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# Natural Dyes as Pigments Harvesting Light for Solar Cells

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**ABSTRACT:** Natural photosynthetic systems contain several dyes such as carotenoids or chlorophylls which are adequately arranged to produce efficient photoinduced charge separation and electron transfer. Several research groups have attempted integrating these natural dyes and photosynthetic systems into functional organic solar cells (OSCs) producing power conversion efficiencies (PCEs) up to 0.99%. The studies presented in this short review emphasize that functionalization of natural dyes can considerably improve their PCEs. For instance, chlorophyll derivatives can yield PCEs up to 2.1%, and copolymers produced with isoindigo as an electron-deficient unit generate high PCEs up to 8%, respectively, when combined with fullerene  $C_{70}$  based electron acceptors in the OSC active layers. An alternative approach for natural dye integration into OSC architectures is to place these light-harvesting antennas at the interface between the active layer and the charge collection layers in these low-cost photovoltaic devices. This strategy produces large PCE increases up to 35% with respect to OSCs prepared without the interlayer. When light-harvesting systems are combined with silver nanoprisms as interlayers, additional localized surface plasmon resonance effects result in high-performance OSCs that integrate natural photosynthetic systems and demonstrate a PCE over the milestone value of 10%.

**KEYWORDS:** pigments, dyes, solar cells, light, photosynthetic, systems, copolymers

## I. INTRODUCTION

Dye-sensitized solar cell (DSSC) was assembled using natural dyes from chlorophyll extracted from spinach as a sensitizer. In this work, the adsorption characteristic has been studied in harvesting sunlight using different solvents. The effect of solvents has been investigated by analyzing the absorption spectrum, bandgap and absorption coefficient of the dyes. From the UV-Vis absorption spectrum, it has been known that chlorophyll extracted with distilled water has the broader region of the visible light spectrum in the range of 400 to 720nm compared to chlorophyll extracted with ethanol. The lowest bandgap of dye also presented by extracted the chlorophyll with distilled water with 1.83eV and the absorption coefficient of 1.59 km<sup>-1</sup>. The photo electrochemical parameter for solar cell by using chlorophyll extracted with DI water solvent showed the open circuit voltage (Voc) of 440mV, current short circuit (Isc) of 0.35mA and a fill factor (FF) of 0.49.[1,2]

The dye-sensitized solar cells (DSSC) deliver a low-cost and dependable alternative for various photovoltaic devices. The extraction process of natural pigments is simple and inexpensive compared with synthetic dyes. Natural plant pigments were extracted from flowers, leaves, roots, and fruits. Thus, this research focuses on the potential of natural dye using cold extraction with methanol. A UV-visible spectrometer was used for analyzing the Inthanin bok (Lagerstroemia macrocarpa) pigments absorption wavelength for the DSSC application. Carotenoids have the highest content, which is  $10.666 \pm 0.324$  µg/ml; followed by chlorophyll-a with  $2.708 \pm 0.251$  µg/ml and lastly chlorophyll-b with  $2.500 \pm 0.102$  µg/ml. Elemental of the TiO<sub>2</sub> nanoparticles and natural dyes was confirmed by energy-dispersive X-ray spectroscopy (EDX). The scanning electron microscope (SEM) and the laser scanning microscope used for analyzed morphological characteristics of TiO<sub>2</sub> nanoparticles and natural dyes. Moreover, the highest efficiency of the pigments extracted from Inthanin bok leaves is  $1.138\% \pm 0.018$ , which the condition of 1 layer of TiO<sub>2</sub> nanoparticles and the temperature; 300 °C.

UV-selective and transparent solar cells demonstrated a power conversion efficiency of 6.17%-8.13% under UV irradiation of 365 nm wavelength and 0.91%-2.70% under white light conditions. Additionally, it has a visible light



transmittance of 60.73%-58.22%, which shows that it is suitable for urban visual comforting without absorbing too much visible light and selectively absorbing more UV and near-infrared (NIR) lights for higher energy conversion. The window-fitted transparent dye-sensitized solar cells (DSSC) are an effective solution for selective absorption of solar irradiation of a particular range of wavelengths owing to the less occupation of land area, adequate visual comfort, and the ease of modification of their elemental compositions.[3,4] Currently, DSSC use metal-free organic dyes, inorganic ruthenium (Ru)-based dyes, perovskite-based sensitizers, quantum-dot sensitizers, and natural dyes sensitizers.

