

Fundamentals and Recent Advancements in DSSC

Charushila C. Revadekar, Swati R. Gawali *

Department of Physics, CES's Dr. Arvind B. Telang Sr. College, Nigdi, Pune.

Affiliated to Savitribai Phule Pune University, Pune.

*Corresponding Author: swati.r.gawali@gmail.com

Abstract: With the industrial revolution; energy demands have been drastically increasing and to meet these energy demands, lot of efforts were taken to find alternate energy sources. The global challenges for clean environment deviated the scientific community towards the first and second generation of photovoltaic solar cells. But the first generation was restricted by cost effectivity and temperature sensitivity whereas the second generation had limitation of low conversion efficiency. Among third generation photovoltaic cells; dye sensitized solar cell (DSSC) was proved to be a most attractive option due to its advantages such as simple preparation method, ease of production, low cost, low toxicity, flexibility and good efficiency under reduced light intensity. O'Regan and Grätzel invented DSSC in 1991 since then a lot of work have been in continuous progress to tailor its chemical and physical properties to increase efficiency and commercialization. This review helps the readers to understand the fundamentals of DSSC, its working and various basic components like photoanode electrode, counter electrode, electrolyte, dye sensitizers and fabrication techniques. All these key components are discussed in detail by considering their essential selection criteria and recent advancements in them. In addition, practical applications are also mentioned and are presented in this paper.

Keywords: Dye-sensitized solar cells, Photoanode electrode, counter electrode, electrolyte, dye sensitizers, fabrication techniques.

1. Introduction:

About 8000 B.C. at the dawn of agriculture, the world population was approximately 5 million which has raised tremendously and world population is more than 8.3 billion. This has increased the demand of basic necessities like food, water, shelter and energy. To meet the energy demands initially fossil fuels and then nuclear energy, bioenergy was used as a primary source. The constant use of these sources has resulted in the emission of hazardous byproducts such as greenhouse gases (GHGs) (methane, nitrogen oxides and oxides of carbon), radioactive wastes (like Technetium-99 isotopes) [1]. This leads to serious deterioration of the natural biosphere thereby changing climate patterns in adverse way [2,3] and impacting the human life. To overcome this condition scientist community moved towards the environmentally friendly, ever lasting renewable energy sources.

Being sustainable and environmentally benign; solar energy is the best alternative source. Nearly 173,000 terawatts of solar energy are continuously striking the Earth's atmosphere. In a single day a huge amount 1.20×10^7 W is received by the Earth which is sufficiently enough to provide power to Earth for two decades [4]. Hence, the Sun will be the most convenient energy source; provided appropriate technology and infrastructure will be developed. So far, there is an appreciable development in the solar energy harvesting. The different generations of photovoltaic cells show the evolution in solar cell technology and the advancement in materials. The First Generation of Photovoltaic Cells is based on silicon-based PV cells. The materials used were monocrystalline silicon, polycrystalline silicon and silicon cells with materials GaAs. The efficiency claimed was in the range of 10 – 30 % but they are

restricted by cost effectivity and temperature sensitivity issues [5-7]. Researchers tried to overcome these challenges and in the Second Generation of Photovoltaic Cells; thin film photovoltaic cells based on materials like CdTe, gallium selenide and copper (CIGS) or amorphous silicon technology were introduced. This generation provided the cost reduction and better improved mechanical properties and additional applications in new areas like electrochemistry [8]. But it has disadvantage of low conversion efficiency as compared to the first-generation photovoltaic cells and the toxicity and scarcity of materials used. The fundamental challenges of the first two generations of solar cells inspired to the development of the current third generation solar cells. The widely studied third generation solar cells are dye sensitized solar cells (DSSCs), perovskites, organic solar cells and quantum-dot solar cells. The discovery of generating electricity at oxide electrodes in electrochemical cell by illuminating organic dye fascinated O'Regan and Grätzel and a new era of third generation solar cell i. e. DSSC was evolved. As compared to first- and second-generation solar cells, DSSCs have an upper hand in multiple points such as simple design, simple manufacturing mechanism, minimum manufacturing expenses and the choice of inexpensive; widely obtainable substances (e.g., TiO₂ and carbon-based components). Additionally, they operate effectively in low and indoor light environments. Boschloo and group had claimed a remarkably high efficiency of 32% under 1000 lux illumination strength for a DSSC with a copper-based electrolyte [9]. This latest advance has widened the manufacturing of DSSC for commercial energy production and also for compact devices.

