STATUS AND PERSPECTIVES OF DYES USED IN DYE SENSITIZED SOLAR CELLS

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ABSTRACT

Dye Sensitized Solar Cells (DSSC) is promising alternative for the development of new generation of solar cells. Currently DSSC is using inorganic (Ruthenium) and organic (natural and synthetic) dyes as a sensitizer. The natures of these dyes together with other parameters have resulted in varying performance. The use of natural pigments have become a viable alternative to expensive and rare Ruthenium dyes because of its low cost, easy attainability, abundance in supply of raw materials and no environment threat. Various components of plants such as fruits, flower petals are being used as natural sensitizers. In this review, commonly used dyes in DSSC have been studied. The principles and components of DSSC are explained and recent progress on inorganic and organic dyes is reviewed. This study will be useful for the researchers working in this emerging technology.

Keywords: dye sensitized solar cells, natural dyes, pigments, Ruthenium dyes.

1. INTRODUCTION

DSSC are the devices for the conversion of visible light into electricity and have attracted much attention as lowcost photovoltaic cells and become a rapidly expanding field with potential applications. In DSSC, the dye as a sensitizer plays a key role in absorbing sunlight and transforming solar energy into electric energy. DSSC photo electrochemical cells have been subjected of a large number of experimental investigations since 1991. DSSC separates the optical absorption and charge separation process by associating a sensitizer with a wide band gap semiconductor of nanocrystalline morphology. According to Kong F et al. conversion efficiency records of 8.12%, 10.10%, 10.40% and 9.90% were announced at Energy Research Centre of The Netherlands, Ecole Polytechnique Federale de Lausanne, Sharp Corporation and Arakawa group, respectively for solar cells with aperture area of $1-5 \text{ cm}^2$. Apparently, the use of high film TiO₂ electrodes led to the highest efficiency till date, standing at 11.10 % (Chiba et al., 2006). Ruthenium based complexes sensitizers have been widely used because they have better efficiency and high durability. However, these advantages are offset by their high cost, their complicated synthetic routes and the tendency to undergo degradation in the presence of water. Also noble

metals are considered as resources that are limited in amount, and their production is costly. Efforts are continually being undertaken to improve the performance of DSSC and hence the competitiveness of this technology in the world market.

2. DSSC OPERATION

DSSC structure is shown in figure 1.It consists of two electrodes one made up of a nanocrystalline semiconductor material having wide band gap (like TiO₂) sensitized with a complex dye and other is platinum doped or graphite coated working electrode. An aqueous electrolyte solution fills the gap in between two. The working principle of DSSC involves some key processes viz. light absorption, charge separation and charge collection. Under sunlight illumination, the dyes will absorb photons (light) and become photo excited. The absorbed dve molecules will inject electrons into the TiO₂ working electrode and thus become oxidized. Charge separation is attained across the semiconductor interface where an electron is located in the TiO₂ and a hole is located in the oxidized dye molecule. The electrons will then percolate through the porous network of TiO₂ and eventually reach the back contact of the working electrode where charge collection and charge extraction occur. The extracted charge can subsequently perform electrical work in the external circuit and eventually return to the counter electrode where reduction of the redox mediator takes place. The liquid redox electrolyte will complete the circuit by reducing the oxidized dye.



Figure 1 Dye Sensitized Solar Cell Structure

3. DSSC COMPONENTS

DSSC converts visible light into electricity based on sensitization of wide band gap semiconductors and is primarily consists of dye, photo electrode, electrolyte, counter electrodes and substrates glass with the transparent conductive oxide (TCO) layer. The optimization of each of them is of great importance in order to improve the overall efficiency.

3.1 Conductive Glass or Substrates

Clear conductive glass are commonly used as substrate because of their relative low cost, availability and high optical transparency in the visible and near infrared regions of the electromagnetic spectrum. Conductive coating (film) in the form of thin TCO is deposited on one side of the substrate. This layer is crucial since it allows sunlight penetrating into the cell while conducting electron carriers to outer circuit. TCO substrates are adopted, including Fluorine-doped or Indium-doped tin oxide (FTO or ITO). The conductive film ensures a very low electric resistance per square. Typical value of such resistance is 10-20 Ω per square at room temperature. The nanostructure wide band gap oxide semiconductor (electron acceptor) is applied, printed or grown on the conductive side.