Especially, the metal-free organic dyes or so-called donor- $\pi$  bridge-acceptor (D- $\pi$ -A) sensitizers have garnered more attention since the last decade and become a promising alternative to Ru-based dyes owing to their very high molar extinction coefficients, free from expensive and toxic Ru metal, easily tunable absorption energies, ease of synthesis, low cost, high stability under elevated temperature, and prolonged luminescence. They consist of three parts viz. donors, linkers, and acceptors. [5,6] Under the irradiation of a specific wavelength, the electron-rich donor moieties give their electrons to the acceptors through  $\pi$ -bond-conjugated linker bridges. researchers synthesized three fluorene donor-based D- $\pi$ -A UV-sensitizing dye with different  $\pi$ -bridges made up of benzene, thiophene, and furan denoted as (E)-3-(4-(9H-fluoren-2-yl) phenyl)-2-cyanoacrylic acid (FPCN), (E)-3-(5-(9H-fluoren-2-yl) thiophen-2-yl)-2-cyanoacrylic acid (FTCN), and (E)-3-(5-(9H-fluoren-2-yl) furan-2-yl)-2-cyanoacrylic acid (FFCN), respectively, for window-fitted PV solar cell.[7,8]

Transparent fluorine (F)-doped tin oxide ( $\text{SnO}_2$ ) (FTO) glass substrates of 2.2 mm thickness and  $8 \Omega/\text{mm}^2$  were cleaned and pre-treated with  $\text{O}_2$  plasma for 10 min and diluted  $\text{TiCl}_4$  solution for 30 min at  $75^\circ\text{C}$ . After that,  $\text{TiO}_2$  paste was screen printed on the FTO glass substrate followed by sintering at  $300^\circ\text{C}$  for 30 min. This  $\text{TiO}_2$  layer acted as the anode of the PV cell (i.e., photoanodes). Subsequently, the photoanodes were dipped in a separate solution of dimethylformamide (DMF) containing 0.7 mM of FPCN, FTCN, and FFCN dyes. A platinum (Pt)-based electrocatalyst was spin-coated and thermally reduced on the FTO substrate using a solution of chloroplatinic acid ( $\text{H}_2\text{PtCl}_6$ ) and 2-propanol. The Pt layer acted as the cathode of the PV cell. In all three D- $\pi$ -A dyes, planar fluorene acted as the donor that enhanced the UV light-harvesting capacity in the short wavelength range, whereas the 2-cyanoacrylic acid acted as the acceptor. All donor molecules exhibited identical frontier molecular orbitals (FMOs) spatial distributions.[9,10]

The valence electrons were at the highest occupied molecular orbital (HOMO) state and spread from the donor molecule to the  $\pi$ -bridge; meanwhile, the excited electrons were localized at the lowest unoccupied molecular orbital (LUMO) state along the  $\pi$ -bridge from the donor molecule towards the acceptor molecule (cyanoacrylic acid). Hence, a continuous charge transfer occurred from the electron donor molecule to the electron acceptor molecule under UV light.[11]

## II. DISCUSSION

The absorption band were 315-400 nm for UV light and 400-420 nm for violet light spectra. The strongest absorption peak was associated with HOMO to LUMO intramolecular charge transfer (ICT). The molar extinction coefficients of FFCN, FTCN, and FPCN dyes in EtOH were 31,000, 32,400, and 36,200  $\text{M}^{-1}\cdot\text{cm}^{-1}$ , respectively, and the corresponding bandgaps were 2.79, 2.79, and 3.14 eV, respectively.[12,13]

