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Dye sensitized solar cells: An experimental aspect

JK Dongre
Department of Physics, Government Post Graduate College, Khurai (Sagar), Madhya Pradesh, India

Abstract
The huge energy demand is increasing due to global population growth and economic development results in fast depletion of traditional energy sources. Therefore we need an alternative energy source which not only satisfy our current energy needs but also have low environmental effect. Among the renewable energy sources solar energy would be best option because sun is the most abundant energy source. The dye-sensitized solar cell (DSSC) is an alternative to common silicon solar cells due to its advantages of easy fabrication, cost effective and high solar to electric conversion efficiencies. In DSSC technology, does not require clean room facility and highly pure materials. An attempt has been made through this article to report the development of key components transparent conducting substrate, photoanode, dye sensitizers, counter electrode and redox electrolyte in dye-sensitized solar cells. Additionally, DSSC operating principal and characterizations are also described in the review article.

Keywords: dye-sensitized solar cell, photoelectrode, sensitizer, electrolyte, conversion efficiency

1. Introduction

Energy problem will be the top one of the 10 critical problems that humans may face in the coming 50 years, which was pointed out by Professor Smalley of Rice University in the USA [1]. The exponential demand for energy due to global population growth and economic development is increasing day to day result in dwindling of fossil fuel supplies and increasing high oil and gas prices [2]. It has been reported that around 80% of energy is supplied by fossil fuel for worldwide consumption. Therefore unlimited worldwide consumption of fossil fuels and the concomitant emission of greenhouse gases, forces us to application of renewable energy sources. The alternative energy sources should sustainable for instance, solar energy, wind energy, hydropower, and tidal wave energy. Among all other non-traditional energy recourses solar energy would be best choice in near future. In 1999, the worldwide energy consumption was about $4x10^{20}$ J, which the Sun supplies to Earth in an hour [3] and now our current rate of energy requirement is around13 TW [4] very less than the energy (120,000 TW) of the sun which is continuously delivers to the earth. It tells that our current energy needs would be satisfied if we cover only 0.1% of the earth's surface with 10% efficient solar cell [5]. Based on the photovoltaic effect solar-cell is a device that converts solar energy to electricity. Up to now several photovoltaic devices have been developed, but their use at large commercial scale is limited. The contribution of solar energy remains negligible around 0.1% of the global energy demand [6]. To utilize the solar energy at the large scale there are two major obstacles namely conversion efficiency and cost. Therefore, efforts should be made to enhance the efficiency of solar cells with economical viability. The solid-state p-n junction solar cell is called first-generation devices. Based on single-crystal silicon for such devices about 18% efficient solar cell is reported at commercial purpose [7]. However, it could not enter in tough market due to high cost of manufacturing and installation. to harvest incident solar photons with greater efficiency researcher are concentrated on second generation devices consisting of $CuInGaSe_2$ (CIGS) and other compound semiconductors, such as gallium arsenide (GaAs) and cadmium telluride (CdTe). However, these devices have suitable band gap match to the solar spectrum and high optical absorbance, but their efficiencies are not more practical at low cost.

Further to reduce the challenges i.e. efficiency and production costs, third-generation solar cells, such as dye-sensitized solar cells (DSSCs) bulk heterojunction cells and organic cells are promising for large-scale solar energy conversion. DSCs are more advantageous as compared with the conventional silicon-based or compound-semiconductor thin-film solar cells, because the device is being built with high efficiency and lower costs.

2. Historical background

Edmund Becquerel is usually credited as the first to demonstrate the photovoltaic effect in 1839 [8]. He used AgCl coated platinum electrodes and upon illumination in acidic solution a small photocreated voltage/current was obtained due to emission of charge carriers from metals. The emission of the charge carries were systematically investigated by H. Hertz and W. Hallwachs in 1886 [9]. A few years later J. Moser reported enhanced photo electrochemical response in a dye-sensitized photo electrode on the base of Becquerel's electrodes, Moser imbrued his halogenated silver plates in an erythrosine solution [10]. Later, metal electrodes were replaced by semiconductors to reduce recombination effects. For example sensitization effects at the interface between ZnO and a dye was investigated by Tributsch *et al.* [11]. After that, A. Fujishima and K. Honda proposed an electrochemical photocell to decompose water into oxygen and hydrogen based on semiconductor TiO_2 electrode in contact with an aqueous electrolyte [12]. Inspired by water splitting, H. Gerischer discussed the application of this type of cell for the conversion of light into electricity in 1975 [13]. After this investigation, there was a concrete start to work in the field of light energy conversion in dye-sensitized photo electrochemical cells. Untill 1980, efficiency of dye-sensitized photo electrochemical cell remained low since measured photocurrents were quite low. The interest in photovoltaic energy conversion through dye-sensitized solar cell finally, exploded by O'Regan and Gratzel in 1991. They reported a device made of sensitized mesoporous TiO_2 with a conversion efficiency of 7:9 % [14]. This triggered a boom in research activities and now the conversion efficiency of 14% has been achieved for rigid glass based DSSCs [15].