The present study highlights significant advancements in DSSC and the current situation by considering system functionality and flexibility with electrolytes used. Additionally, recent developments in electrolyte substances, photoanode materials, dye sensitizers and counter electrode materials are also reviewed. Furthermore, the key role of these key parameters in DSSC manufacturing and development is also discussed.

2. DSSC:

A dye-sensitized solar cell is a type of third-generation thin film solar cell that converts sunlight into electricity using light-absorbing dyes in place of traditional silicon. DSSC works on the principle: The dye catches the intensity of photons emitted by illumination and convert it into electrical power via a series of electron exchanges on its surface.

Construction of DSSC:

The four main components of a DSSC includes a working electrode, sensitizer (dye), redox-mediator (electrolyte) and a counter electrode. The most common n-type DSSC consists of a thin layer of porous titanium dioxide nanoparticles which is covered with a molecular dye. The dye absorbs sunlight similarly like the chlorophyll in green leaves. The titanium dioxide is immersed in an electrolyte solution, above which is a platinum-based catalyst. Thus like a conventional alkaline battery, an anode; the titanium dioxide acts as an anode and the platinum as a cathode. They are placed on either side of the electrolyte acting as a liquid conductor.

The working principle for n-type DSSC:

The working principle for n-type DSSC can be summarized into four basic steps:

Step 1: Photoexcitation of the electron in the dye

After passing through the transparent electrode the sunlight enters the dye layer. The dye molecules on the metal oxide conductor absorb wide spectrum of sunlight. TiO_2 is a wide band gap semiconductor which absorbs the wavelengths only below 400 nm and is transparent to visible and near infra-red (NIR) light. This allows the dye molecules to harvest a broadband region of the solar spectrum. The absorbed photon excites electrons of the dye molecules in the excited state.

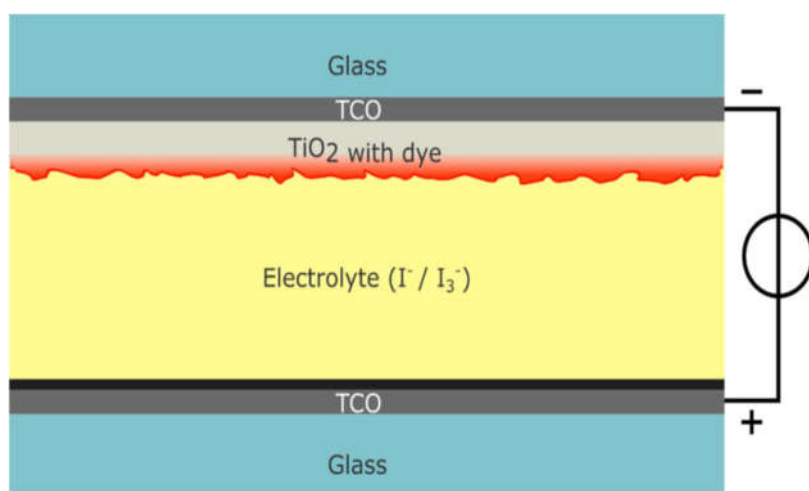


Figure 1: Construction of n-type DSSC [10]

Step 2: Electron injection

As the excited state of dye molecules have nearly same energy as that of the conduction band of titanium dioxide, therefore these electrons are injected into the conduction band of the n-type semiconductor (titanium dioxide). These injected electrons then transferred to the TCO surface and in this process electron hole pairs are generated.

Step 3: Dye regeneration

After losing an electron the dye gets oxidized and readily accepts electrons from the electrolyte i.e. the redox pair $\frac{\text{I}^-}{\text{I}_3^-}$ thereby making the ground state of dye molecules available. Thus, the dye molecules are ready again to absorb incoming photons and generate excited electrons continuously.

