily reproducible by using a simple method which is eventually imperative for the industrial manufacture of QDSSC (Zhang et al., 2009).The methods in preparing TiO_2 such as the hydrothermal method (Gopinathan, & Pandi, 2008; Vijayalakshmi & Rajendran, 2012; Wu et al., 2013), the sol gel method (Behnajady & Eskandarloo, 2013; Guo, Liu, Hong, & Jiang, 2005; Sabataitytė, Oja, Lenzmann, Volobujeva, & Krunks, 2006) and anodization (Tang et al., 2008) have been studied extensively in order to produce excellent characteristics of TiO_2 as a photoelectrode.

Sol-gel method

The sol-gel method for TiO_2 synthesis is a very useful tool for photo-induced molecular reaction due to the special variables such as particle size, incident light, phase composition and convenient preparation method (Karami, 2010). Titanium (IV) isopropoxide (TIPP) is usually used as a starting material in this method(Manoharan & Venkatachalam, 2015; Zeng, Chen, Su, Li, & Feng, 2014). The mixture will undergo an aging period and it is kept in the oven to obtain the colloidal solution (Zeng et al., 2014). Next the solution is dried and calcined to get TiO₂ nanocrystal powder (Hu, Tang, He, Lin, & Chen, 2014; Laranjo et al., 2014; Zhu, Zhang, Gao, & Cao, 2000). Figure 3 shows TiO2 nanoparticles prepared by the sol gel method at different levels of concentration.



Figure 3: SEM surface images of three different kinds of TiO₂ concentrations of (a) 17 wt.%, (b) 20 wt.%, and (c) 24 wt.% films on the SnO2:F glass after sintering process(Lee et al., 2009).

Hydrothermal method

Hydrothermal method is one of the most popular methods to prepare TiO_2 nanostructure. Other than TiO_2 nanoparticle, other nanostructures such as nanotube and nanorod can also be synthesized via the hyrothermal method (Lee, Lee, Rhee, & Park, 2014). One of the suggested methods is TIPP where it is mixed and stirred with nitric acid, ethanol and distilled water through the sol-gel method. The product produced from the sol-gel method will undergo hydrothermal treatment in the teflon-lined autoclave to produce TiO_2 powder and the powder will receive further treatment for calcination to

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achieve the desired size and crystallinity (Manoharan & Venkatachalam, 2015). Figure 4 shows SEM images of TiO_2 nanorod arrays formed by the hydrothermal method by optimization of the seed layer.



Figure 4: SEM images of TiO₂nanorod arrays grown by hydrothermal method on (a) bare FTO,
(b) FTO immersed in 0.05 M TiCl₄ solution, (c) FTO immersed in 0.1 M TiCl₄ solution,
(d) FTO immersed in0.15 M TiCl₄ solution, (e) FTO immersed in 0.2 M TiCl₄ solution,
respectively (Wang et al., 2013)

Electrochemical method

The Electrochemical method is an impressive technology to develop the nanotube or nanoporous layer as an electrode especially in QDSSC. TiO₂ nanotube can be formed by an anodization of the titanium whose capability is strongly influenced by the variation of parameters. The quality and ability of TiO₂ nanotubes also depends on their very own properties such as crystallite size, morphology and the lattice strain. Yulian Zhang et al.,(2015)reported the frequent used of ammonium fluoride (NH₄F) as an electrolyte and indicated that high NH₄F concentration is beneficial to the growth of ribs around the nanotubes. Figure 5 shows the FESEM images of surface morphologies and cross-section of TNTs obtained in electrolytes with different NH₄F concentrations. Meanwhile, Munirathinam, Pydimukkala, Ramaswamy, & Neelakantan (2015) reported on the development of TiO₂ nanotubes by the anodization process using the two electrode system whereby titanium was used as anode and a stainless steel plate as a cathode at a specific distance. In this research, two different electrolytes which are hydrofluoric acid, HF (acidic medium) and sodium sulfate, Na₂SO₄ (neutral) were used and then followed by annealing at 450°C for 2h. The result clearly indicated nanotubes formed from the neutral bath are four times longer than the ones synthesized from the acidic bath.

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Figure 5: FESEM images of surface morphologies and cross-section of TNTs obtained in electrolytes with NH₄F concentrations of (a) (d) 0.2 wt%, (b) (e) 0.4 wt%, (c) (f) 0.6 wt%, respectively (Zhang et al., 2015)

Approach in improving TiO₂photoanode in QDSSCs

Although QDSSC raised tremendously high attention among researchers in order to improve solar cell performance, energy conversion efficiency remains under 10% as reported in figure 1. A lot of work has been done in the approach to improve QDSSC. One of the important approaches to increase the energy conversion efficiency yield is the architecture of the photoanodes. This is because photoanode material like TiO_2 has wide band gap (3.20 eV for anatase and 3.02 eV for rutile TiO_2) that limit its usage at UV light region(Maheswari & Venkatachalam, 2015). Other than that, optimal nanoparticle interconnection and pores size can control the charge carrier transport to ensure an efficient electrolyte penetration (Yacoubi, Samet, Bennaceur, Lamouchi, & Chtourou, 2015).