2.1 Operational Principle

Recently, DSSCs emerge as the most promising energy technology for light-to-electrical energy conversion at low cost. A typical DSSC is mainly consisting of four components: i.e. a photoanode having large band gap mesoporus metal-oxide semiconductors that could not absorb large fraction of solar radiation itself, dye molecules attached on the semiconductor surface known as a sensitizer, an electrolyte containing most commonly iodide/triodide (I^-/I_3^-) and a conducting substrate coated with a catalyst (platinun or carbon) is used as cathode. The schematic representation of a dye-sensitized solar cell and operation are shown in Fig. 1. A charge separation process in a DSSC consists of the following steps [1].

1. When the device DSSC exposed to sunlight, the dye sensitizer gets excited i.e. the electrons in the dye become exited from ground state to the excited state.
2. The exited dye (S^*) then injects an excited electron into the conduction band of the semiconductor (on a femto- to picosecond timescale) leaving an oxidized dye (S^+). For DSSC device performance, the time scales of the injection process should be compared with decay of the excited state of the dye to the ground state.

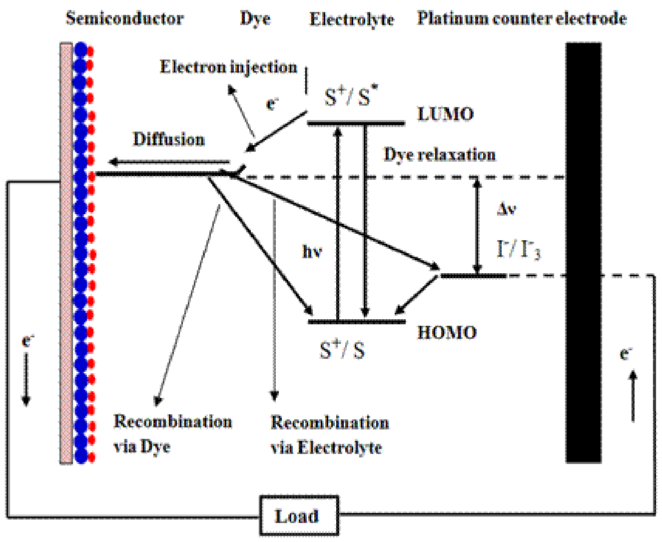

Fig 1: Schematic illustration of operation principle of dye sensitized solar cell.

3. These inject electrons diffuse and flow across the semiconductor network until arriving at the back contact electrode.
4. Due to ejection of an electron dye gets neutral (S^+). The neutrality of the oxidized dye $(S^{+)}$ is regenerated by iodide ion in the electrolyte within a few microseconds that donates electrons to the dye restoring it to the initial state which oxidizing the iodide to iodine. This iodine becomes tri-iodide ion under excess iodide conditions.
5. The tri-iodide ion diffuses to the counter electrode, receives an electron and becomes an iodide ion again, and the system returns to the original state.

Within completed circuit, the DSSC generates electric power from light and does not cause permanent chemical changes or transformation. This complete cycle of charge transfer is known as forward charge transfer process. The performance of cell depends on the forward charge transfer process and if recombination losses called backward electron transfer processes as depicted in the Fig.1 are prevented. To prevent the backward electron transfer it should necessary that dye injects electrons into the semiconductor with a quantum yield of unity [16]. Photocurrent density is dependent on the energy separation between the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) which ensures absorption of low energy photons in the solar spectrum. Therefore for effective electron injection the level of LUMO should be more negative than the bottom of conduction band of the semiconductor. Apart from this, the energy difference between the HOMO and redox chemical potential must be more positive for effectively accept the donated electrons from the redox mediator [17].