Step 4: Regeneration of the electrolyte

The electrons from titanium dioxide then flow toward the transparent electrode and collected for powering a load. After flowing through the external circuit, the electrons are re-introduced into the cell on a metal electrode on the back (known as the counter electrode) and then into the electrolyte; thereby reducing the tri-iodide I_3^- ion in the electrolyte. Thus, the electrolyte

recovers its ground state by accepting electrons back and regenerates the oxidized dye. Thus the cycle of regeneration is completed.

The reactions taking place are as below:

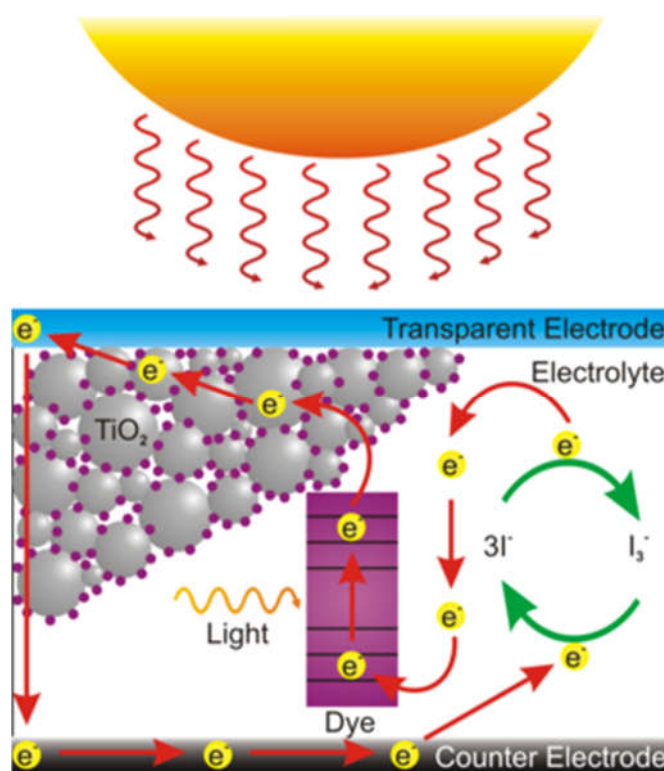
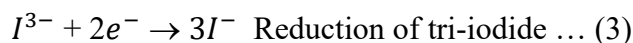
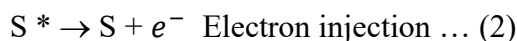


Figure 2: Working Mechanism of DSSC [10]

Continuous engineering experiments are in progress to improve the efficiency of DSSC. The performance of a DSSC is governed by materials used in the photoanode, dye sensitizer, electrolyte, and counter electrode [11]. Apart from the fabrication techniques, specific criteria must be satisfied by these materials to ensure various steps such as effective light harvesting, fast electron transport, minimum recombination of electron hole pairs, long-term stability and so on; to enhance the output efficiency of DSSC [12].

3. Fabrication Techniques:

Fabrication of a highly efficient DSSC is a careful integration of layer-by-layer processing in material science. The routine straight way of fabrication includes following steps:

The canonical device stack — TCO glass (or flexible TCO film) → Compact TiO₂ blocking layer → mesoporous TiO₂ scattering/photoanode → Dye monolayer → Electrolyte (or solid HTM) → Counter electrode. But in practice each step is highly demanding as it strongly affects the steps of charge injection, transport, electron hole recombination and long-term stability [13]. The fabrication of DSSC begins with the substrate.

a) **Substrate:** Glass is coated with fluorine-doped tin oxide (FTO) as it provides decent conductivity, optical transparency and thermal stability during high-temperature processing [14]. For temperatures below 150 °C, ITO/PET or ITO/PEN and emerging flexible TCOs are used. These alternative sintering and chemical strategies make the DSSC more flexible or wearable [13].

b) **Preparation and deposition of TiO₂ paste:** The heart of the DSSC is the mesoporous TiO₂ electrode. The main function of this electrode is to facilitate maximum dye loading by providing high internal surface area and thus forms a good percolating network for electron diffusion. On the basis of desired morphology and film thickness; different lab and pilot-scale fabrication techniques are used [15]. Few of them are mentioned below.

i) Tape casting/ Doctor blade method: Being simple and cost effective it is widely used in laboratories. The thickness of thin film is controlled by tape gap and blade [13].

ii) Screen Printing method: It is mainly used in industries for large scale and large area thin film fabrication. It is common in module prototype [16].

iii) Spin Coating: This technique is ideal in research for preparing uniform thin films but is not scalable [15].

iv) Slot-die and blade coating: The method is in demand for scalable manufacturing and continuous processing [17].

v) Spray pyrolysis / Spray coating and ink-jet printing: The technique allows patterned deposition and is more compatible with roll-to-roll pattern [18].