3.2 Photo Electrode

The photoelectrode or working electrode in a DSSC consists of a nanostructure semiconductor material, attached to a transparent conducting substrate. The most extensively utilized semiconductor material is TiO₂ (anatase band gap 3.2 eV). TiO₂ is an inexpensive, nontoxic abundant material. The electrode consists of interconnected nanoparticles, in the size of 15-30 nm. They form a transparent porous electrode, with a typical thickness of 10-15 µm. The deposition methods predominantly used for the film preparation are screen printing and doctor blading. Both techniques involve the deposition of viscous colloidal TiO₂ paste onto a substrate prior to the sintering process. Sintering is usually performed at temperatures of 450-500°C. The high temperature results in electrical interconnection between the nanoparticles and ultimately forms the nanostructure porous electrode. The dye sensitization is performed by immersing the electrode into a dye solution for a given time.

3.3 Counter Electrode

Counter electrode is another critical component of a DSSC. The main purpose of the counter electrode is the reduction of triiodide. The reduction reaction rate at the cathode is important because after the reduction of triodide, iodide is formed which is used to regenerate the oxidized dye molecule at anode side of the cell. For having a feasible cell, reaction at the anode side must be slow and reaction at the cathode side which is the counter electrode must be fast. Counter electrode is near the equilibrium potential of the redox couple but anode side is far from the equilibrium potential. This phenomenon

creates a voltage difference in the DSSC causing triiodide reduction. Although many materials like platinum, carbon, graphite, conductive polymers are used as counter electrode, platinum is the still catalyst of choice. Platinum is a better catalyst for iodide/triiodide redox couple. Also light reflection value of platinum is also higher than carbon coating which causes reflection of more light into the cell.

3.4 Electrolytes

The electrolyte plays a very important role in the DSSC by facilitating the transport of charge between the photo electrode and counter electrode. The ideal electrolyte solvent is one that has very low viscosity, negligible vapor pressure, high boiling point and high dielectric properties. From industrial perspective, factor such as (chemical inertness). robustness environmental sustainability, and easy processing are also important. At present the most successful redox mediator used in DSSC consists of a liquid electrolyte containing the redox couple iodide/triiodide. The redox electrolyte consists of iodides, iodine and often additional additives. Ionic liquids are a promising alternative electrolyte providing advantages such as non volatility, high thermal and chemical stability and excellent ionic conductivity. To find a superior redox couple is one of the main challenges for future DSSC research.

3.5 Dyes

The sensitizing dye is responsible for the capture of sunlight in DSCC. Due to the central role of the sensitizing dye in DSCC, much effort is invested in the synthesis and investigation of novel dyes. The ideal sensitizer should be stable and be able to attach to the surface of the electron-conducting material. They should able to absorb light at all wavelengths (λ) below 920 nm, thus spanning across the range of light that reaches the Earth's surface, and maximizing efficiency. Dyes are a critical component of DSSCs and it is no surprise that they have been a subject of intense interest.

4. REVIEWS OF DYES USED IN DSSC

The dyes used in DSSC are divided into two types: organic and inorganic dyes according to the structure. Inorganic dyes include metal complex, such as polypyridyl complexes of Ruthenium and Osmium, metal porphyrin, phthalocyanine and inorganic quantum dots, while organic dye includes natural and synthetic organic dyes.

4.1 Inorganic Dyes

In recent years, Ruthenium (Ru) dyes are characterized by their high efficiency, expensive cost as well as their difficult purification and have achieved the best results. To date, DSSCs with Ru bipyridyl complexes (N3 and N719) and the black Ruthenium dye as photo sensitizers have achieved power conversion efficiencies up to 11.2% and 10.4%, respectively (Nazeeruddin et al., 2005 ; Nazeeruddin et al., 2001), compared to just 1% twenty years ago. Further to superior light harvesting properties and durability, a significant advantage of these dyes lies in the charge transfer transition through which the photoelectric charge is injected into TiO₂. In Ruthenium dyes, this transfer takes place at a much faster rate than the back reaction, in which the electron recombines with the oxidized dye molecule rather than flowing through the circuit and performing work. The efficiency for different Ru complex such as N3, N712, N719, Z910, K19, N945, K73, N621, Z907, Z955, HRS-1 and Black dye is 10.0%, 8.2%, 11.2%, 10.2%, 7.0%, 9.6%, 9.0%, 9.6%, 7.3%, 8.0%, 9.5% and 10.8% respectively.