3. Synthesis and Materials for DSSCs

Synthesis of DSSCs is relatively simple as compared to that of conventional Silicon based solar cell.

3.1 Transparent conducting substrate

Primarily it requires to two sheets of conductive transparent materials, one of them uses for the deposition of semiconductor film and other one for catalyst, as current collectors. The transparency (>80%) of conducting glass must be high enough, which passage the maximum sunlight into cell region. Normally, two types of conducting glass substrate fluorine tin oxide (FTO) and indium tin oxide (ITO) have been used as substrates. To minimize energy losses and for efficient charge transfer the electrical resistivity of the substrates should low. The ITO films coated glass has a sheet resistance value 18 Ω/cm^2 whereas FTO films show sheet resistance value $8.5\Omega/cm^2$. In an experiment it was reported that the sheet resistance value of ITO increased from the 18 Ω/cm^2 to 52 Ω/cm^2 upon sintering, whereas sheet resistance value of FTO remained constant at 8.5 Ω/cm^2 [18]. It was also reported that an efficiency of DSSC based on FTO has larger value 9.4% as compared to that of ITO based cell at identical condition. However, ITO and FTO both transparent conducting oxide substrates have been used for the fabrication of DSSC, but their use in the commercialization purpose is restricted due to their high manufacturing cost, sensitivity to thermal treatment and limited transparency in the near-infrared region [19].

Now a day's plastic substrates have also been used for the fabrication of DSSCs as an alternative to ITO and FTO [20]. A maximum power conversion efficiency of 4.2% was obtained for the plastic based flexible DSSC prepared using the ball milling technique [21]. But polymer substrate is not suitable for preparing electrodes by thermal treatment at high temperature above 200 ^{0}C [22]. The ultrathin film of graphene can also be used as an alternative to ITO and FTO because of their high conductivity and transparency in near infrared region [23]. It has several advantages over FTO and ITO such as chemical and thermal stability, ultrasmooth surface and low cost make it promising candidate for DSSC in the future [24].

3.2 Semiconductor oxide film: photoanode

The performances of the photoanodes depend on the band gap, morphology, composition of metal oxides and thickness of metal oxide layers [25].

To prepare photoanode, a semiconductor nanoparticle is deposited onto a conducting substrate by means of a deposition technique and the as grown substrate is calcined at temperature above 300 °C. The huge surface area of the semiconductor film, support the monolayer of a sensitizer and the medium of electron transfer to the conducting substrate. Metal oxides like titanium oxide (TiO2) [26] zinc oxide (ZnO) [27], and stannic oxide (SnO2) [28] have been used as the semiconductor material.

Among the entire mesoporous oxide semiconductor, TiO_2 has been considered best semiconductor material for DSSCs since it gives the highest efficiencies. Apart from this, TiO_2 is a nontoxic material, stable, and has a high refractive index (n = 2.4-2.5). TiO_2 can be synthesized in its three natural forms namely rutile, anatase and brookite. Among them, however rutile is the most stable form of TiO_2, but anatase crystalline form exhibited better performance due to its chemical activeness when used in DSSC. In a comparative study, anatase-based cell exhibited 30% larger short-circuit photocurrent than that of a rutile-based solar cell having same film thickness [29]. Relatively smaller surface area and slow electron transfer rate in the rutile film as compared to that of anatase film would be cause of lower photocurrent. The smaller surface area adsorbed lower amount of dye on the semiconductor surface leads less efficient solar cell [30].

Interfacial charge recombination as shown in Fig.1 causes a loss of photogenerated electrons results in lowering the energy conversion efficiency of DSCC [31]. Several attempts have been made to overcome the problems of recombination. The film morphology and network plays important role on the electron transport dynamics of DSSCs. The morphology of the films greatly increase the electron life time and reduce recombination. Liao *et al.* [32] reported that DSSC based on the hierarchical anatase TiO2 sphere photoelectrode shows higher power conversion efficiency (9.35%) as compared to that of nanoparticles (7.37%), nanofibers (8.15%) and ellipsoid TiO2 spheres (7.93%). The larger dye loading, superior light scattering ability, faster electron transport and longer electron lifetime, superior characteristics of the hierarchical sphere-based DSSC as compared to other nanostructures causes the significant improvement in the cell parameters.