The method of paste formation decides various properties such as particle size, thin film uniformity, porosity and so on. Hence optimization of paste chemistry is very important in every method [15,18].

c) **Sintering:** In conventional DSSC sintering is a critical process as it ensures the proper bonding, stability of the electrode layer and prevent leakage from the central portion of the cell. Generally sintering temperature is high up to 400 °C which limits substrates and raises cost [17]. Consequently, low temperature sintering methods such as laser sintering, chemical sintering, photonic (flash) sintering, plasma treatments and so on are developed. It lowers the sintering temperature to 100 – 200 °C. Due to this DSSC become flexible and can be fabricated at large area [15,17-18].

d) **Dye sensitization:** In deciding the efficiency and performance of a DSSC, dye sensitization of mesoporous thin film is very important. The photoanode captures sunlight and facilitate the process of charge transport with the help of dye molecules adsorbed on the photoanode surface. Generally used dye solutions are N719, natural dye or organic dye. Interfacial treatments are not only useful in for the purposes of enhancing dye's stability, preventing aggregation but also lead to improved efficiency and longevity of solar cell. These treatments can include use of different solvents, additives and coatings to optimize dye sensitive interface and improve the overall performance of DSSC.

4. Material Specifications:

The choice of materials used for photoanode, counter electrode, electrolyte and dye play a crucial role in deciding the performance of a DSSC [19]. Each material must fulfill specific

conditions to ensure overall improvement in the performance of a DSSC. In this section the recent advancements and required criteria of these materials is discussed in detail.

4.1 Photoanode Material:

A working electrode or photoanode is deposition of a thin layer of semiconducting oxide material on a transparent conducting glass plate made of FTO or ITO. Semiconductor oxide materials such as titanium dioxide (TiO_2), niobium pentoxide (Nb_2O_5), zinc oxide (ZnO), n-type tin oxide (SnO_2) and p-type nickel oxide (NiO) were reported for enhanced electron mobility or modified light scattering properties [20]. Recent advancement in these materials is use of nano-structures such as nanoparticles, nanorods, CNTs and hierarchical architectures, which result into improved surface area and dye loading producing increased photocurrent [21].

Essential selection criteria for photoanode material are as below [22,23]:

- The material must possess a wide bandgap (3 to 3.2 eV)
- The conduction band edge must be lower than the LUMO of the dye
- The material must have high surface area and porosity.
- The material must provide fast electron mobility and transport.
- The material must be photo corrosion resistant and chemically stable under UV and electrolyte exposure.

4.2 Electrolyte Material

The electrolyte plays a very critical role in DSSC by mediating charge transport and regenerating oxidized dye molecules thereby directly determining efficiency, voltage and long-term stability. They act as an ionic conducting bridge between photoanode and counter electrode. Most commonly used electrolyte materials are I^-/I^{3-} , Br^-/Br^{2-} , SCN^-/SCN_2 and Co(II)/Co(III) [24-26].

Essential selection criteria for electrolyte material are as below:

- The electrolyte should provide a high ionic conductivity between photoanode and counter electrode.
- Redox couple must be able to regenerate the oxidized dye efficiently.
- It must have low viscosity, long term chemical and thermal stability.
- Optical properties of electrolyte must be different than the dye used so that their absorption spectra will not overlap.

Though I^-/I^{3-} is highly efficient redox pair; due to limitations like high volatility, corrosive to electrode material, photo degradation and poor long-term stability new electrolytes such as cobalt complexes with polypyridyl ligands, organic redox pairs, Fc^+/Fc , solid state electrolytes and so on are used [27,28]. Apart from this long-term light soaking tests on sealed cells are also being conducted to minimize failure of the redox electrolyte [29].