Ruthenium dyes are currently considered the best dyes for the production of efficient DSSC. By implementing such improvements, the efficiency of a DSSC can dramatically improve and efficiency values of 10-11% have been reported in the literature. However, the noble metal Ru is a limited resource and is expensive. Due to the high price of Ruthenium, other metal centers such as Copper and Zinc porphyrins are also gaining interest (Alibabaei et al., 2010). Pendant ligands play a huge role in the efficiency due to their affect on the anchoring efficiency of the dyes, their stability in solution and on the semiconductor surface (preventing aggregation) as well as playing a big role in reducing recombination. Traditionally carboxylic groups have been used to anchor dyes to the surface but it has been recently shown that phosphonic acid groups might be better suited for the role (Bae & Choi, 2005). The use of hydrophobic side chains like that of Z-907 increases the long term stability of the dyes in solution as well as on the surface of titania which is partly due to the prevention of water adsorption on the titania surface (Wang et al., 2003). The grand challenge as well as the holy grail of this field is to extend the absorption spectra such that more light and thus more current is generated. Table 1 shows the status review of Photo electrochemical parameters of Ruthenium based dye sensitizers till date.

4.2 Organic Dyes

DSSC sensitized with organic dyes have relatively lower power conversion efficiencies than those sensitized with metal complexes. However pure organic dyes have many advantages for their application in DSSC such as lower cost, high absorption coefficient and easy control of redox potential. In order to obtain even cheaper dyes for DSSC, metal free organic dyes are strongly desired. Metal free organic dyes offer superior molar extinction coefficients, low cost, and a diversity of molecular structures. Recently, novel photosensitizers such as coumarin, merocyanine, cyanine, indoline, hemicyanine, triphenylamine, dialkylaniline, phenothiazine, tetrahydroquinoline, and carbazole based dyes have achieved solar to electrical power conversion efficiencies up to 5–9%.

Mosurkal et al. studied various commercially available organic dyes with catechol groups, triphenylmethyne

dye, anthraquinone dye and xanthene dye, as sensitizers for dye sensitized solar cells (Mosurkal et al., 2004). The absorption maxima of these dyes ranges from 470 to 570 nm with cut off wavelengths at around 700 nm which makes them potential candidates for light harvesting in dye sensitized solar cells. All dyes showed quasireversible wave with oxidation potentials ranging from 0.6 to 0.9 V. The short-circuit photocurrent density (I_{sc}), the open-circuit photovoltage (V_{oc}), the fill factor (FF) and the overall energy efficiency (η) were estimated to be 3 mA/cm², 443 mV, 33% and 0.74%, respectively.

Wang et al. demonstrated that co sensitization of plural organic sensitizer dyes resulted in wider and higher photocurrent action spectrum than the sensitization by single dye. By optimizing the components of dye solutions the power conversion efficiency of 6.5% was obtained which was one of the highest efficiency to date for DSSC based on co sensitization of plural organic application dyes. of novel The cyanine trimethylcyanine pentamethylcyanine derivative, derivative and their mixture as a photosensitiser were also investigated by Cai group (Li et al., 2005). The highest photoelectric conversion of 3.4% was obtained for DSSC based on their mixtures.