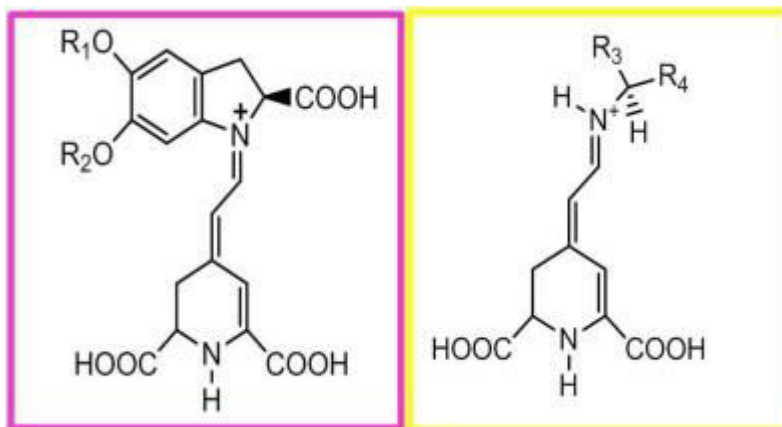
Furthermore, all three dyes exhibited a lower HOMO state than the redox potential of -4.80 eV, indicating high electron gain from the electrolyte and complete regeneration ability. Similarly, they exhibited a higher LUMO state than the conduction band of the  $\text{TiO}_2$  layer, i.e., -4.00 eV, indicating that all three dyes spontaneously and efficiently transferred exciting electrons into the  $\text{TiO}_2$  layer. The researchers synthesized metal-free organic so-called D- $\pi$ -A



UV-sensitizing transparent dye containing solar cells that can be fitted onto window glass, which can selectively harvest energy from UV light in a wavelength range of 315-420 nm without affecting the transmission of visible light.

The lower HUMO state than redox potential and higher LUMO state than the conduction band of the  $\text{TiO}_2$  layer indicated high regeneration ability efficient electron transfer efficiencies, respectively. Additionally, a high power conversion efficiency of 6.17%-8.13% for UV and high visible light transmittance of 60.73%-58.22% make it a promising dye for window-based solar cells. Dye-sensitized solar cells (DSSCs) are potentially economical and environmentally friendly alternatives to silicon based solar photovoltaics. However, reliance on ruthenium based metal-organic dyes to sensitize nanocrystalline  $\text{TiO}_2$  greatly mitigates their economic and environmental advantages. In our lab, we are replacing the costly ruthenium-containing sensitizers with plant pigments from the betalain family, which are found in plants of the order Caryophyllales: such as beets, bougainvillea, amaranth and cactus pear.[14,15]

Betalains are found in nature in two forms: as purple compounds, called betacyanins, or yellow ones called betaxanthins. The former vary as to the sugar group  $\text{R}_1$  or  $\text{R}_2$ , attached to the aromatic ring, and the latter are Schiff's base adducts of betalamic acid with various amino acids.



Using natural dyes as sensitizers has advantages and disadvantages. The potential advantage is that organic molecules can undergo two-electron, two-proton redox chemistry resulting in higher IPCE than a metal-centered dye which undergoes one-electron redox chemistry. However, the oxidation of organic compounds can be irreversible, which is detrimental to the stability of a DSSC.

1. Natural pigments are photosensitizers in dye-sensitized solar cells (DSSCs).
2. Efficiency is still lower compared to synthetic pigments.
3. The use of natural pigments such as carotenoids and polyphenols is cheap.
4. General advantages of DSSCs are flexibility, color and transparency.
5. Usage under diffuse light and therefore, indoor applications are possible.[16,17]



### III. RESULTS

Dye-sensitized solar cells (DSSCs) which are also called Graetzel cells are a novel type of solar cells. Their advantages are mainly low cost production, low energy payback time, flexibility, performance also at diffuse light and multicolor options. DSSCs become more and more interesting since a huge variety of dyes including also natural dyes can be used as light harvesting elements which provide the charge carriers. A wide band gap semiconductor like  $\text{TiO}_2$  is used for charge separation and transport. Such a DSSC contains similarities to the photosynthetic apparatus. Therefore, we summarize current available knowledge on natural dyes that have been used in DSSCs which should provide reasonable light harvesting efficiency, sustainability, low cost and easy waste management. Promising natural compounds are carotenoids, polyphenols and chlorophylls.[18]