The dark current which decreases the efficiency of DSSCs, can also be minimized by use of blocking layers of wide band-gap inorganic oxides such as ZrO_2 [33], Al_2O_3 [34], SiO_2 [35], and Nb_2O_5 [36] onto TiO_2 surface. This blocking layers act as barrier layers for interfacial electron transfer between injected electrons and redox couple since it increases physical separation between them [37].

3.3 Dye (Sensitizer)

In DSSC assembly dye is a backbone of the device which plays an important role to harvesting the incident light for the photon-to-electron conversion. The FTO/ITO substrate coated with porous semiconductor films are incubated in a solution of sensitizing dye for several hours to allow dye adsorption at the semiconductor surface. The dye loading process directly affects the light harvesting ability of the device and thus performance of DSSC. Various dye loading parameters such as dye adsorption time, dye concentration and the incubation temperature affects the electrical response of DSSCs [38, 39]. The quantity of dye molecules adsorbed on the semiconductor surface monitoring by the impregnation time also responsible for device performance. Various kinds of method such as fluxing, pumping of dye solution have been adopted to sensitize a semiconductor photoanode. These methods are not time limiting process. The impregnation time of dying process should minimize in view of the commercial manufacture. Recently a novel ultrafast dye loading process has been reported. This method is based on a microfluidic approach which allows monitoring the dye adsorption mechanisms and to optimize the impregnation times required for the DSSC fabrication [40].

Dye acts like an electron pump and inject an electron to the conduction band of semiconductor. As the hart in DSSC, many requirements should be met [41, 42]:

1. The sensitizer should be able to absorb wide range of the solar spectrum that cover the whole visible region and the near-infrared (NIR) region.
2. It should carry anchoring groups (-COOH, -H2PO3, -SO3H, etc.) to strongly bind the dye onto the semiconductor surface.
3. The extinction coefficient of dye must be as high as possible to absorb sufficient light with minimum quantity of dye material namely panchromatic absorption.
4. The dye posses appropriate steric properties to suppress charge recombination at the semiconductor /electrolyte interface and dye aggregation
5. LUMO of the dye must be more negative than the conduction band of semiconductor for efficient electron injection.
6. HOMO of the dye must be more positive than the redox potential of electrolyte for the efficient regeneration of the oxidized dye.
7. The photo, electrochemical and heat stability of the dye are required for longer lifetime of DSCs.
8. The dye should be cheap, non-toxic, and exhibiting flexible properties like solubility.

Based on these requirements, several kinds of sensitizer have been used for better photovoltaic performance and long-term stability. For better conversion yield in DSSC device three types of sensitizers including metal complex sensitizers, metal-free organic sensitizers, and natural sensitizers have been designed and applied in the past decades.

For a good sensitizer should have anchoring ligands (ACLs) and ancillary ligands (ALLs). Anchoring groups (such as carboxylic acids, acety lacetonate, catechol, silanol groups) guarantee the strong adhesion of photosensitizers on the surface of semiconductor and ALLs groups such as bipyridines or terpyridines have been employed to improve the photovoltaic performance.

Metal complex sensitizers possess both ACLs and ALLs. Generally, a sensitizer has structure $ML_2(X)_2$ where M is used for Ru, L is 2, 2'-bipyridyl-4, 4'-dicarboxylic acid and X represents halide, cyanide, thiocyante, acetyl acetonate etc. Among the metal complexes, Ru complexes have been exhibited the best photovoltaic performance. The ruthenium complex cis-RuL_2 $(NCS)_2$ known as 719 N dye is most studied sensitizer and considered as reference dyes for DSSC [43]. By changing ancillary ligands in N719 dyes other Ru complexes photosensitizers have been optimized to further improve the efficiency of DSCs. Nazeeruddin *et al.* made a dye namely N749 dye (black dye) in which bipyridyl ligands was changed by terpyridyl ligands that enhance the performance DSSC dramatically by the broadening of the absorption area [44]. Using N719 as reference dyes Wang *et al.* have been prepared A Z series dye by introducing hydrophobic alkyl chain to bipyridine [45]. However, the efficiency and stability of DSSC of Ru based dyes are good but the high cost and lace of ruthenium oriented to find the other options.