4.3 Natural Dyes

The use of expensive Ruthenium metals, derived from relatively scarce natural resources corresponds to a relatively heavy environmental burden. Hence, it is necessary to use natural dyes as alternative sensitizers with appreciable efficiencies. Natural dyes provide a viable alternative to expensive organic based DSSC. Various fruits, flowers vegetables have been tested over the last decades as suitable sensitizers. Natural dyes are exclusively used for educational purposes representing a low cost and environmental friendly alternative to conventional Ruthenium sensitizer. Extracted dyes might also be a good starting point to evaluate to decide which dye classes are potentially interesting for sensitization. Plant pigmentation occurs due to the electronic structure of the pigment interacting with sunlight to alter the wavelengths that are either transmitted or reflected by the plant tissue. Anthocyanin is a group of naturally occurring phenolic compounds responsible for the colour of many flowers, fruits (particularly in berries) and vegetables. Sometimes they are present in other plant tissues such as roots, tubers and stems. Anthocyanins various plants give different sensitizing from performances. They have been of recent interest to research because of their ability to absorb light and convert it into electrons. The use of natural pigments as sensitizing dye for the conversion of solar energy into electricity is interesting because, on one hand it enhances the economical aspect and on the other, it has significant benefits from the environmental point of view. There remains the need for alternative sensitizers for use with TiO₂ based photovoltaic devices, especially due to the high cost of Ruthenium complexes and the long-term unavailability of these noble metals. Nonetheless, the natural dyes found in flowers, leaves, and fruits can be extracted by simple procedures. Due to their low cost, non-toxicity, and complete biodegradation, natural dyes have been a popular subject of research. Therefore, investigation of low cost, readily available dyes as efficient sensitizers for DSCC still remains a scientific challenge.

Cherepy et al. reported that a photo electrochemical cell utilizing flavonoid anthocyanin dyes extracted from blackberries was shown to convert sunlight to electrical power at an efficiency of 0.56% under full sun (Cherepy et al., 1997). The open-circuit voltages of 0.4-0.5 V, and short-circuit photocurrents of 1.5-2.2 mA/cm² were remarkable for such a simple system and suggest efficient charge carrier injection.

Olea et al. investigated the functioning of black berry extract to sensitize TiO₂ for light absorption (Olea et al.,1997). The black berry extract sensitized TiO₂ exhibited an increase in the photocurrent response indicating excess generation of photoelectrons due to light absorption by the extract. The maximum current (I_{max}), the maximum voltage (V_{max}), and the overall energy efficiency (η) of dye-sensitized solar cell for a 4 cm² active area cell were estimated to be 4 mA, 300 mV, and 1%, respectively.

Garcia et al. presented that fresh extracts of chaste tree fruit, mulberry and cabbage-palm fruit were employed as TiO_2 sensitizers in thin-layer sandwich-type photo electrochemical solar cells (Garcia et al., 2003). Conversion of visible light into electricity was accomplished with natural sensitizers resulting in I_{sc} and V_{oc} values similar to those obtained employing traditional

synthesized dyes. The short-circuit current (I_{sc}) obtained from 0.5 cm² of dye-sensitized solar cells using the fresh extracts of chaste tree fruit, mulberry and cabbage-palm fruit as sensitizers were 1.06, 0.86 and 0.37 mA, respectively and the open-circuit voltage (V_{oc}) obtained from 0.5 cm² of DSSC using the fresh extracts of chaste tree fruit, mulberry and cabbage-palm fruit as sensitizers were 390, 422 and 442 mV respectively.

Hao et al. reported that DSSCs were assembled by using natural dyes extracted from black rice, capsicum, erythrina variegata flower, rosa xanthina, and kelp as sensitizers (Hao., 2006). The short circuit current (I_{sc}) from 1.142 mA to 0.225 mA, the open-circuit voltage (V_{oc}) from 0.551 V to 0.412 V, the fill factor from 0.52 to 0.63, and maximum power (P_{max}) from 58 μ W to 327 μ W were obtained from the DSSC sensitized with natural dye extracts. In the extracts of natural fruit, leaves and flower chosen, the black rice extract performed the best photosensitized effect, which was due to the better interaction between the carbonyl and hydroxyl groups of anthocyanin molecule on black rice extract and the surface of TiO₂ porous film.

Fernando et al. (Fernando et al., 2008) studied dyes extracted from tropical flowers as possible sensitizers for TiO_2 by assembling DSCC (Fernando et al., 2008). Photocurrent densities ranging from 1.1 to 5.4 mA/ cm² are obtained with photo voltages ranging from 390 to 410 mV. The overall efficiency and fill factor of these cells varied from 0.2 to 1.1 and 0.53 to 0.64 respectively.