Dye-sensitized solar cells (DSSCs) belong to the group of thin-film solar cells which have been under extensive research for more than two decades due to their low cost, simple preparation methodology, low toxicity and ease of production. Still, there is lot of scope for the replacement of current DSSC materials due to their high cost, less abundance, and long-term stability. The efficiency of existing DSSCs reaches up to 12%, using Ru(II) dyes by optimizing material and structural properties which is still less than the efficiency offered by first- and second-generation solar cells, i.e., other thin-film solar cells and Si-based solar cells which offer ~ 20–30% efficiency.

Dye-sensitized solar cells (DSSCs) have arisen as a technically and economically credible alternative to the p-n junction photovoltaic devices. In the late 1960s, it was discovered that electricity can be generated through illuminated organic dyes in electrochemical cells. At the University of California at Berkeley, chlorophyll was extracted from spinach (photosynthesis). First chlorophyll-sensitized zinc oxide ( $\text{ZnO}$ ) electrode was synthesized in 1972. For the first time, through electron injection of excited dye molecules into a wide band gap of semiconductor, photons were converted into electricity [1]. A lot of research has been done on  $\text{ZnO}$ -single crystals [2], but the efficiency of these dye-sensitized solar cells was very poor, as the monolayer of dye molecules was able to absorb incident light only up to 1%. Thus, the efficiency was improved by optimizing the porosity of the electrode made up of fine oxide powder, so that the absorption of dye over electrode could be enhanced and as a result light harvesting efficiency (LHE) could also be enhanced. As a result, nanoporous titanium dioxide ( $\text{TiO}_2$ ) electrodes with a roughness factor of ca.1000 were discovered, and in 1991, DSSCs with 7% efficiency were invented [3]. These cells, also known as Grätzel cells, were originally co-invented in 1988 by Brian O'Regan and Michael Grätzel at UC Berkeley [3] and were further developed by the aforementioned scientists at Ecole Polytechnique Fédérale de Lausanne (EPFL) till 1991.

Brian O'Regan and Michael Grätzel fabricated a device based on a 10- $\mu\text{m}$ -thick, high surface area and optically transparent film of  $\text{TiO}_2$  nanoparticles, coated with a monolayer of a charge transfer dye with ideal spectral characteristics to sensitize the film for light harvesting. The device harvested a high proportion of the incident solar energy flux of 46% and showed exceptionally high efficiencies, even more than 80% efficiencies for the conversion of incident photons to electrical current. The overall incident photon to current conversion efficiency (IPCE) yield was 7.1–7.9% in simulated solar light and 12% in diffuse daylight. A large short circuit current density  $J_{\text{SC}}$  (greater than  $12 \text{ mAcm}^{-2}$ ) and exceptional stability (sustaining at least five million turnovers without decomposition) and low cost made the practical application feasible [3]. In 1993, Grätzel et al. reported 9.6% efficiency of cells, and then in 1997, they achieved 10% at the National Renewable Energy Laboratory (NREL). The sensitizers are usually designed to have functional groups such as  $-\text{COOH}$ ,  $-\text{PO}_3\text{H}_2$ , and  $-\text{B}(\text{OH})_2$  for stable adsorption onto the semiconductor substrate [4, 5]. Recently in 2018, an efficiency of 8.75% was reported for hybrid dye-titania nanoparticle-based DSSC for superior low temperature by Costa et al. [6]. In a traditional solar cell, Si provides two functions: acts as source of photoelectrons and provides electric field to separate the charges and create a current. But, in DSSCs, the bulk of semiconductor is only used as a charge transporter and the photoelectrons are provided by photosensitive dyes. The theoretically predicted power conversion efficiency (PCE) of DSSCs was



approximately 20% [7, 8]; thus, an extensive research has been made over the years on DSSCs to improve the efficiency and to augment its commercialization. However, in the last few decades, a lot of experiments were carried out to improve the performance of DSSCs. For instance, if one goes through the review articles or papers published around 1920 and 1921, a remarkable difference may be observed in the performance as well as fabrication of these cells. Few review papers are discussed below with the objective and main results shown in a respective article to get an idea how the performance of these cells has been improved and, thus, how the DSSCs became a hot topic for researchers.[19]