4.3 Counter Electrode Material

The counter electrode acts as appositve electrode in DSSC. It plays a crucial role in collecting electrons from the external circuit and catalysing the reduction of the redox electrolyte. Being catalytically high active and excellent conductive; Platinum has set up a benchmark. But it has limitations of extremely high cost and scarcity advancements are done in the counter electrode materials. Lee et.al [30] reported low cost; flexible graphene film coated with a conducting polymer as a counter electrode; Banerjee et al. [31] reported nanoneedle-array of nickel cobalt sulphide as an alternative for Pt electrode. A highly conducting 1-D aligned poly ethylene dioxy thiophene (PEDOT) coated on the inner and outer surfaces of a hollow carbon nanofiber is used by Anothumakkool et al. as a counter electrode in a DSSC [32].

Essential selection criteria for counter electrode material are as below:

- High catalytic activity to minimize energy loss.
- High electrical conductivity.
- Low charge transfer resistance.
- Good chemical stability or low corrosion in electrolyte environment.
- Cost effectiveness.

4.4 Dye Sensitizers:

The element responsible for the maximum absorption of the incident light in DSSC is the dye. It absorbs photons and injects excited electrons into the semiconductor conduction band.

Essential properties of a dye material are as below:

- A dye must be luminescent.
- A dye must have strong absorption in the visible and near-infrared region.
- The dye's Lowest Unoccupied Molecular Orbital (LUMO) must be above the conduction band of TiO_2 for easy electron injection.
- The Highest Occupied Molecular Orbital (HOMO) must be below the electrolyte redox potential for proper regeneration.
- The dye must have functional groups such as carboxylic acid, phosphonic acid for efficient charge transfer.
- The dye must be stable under prolonged light exposure.
- The dye must provide aggregation prevention. For this, co-absorbents like chenodeoxycholic acid (CDCA), anchoring groups like alkoxy-silyl [21], phosphoric acid [33] and carboxylic acid group [34,35] were inserted between the dye.

Comprehensive literature reports have consistently emphasized the need to optimize the dyes used. On the basis of key parameters such as optimization of optical response and absorption, electron injection efficiency, stability, durability, cost and so on there are different types of dyes used in DSSC. There are plenty of sensitizers under potential study. Common are organometallic dyes, metal free dyes, natural dyes, quantum dot sensitizers, mordant dyes etc.

I] Organometallic Dye:

The transition elements like Ru, Os and Ir and organic ligands make up organometallic dye. The commonly used organic ligands are bipyridine derivatives, thiocyanate, hydrophobic

ligands, cyclometalated ligands and so on. Ru (II) possesses an upper hand in tunable electrochemical, photochemical properties, oxidation level, sustainability and cost effectivity thereby it is most commonly used transition element in DSSC. The widely exploited Ru dyes include (N3-cis-di(thiocyanato) bis (2,2-bipyridine-4,4-dicarboxylate) ruthenium (N3 dye), (di-tetrabutyl ammonium cis-bis (isothiocyanato) bis (2,2'-bipyridyl-4,4'-dicarboxylato) ruthenium (II) (N719 dye), and tris (N,N, N-tributyl-1-butanaminium) [2,2''6,2-terpyredine]-4,4,4''-tricarboxylato(3-)-N1,N1,N1] tris (thiocyanato-N) hydrogen ruthenate (N749 or black dye). Various studies had claimed modified dyes due to limitations of transition elements [36]. One of the studies conducted by Hore et al. [37] had used modified N3 dye by integrating it with traditional aniline-based dyes A1–4 and obtained a conversion efficiency of 7%. In another study, [38] an advanced heteroleptic dual-anchored Ru (II) complex (RNPD) attached to 4-nitro-phenylenediamine (NPD-PC) Schiff compound as a ligand was used as a dye. However, despite the keen interest of scientific community in Ru complexes, efforts have been advanced in finding potential candidates which can be used as sensitizers and replace Ru effectively. Metal free dyes are identified as feasible substitutes.