Dye	J _{sc} ma/cm ²	V _{oc}	FF	η (%)	Reference
N3	18.20	720	0.730	10.00	Li et al.
N719	17.73	846	0.750	11.18	Lu et. al.
Black Dye	20.53	720	0.704	10.40	Wang et al.
Black Dye	20.90	736	0.722	11.10	Wang et al
Z907	13.60	721	0.692	6.80	Li P Hironori
Z907	14.60	722	0.693	730	Hironori et al
K8	18.00	640	0.750	8.64	Hironori et al
K19	14.61	711	0.671	700	Yang et al
N945	16.50	790	0.720	9.60	Weon et al
Z910	17.20	777	0.764	10.20	Bandaranayake
K73	17.22	748	0.694	9.00	Panitat et al
K51	15.40	738	0.685	7.80	Wang et al

Table 1 Photo electrochemical parameters of Ruthenium based dye sensitizers

Dye	Jsc(ma/cm ²)	V _{oc} (V)	FF	η(%)	Ref.
Festucaovina	1.189	0.548	0.699	0.46	(Martinez et al., 2012)
Tageteserecta	2.891	0.475	0.606	0.8	
Bougainvillea spectabilis	2.344	0.26	0.738	0.46	
Punicagranatum peel	3.341	0.716	0.776	1.86	
Rosella	1.63	0.40	0.57	0.37	(Wongcharee et al., 2007)
Blue pea	0.37	0.37	0.33	0.05	
Mixedrosella blue pea	0.82	0.38	0.47	0.15	
Black rice.	1.14	0.55	0.55	-	(Hao et al., 2003)
Bixin	1.10	0.57	0.59	0.37	(Gomez-Ortiz et al.,2009)
Crocetin	2.84	0.43	0.46	0.56	(Yamazaki et al., 2007)
Crocin	0.45	0.58	0.60	0.16	
Fruit of calafate	6.20	0.47	0.36	-	(Polo et al., 2006)
Syrup of calafate.	1.50	0.38	0.20	-	
Skin of jaboticaba.	7.20	0.59	0.45	-	
Red Sicilian orange	3.84	0.34	0.50	-	(Calogero et al., 2006)
Purple eggplant extract	3.14	0.35	0.40	-	
Dragon fruit	0.20	0.22	0.30	0.22	(Ali et al., 2010)
Pomegrante	0.20	0.40	0.45	1.50	(Bazargan, 2009)
Red turnip	9.50	0.43	0.37	1.70	(Calogero, 2010)
Wild Sicilian prickly pear	8.20	0.38	0.38	1.19	
Bougainvillea	2.10	0.30	0.57	0.36	
Shisonin	3.56	0.55	0.51	1.01	(Kumara et al., 2006)
Shisonin and Chlorophyll	4.80	0.53	0.51	1.31	
Chlorophyll	3.52	0.43	0.39	0.59	
Hibiscus surattensis.	5.45	0.59	0.54	1.19	(Fernando et al., 2008)
Sesbania grandiflora.	4.40	0.41	0.57	1.02	
Begonia	0.63	0.54	0.72	0.24	(Huizhi Zhao et al., 2011)
Violet Bougainvillea glabra.	1.86	0.23	0.71	0.31	
Red Bougainvillea spectabilis	2.29	0.28	0.76	0.48	
Violet Bougainvillea Spectabilis	1.88	0.25	0.73	0.35	
Spinach	0.47	0.55	0.51	0.13	(Chang et al., 2010)
Ipomoea	0.47	0.55	0.51	0.13	
Bougainvillea brasiliensis	5.00	0.25	0.36	0.45	(Lai et al., 2008)
Garcinia suubelliptica	6.48	0.32	0.33	0.69	
Ficus reusa	7.85	0.52	0.29	1.18	

Table 2 Photo Electrochemical Parameters of Natural Dye Based DSSC

In year 2007, Wongchareea et al. fabricated DSSCs using natural dye extract from rosella, blue pea and a mixture of the extracts and the efficiency is 0.37%, 0.05% and 0.15% respectively (Wongchareea et al. ,2007). Table 2 shows current status of photo electrochemical parameters of DSSC based on natural dyes till date.