Anandan reviewed the improvements and arising challenges in dye-sensitized solar cells till 2007 [9]. The main components of his review study were light harvesting inorganic dye molecules, p-CuO nanorod counter electrodes, and self-organization of electroactive polymers, and he showed how these materials perform in a rationally designed solar cell. However, the maximum IPCE of 7% was discussed in the review paper for naphthylidine coordinated Ru complex [10] which was good till 2007 but is almost half to the efficiencies shown in later work.

#### IV. CONCLUSIONS

The main aim of this study was to put a comprehensive review on new materials for photoanodes, counter electrodes, electrolytes, and sensitizers as to provide low-cost, flexible, environmentally sustainable, and easy to synthesize DSSCs. However, a brief explanation has been given to greater understand the working and components of DSSCs. One of the important emphases in this article has been made to establish a relation between the photosensitizer structure, the interfacial charge transfer reactions, and the device performance which are essential to know as to develop new metal and metal-free organic dyes. In terms of low stability offered by DSSCs, two major issues, i.e., low intrinsic stability and the sealing of the electrolytes (extrinsic stability), have been undertaken in this study. To fulfill huge demand of electricity and power, we have two best possible solutions: this demand should be compensated either by the nuclear fission or by the sun. Even so, the nuclear fission predicted to be the best alternative has great environmental issues as well as a problems associated with its waste disposal. Thus, the second alternative is better to follow. DSSCs are developed as a cheap alternative but the efficiency offered by DSSCs in the field is not sufficient. Thus, we have to do a wide research on all possible aspects of DSSCs. We proposed to develop DSSCs based on different electrodes viz. graphene, nanowires, nanotubes, and quantum dots; new photosensitizers based on metal complexes of Ru or Os/organic metal-free complexes/natural dyes; and new electrolytes based on imidazolium salts/pyridinium salts/conjugated polymers, gel electrolytes, polymer electrolytes, and water-based electrolytes. In summary, so far, extensive studies have been carried out addressing individual challenges associated with working electrode, dye, and electrolytes separately; hence, a comprehensive approach needs to be used where all these issues should be addressed together by choosing appropriate conditions of electrolyte (both in choice of material and structure), optimum dye, and the most stable electrolyte which provides better electron transportation capability.[18,19]

In terms of their commercial application, a DSSC needs to be sustainable for >25 years in building-integrated modules to avoid commotion of the building environment for repair or replacement and a lifespan of 5 years is sufficient for portable electronic chargers integrated into apparel and accessories [19]. However, DSSCs are being quite bulky due to their sandwiched glass structure, but the flexible DSSCs (discussed elsewhere) that can be processed using roll-to-roll methods may come as an alternative but then has to compromise with the shorter lifespan. Although the stability and lifetime of a DSSC most probably depend on the encapsulation and sealing as discussed above. Apart from the usage of expensive glass substrates in the case of modules and panels, one of the biggest hurdles is to manufacture glass that is flat at the 10  $\mu\text{m}$  length scale over areas much larger than  $30 \times 30 \text{ cm}^2$  [18] and the humidity. Another challenge is to choose which metal interconnects in the cells that are more or less corroded to the electrolyte, and high degree of control over cell-to-cell reproducibility is required to achieve same current and/or voltage for all the cells in the module. If the abovementioned challenges would be overcome, then there is no roadblock for the commercial applications of DSSCs, which has been restricted up to an



amicable extent. G24i has introduced a DSC module production of 25 MW capacity in 2007 in Cardiff, Wales (UK), with extension plans up to 200 MW[20]

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