II] Metal-Free Dye:

Metal-free organic dyes are developing fast as they have higher stability and can design efficient DSSC at lower price though they offer less efficiency in comparison to Ru dyes. Structure variations, simple preparation process, higher absorption coefficient and environmental friendliness are other advantages associated with organic dyes. Hence there is a huge demand to develop new pure organic dyes for the commercialization of DSSCs. Electron donor, electron acceptor or anchoring component are modified to enhance the efficiency of DSSC. Tontapha et al. and group [39] have presented an outstanding overview of TTF- and DTF-based donor dyes and showed sulphur containing chemical tetrathiafulvalene (TTF) as a powerful electron donor. Xia et al. [40] used N, N'-para-amino benzoic acid (PABA) built on N, N-dimethyl-4-vinyl aniline with 4-amino benzoic acid as an acceptor photo sensitizer for a TiO₂ photo electrode. SPSGOD3 and SPSGOD4 are two D- π -A natural sensitizers created by researchers [41] in which their anchoring sites; cyan acrylic acid and rhodanine-3-acetic acid contrast each other on a conventional doner. However, metal free dyes also have certain limitations such as poor stability at high elevated temperatures. Hence advancements in predominant light-harvesting abilities whole visible region and NIR to get a large photocurrent are necessary.

III] Natural Dye:

Flowers, fruits, leaves of various plants and variety of microorganisms possesses different pigments which can be harvested and used in DSSC as a natural dye. Even though the performance natural dye is less as compared to Ru-based dyes they are explored significantly due to characteristics such as abundant occurrence, ecological compatibility, simple synthesis techniques and strong absorption coefficients in the visual range [42]. Chlorophyll, anthocyanins extracted from berries, carotenoids, flavonoids, betalains and so on are pigments exacted from different plants and are commonly used in DSSC as used a dye. Using beetroot dye an efficiency of 1.3% had been reported by Sathyajothi et.al. [43], Kumar et al. [44]

reported an efficiency of 0.11%. Yadav et al. [45] claimed enhanced light harvesting and charge transfer with Butea monosperma dye. Soni et al. [46] reported a substantial increase in efficiency by using C, N, and S co-doped TiO₂, from betalain dyes. Recent advancements in natural sensitizers primarily focus on increasing the light harvesting efficiency of DSSC without reducing environmental impact.

5. Practical Applications of DSSCs:

DSSCs have got significant attention in the field of internet of things (IoT), wearable and high integrated microelectronic devices, portable power supply and so on [47]. IoT is facilitating simplified, comfortable, and automated life to human being. For this IoT devices need an interrupted power supply in sensors and communication devices which can be provided by DSSC. DSSC also has an advantage of high performance in indoor and ambient light conditions. Hence, they can be used efficiently in the low illumination places such as low-light intensity conditions with light intensity 50 lux, dim living room with light intensity 200 lux, brightly-lit supermarkets with light intensity 1000 lux and are suitable in the ecology of such IoT applications. In the exponentially growing solar cell market; DSSC is a milestone in indoor lighting devices such as light-emitting diodes (LEDs), fluorescent light and incandescent light and in the automation technologies [48]. DSSC have proved their utility in sensors used in various applications like industrial automation, healthcare systems and environmental monitoring [49]. In biomeccal field, DSSCs are used in photodynamic therapy in which light sensitive drugs are used to prohibit the growth of cancerous cells. The light produced by device activates photosensitive drug to generate oxygen which kills the cancer cells [50]. In biomedical implants like pacemakers, drug delivery system DSSC can be used as a power supply [51]. As a DSSC has ability to generate electricity in indoor conditions, it is also used as a power source in the regions where diffused light predominates over direct solar light [52]. The translucent, thin sheets of DSSCs are installed in common windows, glass facades, skylights and buildings wall and turn them into electricity generators [53]. Multiple studies have shown that besides energy savings; DSSCs can be integrated with windows and other construction materials due to their attractive aesthetics.

6. Conclusion:

This review has demonstrated that DSSC represents a practicable and versatile photovoltaic technology with special characteristics in terms of cost, flexibility and adaptability to various lighting illuminations. Their ability to perform efficiently in low-light illumination environments makes it a potentially strong candidate in many applications related to indoor energy harvesting, building-integrated photovoltaics, portable devices and biomedical implants. Focussing continuous improvements in the key components such as photoanode and counter electrodes, electrolyte structures, dye sensitizers is still necessary to overcome the challenges. The ongoing research in novel dye combinations, alternative catalysts, fabrication methods and solid-state electrolytes show promising results. These interdisciplinary efforts made DSSC as an efficient alternative for photovoltaic solar cell technology and thus contribute in achieving sustainable development goals.

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