Among studies that have been done recently are doping TiO_2 electrode with Ni (Maheswari & Venkatachalam, 2015), Au (Liu et al., 2014), Co (Brigham, Achey, & Meyer, 2014; C. Wang et al., 2014), Fe (Wang et al., 2014) and Mn (Wang et al., 2014). Doping TiO_2 with impurities dopants will broaden the use of the PV to the visible region and at the same time provide a good surface for the deposition of QDs (Maheswari & Venkatachalam, 2015; Yacoubi et al., 2015). The dopants also act as a light harvesting material which means the light will be scattered and trapped in order to increase the effective path length of incident light of the absorption of the semiconductor (Liu et al., 2014). Some dopant like Fe have been reported as having the ability to increase charge carrier density of TiO_2 leading to good carrier transportation and separation and relatively long electron lifetime(Wang et al., 2014).

Beside dopants, surface modification also play an important role in order to avoid or reduce recombination of excited electron whereby it is a major problem in QDSSC(Kim et al., 2012). TiCl₄is usually applied on the substrate at multiple times of immersion, followed by annealing at 450° C for 30 min before depositing TiO₂ paste (Kim et al., 2012). Nevertheless, TiCl₄ treatment decreases average

pore size whereby it can lower the recombination and increase the current (Guo et al., 2014). Recently, a study focused on the treatment of TiO_2 hydrolysed by hydrochloric acid (HCl) in preparation of $TiCl_4$ stock solution has been reported for the synthesis of nanosized crystalline TiO_2 (Lee & Yang, 2005). The result from this study showed that the brookite phase was transformed to the rutile phase with increase reaction time, while through a heat treatment, it was transformed to rutile via anatase phase (Lee & Yang, 2005).

Previously, there were so many efforts made to optimize TiO₂ structures to enhance QDSSC performance. The first effort made was by creating large pore size distribution of TiO_2 to make the loading process of quantum dots much easier due to the small size of quantum dots (Salant et al., 2012). Secondly, the surface area of TiO_2 was increased in order to increase quantum dots loading. Moreover, high surface area of photoanaode may improve the quantum dots coverage and retard unnecessary interface recombination (Song et al., 2012). Thirdly, an additional layer of coating was also added to improve the electron transport path in order to enhance the QDSSC performance. However, previous researches reported that an additional layer of TiO_2 can hardly balance the required qualities of TiO₂ (Wu et al., 2015). Therefore, further studies on optimization of TiO₂ should be done to meet the demand in QDSSC. For example, Wu et al. (2015) designed a multi-dimension titanium dioxide made up of mesoporous nanoribbons consisting of oriented aligned nanocrystal. This impressive development resulted in increased surface area of TiO_2 that led to a high photocurrent efficiency of 4.15%. Meanwhile, in another study an attempt has been done by doping TiO₂ nanocrystal with two dimensional graphene in order to improve the photovoltaic performance owing to the graphene unique characteristics such as good thermal conductivity, good mobility charge carriers and specific surface area (Chen, Tuo, Rao, & Zhou, 2014). The incorporation of graphene with TiO₂ increase the photocurrent efficiency by 37% compared to the pure TiO₂ and eventually increased the ODSSC performance.

Conclusions and future directions

The review on TiO₂ as a working electrode in QDSSC demonstrated high potential inorder to increase energy conversion efficiency in a novel QDSSC system. The study of photoanode configuration is critically important because the significance can be of high impact particularly in providing high QD loading, strong light scattering, quick electrolyte diffusion and efficient electron transport (Zhou et al., 2014). Different nanocrystal structure such as nanotubes, nanorods and nanowire have been developed whereby particular control is given to recombination and this eventually improves PV performance. Currently, a lot of studies are focused on developing low cost high ability nanocrystal material for PV application and this will no doubt raise its potential when developed and applied in the academia and industry. The low cost nanocrystal material used make the current price of QDSSC cheaper than DSSC (\$3/Wp–\$4/Wp) and silicon solar cell (\$3/Wp) (Kalowekamo & Baker, 2009). Future work should be focused on improving the solar cell efficiency as mentioned in this review paper. Many modifications on QDSSC have been developed however, they are still in their early stages and many other new developments can be done in order to improve the efficiency, robustness and potential of the thin-film-type material. No doubt, as the understanding of the topic continues, more possible ideas can be conceived to improve QDSSC potential.

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