Metal-free organic sensitizers are an attractive alternative to ruthenium based sensitizers. Organic sensitizers are free from toxicity and less expensive material. Apart from this, they posses higher extinction coefficients as compared to Ru(II) polypyridyls. By using organic dye such as coumarine and indoline dyes the efficiency around 8-9% has been achieved [46, 47]. In an experiment it is reported that dye C219 showed efficiency greater than 10% [48].

3.4 Counter electrode

Counter electrode is also an essential component for regeneration of the electrolyte. A layer of catalytic prepared onto a conducting glass substrate. Platinum (Pt) is the most popular used as the catalyst on the counter electrode substrate because of its high exchange current density, good catalytic activity, and transparency. For counter electrode Pt has been deposited onto conducting substrates by different techniques such as electrode position, spray pyrolysis, sputtering, and vapor deposition, thermal decomposition of the H_2PtCl_6 solution and screen printing and annealing of different Pt-based pastes. But among the methods thermal decomposition is good for best performance and long-term stability of DSSC. Due to high cost of Pt various promising alternatives have been widely investigated to replace it. The most commonly used alternatives are carbonaceous materials like graphite, grapheme, carbon black, carbon nanotubes. Apart from this, other catalysts like conducting polymers [such as poly (3,4-ethylenedioxythiophene)] and recently cobalt sulfide [49] also considered for counter electrodes.

3.5 Electrolyte

The electrolytes are one of the key components, which greatly affect the stability as well as conversion efficiencies of the DSSCs [50]. It provides a medium to charge transport. It also uses for dye regeneration. The electrolyte must have high electrical conductivity and low viscosity. It should make good interfacial contact with both the electrodes (i.e. working and the counter electrode). It should not harmful and evaporate at large temperature. The chemical, thermal, optical, electrochemical and interfacial stability of electrolyte must be long enough, so that desorption and degradation of the dye from the oxide surface would be minimized. The electrolyte should not exhibit a significant absorption in the range of visible light. According to its physical states, an electrolyte can be categorized into three types: liquid electrolyte, quasi-solid-state electrolyte, and solid-state electrolyte.

Two types of liquid electrolytes i.e. organic solvent-based electrolytes and ionic liquid-based electrolytes have been widely used and investigated in DSSCs. Generally, the liquid electrolyte consists of a solvent, a redox couple and additives. The most frequently used organic solvent electrolytes are acetonitrile, valeronitrile, 3-methoxypropionitrile, ethylene carbonate, propylene carbonate, N-methylpyrrolidone and g – butyrolactone etc. [51]. The iodide/triiodide (I^-/I_3^-) is studied the most common redox couple for DSSC application [52]. It has proven to be one of the most versatile because of its excellent electrochemical properties, such as fast oxidation of I^- and slow reduction of I_3^-, relative high stability, low cost and easy preparation [53].

However, recently I^-/I_3^- based electrolyte show power conversion efficiency more than 12% [54], but few drawbacks often limit its adaptability for DSSC application. The I^-/I_3^- based electrolytes are very corrosive and its relatively high vapor pressure create difficulties in the encapsulation of the cells. Besides, a part of incident visible light at 430 nm is absorbed by ion and thus may diminish the efficiency of the cell. As a result, to replace the I^-/I_3^- system several alternative electrolytes have been quested such as $LiBr/Br_2$, Br^-/Br_3^-, $SCN^-/(SCN)_3^-$, $SeCN^-/(SeCN)_3^-$, Co(II)/(III), Cu(I)/(II), Ni(III)/Ni(IV). It is also envisaged from the literatures that by introducing additives to the electrolyte such as 4-tert-butylpyridine (4TBP), guanidiumthiocyanate, and methylbenzimida- zole (MBI) the efficiency and stability can be can improved. To solve the stability problem a variety of quasi-solid and solid electrolytes have been investigated for DSSC applications. Quasi-solid electrolyte is made by adding inorganic nano materials, organic molecular gel, or polymer into liquid electrolyte by which liquid electrolyte is solidated in 3D space network [55].

4. Characterization of solar cell performance

Solar cells are characterized by various parameters such open-circuit voltage (Voc), short-circuit current (Isc), fill-factor (ff), efficiency (η) etc.