According to Table 2, the best performance has been produced by Red turnip based on the work performed by Calogero (Calogero et al., 2010). This work resulted in a remarkable J_{sc} of 9.5 mA/ cm² and a efficiency of 1.70%. The use of raw betalains, having high concentration of betaxantins, gives a promising result. The presence of carboxylic groups in the betalains presents, together with the higher oxidation potential, an advantage for anchoring. Red turnip contains a high concentration of betalain pigments. Work done by Bazargan (Bazargan, 2009) produced the second best efficiency. DSSC sensitized with pomegranate juice produced an efficiency of 1.50%.Pomegranate juice mainly contains cyanin derivatives and exists as flavylium at natural pH. The third best performance has been delivered by shiso leaf pigments. The shiso plant is well known in Japan as a vegetable. Leaves of this plant contain two anthocyanin pigments referred to as shisonin and malonylshisonin. An efficiency as high as 1.30%, J_{sc} of 4.80 mA /cm² and V_{oc} of 0.53 V is obtained for this dye. Shisonin also produced a promising performance of 1.01% (Kumara et al., 2006). Natural dyes were extracted from natural materials such as flowers, leaves, fruits, traditional Chinese medicines and beverages and used as sensitizers to fabricate DSSCs. The photoelectron chemical performance of the DSSC based on these dyes showed that V_{oc} varied from 0.38 to 0.69 V and Jsc ranged from 0.14 to 2.69 mA /cm². Specifically, a high V_{oc} of 0.69 V was obtained from the dye extracted from mangosteen pericarp sensitizer (Zhao et al., 2011). Sesbania grandiflora and Hibiscus rosasinesis also produced promising efficiency of 1.02%. A variable amount of flavonolglycosides was obtained from the two extracts (Lawrence & Kazmerski, 1997). Various natural pigments containing anthocyanins were extracted from tropical flowers and obtained J_{sc} ranging from 1.1 to 5.4 mA/ cm² and V_{oc} ranging from 0.39 to 0.41 V. The overall efficiency and FF of these cells varied from 0.20% to 1.14% and 0.53 to 0.64 respectively. The extract from Hibiscus surattensis gave the best photosensitized effect of 1.14% (Lawrence & Kazmerski, 1997). Another best results so far are the anthocyanins extracted from Jaboticaba and Calafate yielding $I_{SC} = 9 \text{ mA/cm}^2$, $V_{OC}=0.59 \text{ V}$ and 6 mA/cm^2 , 0.47 V respectively. Other antocyanins extracted from blackberries gave a conversion efficiency of 0.56 %. Another intresting class of natural dyes is tannis because of their photochemical stability. DSSCs using tannins and other polyphenols extracted from Ceylon black tea gave photocurrents of up to 8 mA/cm². Calogero et al. reported that a conversion efficiency of 0.66% was

obtained using red Sicilian orange juice dye as sensitizer (Calogero et al., 2010).

Furthermore, Roy et al. indicated that when using Rose Bengal dye as sensitizer, the J_{sc} and V_{oc} of their DSSC reached 3.22 mA/cm² and 0.89 V respectively resulting in a 2.09% conversion efficiency (roy et al., 2008) & (Wongcharee et al., 2007). Thus so far, several natural dyes have beenutilized as sensitizers in DSSC. It is found that the DSSCs contained different pigments which produced diverse photosensitizing effect, justifying that only selected pigments are capable of converting sunlight into electricity. The best performance is found for betalain pigment due to better interaction between betalain and TiO₂.

5. CONCLUSIONS

Ruthenium dyes are currently considered as the best dyes for the production of efficient DSSC having efficiency of 10-11%. However, the noble metal Ruthenium is a limited resource and is expensive. In order to obtain even cheaper dyes for DSSC, metal-free organic photosensitizes are strongly desired. Hence natural dyes extracted from different easily available flower and fruits are the suitable alternative for possible application as sensitizers to inorganic dyes in DSSC. Although the efficiencies obtained with the natural dyes are below the requirements for large scale production, the results are hopeful and can boost additional studies oriented to the search of new natural sensitizers and to the optimization of solar cell components compatible with such dyes. Hence there is still remains scope for further development for this technology.

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