Among the parameters used to characterize solar cell performance power conversion efficiency (η) is the most important. Figure (2) represents a typical I-V curve which is also known as photovoltaic power output characteristics. The overall power conversion efficiency can be estimated as [56];

$$\eta(\%) = \frac{P_{(out)\max}}{P_{in}A} \times 100 = \frac{V_{\max} \times I_{\max}}{P_{in}A} \times 100 \qquad (1)$$

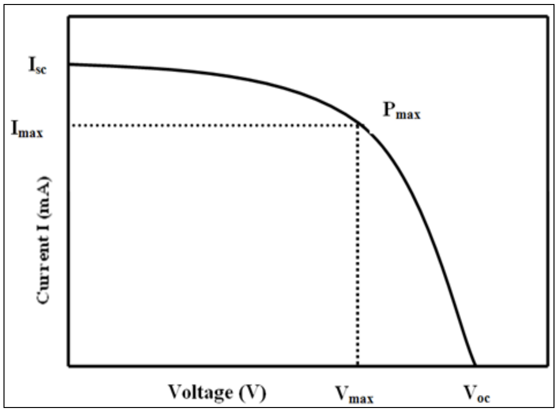

Fig 2: A typical J–V curve of a DSC.

Where V_{max} and I_{max} are the current and voltage at the point of maximum power respectively of solar cell, A is the illumination area of the photoelectrode and P_{in} is the incident solar flux. A parameter known as fill factor (ff), measures the 'squareness' of the photovoltaic output curve and is given as;

$$ff(\%) = \frac{V_{max} \times I_{max}}{V_{oc} \times I_{sc}} \times 100 \tag{2}$$

Where I_{sc} and V_{oc} are short circuit photocurrent and open circuit voltage, respectively. The I_{SC} is the photocurrent per unit area (mA cm^{-2}) when the applied bias potential is zero. When no current is flowing through the cell, the potential equals the V_{OC}.

To better evaluate the I_{SC}, another important parameter, incident photon to current conversion efficiency IPCE, has to be taken into account. The IPCE, sometimes referred to also as the "external quantum efficiency" (EQE), is an important characteristic of a device. The IPCE is a measure of the efficiency of the solar cell to convert the incoming photons to photocurrent at different wavelengths. This is done by measuring the resulting photocurrent of the solar cell when illuminated by monochromatic light. A typical IPCE spectrum is shown in Fig. 3. It can be calculated for each wavelength (λ) by the following equation:

$$IPCE = \frac{1240 \times I_{sc}(mAcm^{-2})}{\lambda(nm) \times P_{in}(mWcm^{-2})} \tag{3}$$

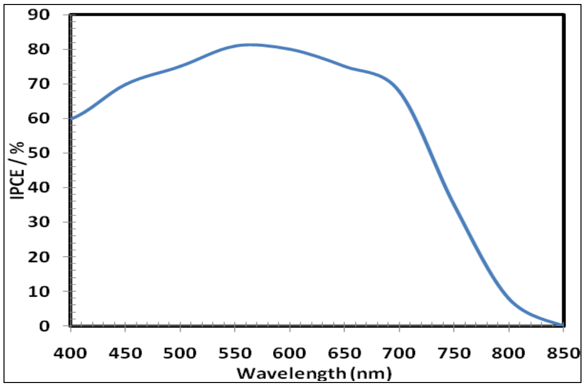

Fig 3: A typical IPCE curve of a DSC

Generally, a low value of IPCE results in low I_{SC}. Panchromatic dyes can absorb incident light at a longer wavelength which improve the short-circuit current I_{sc}, and hence the IPCE value [57].

5. Concluding remark

Among the renewable sources of energy solar energy is the best choice. As a novel photovoltaic technology, dye-sensitized solar cells have potential to compete with conventional solid state junction devices for the conversion of solar energy to electricity. For the commercialization of DSSC, the manufacturing cost should be further minimized by applying low-cost materials required for fabrication. Besides, efficiency and stability are the two major factors that should be optimized from the use of novel materials necessary for DSSC. To gain high cell efficiency, both J_{sc} and V_{oc} must be further improved. The J_{sc} can be improved by proper selection or development of new sensitizers that can absorb in the near-IR region with relatively large absorption coefficients. The V_{oc} of DSSC can be further improved by molecular design of the sensitizers which suppress charge recombination. After the test of stability, DSSC will be entered in the tough PV market.

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