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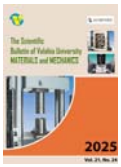

DYES FOR DYE-SENSITIZED SOLAR CELLS (DSSCs): A REVIEW

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Abstract: *Dye-Sensitized Solar Cells (DSSCs), also known as Grätzel cells, represent a promising technology in the field of solar energy. These cells convert solar energy directly into electrical energy in an efficient and environmentally friendly manner. A crucial component of these cells is the dye. The dye, essentially, acts as a molecular antenna, absorbing sunlight and thus initiating the energy conversion process. The absorption of a photon by the dye molecule elevates an electron to a higher energy level.This excited electron is then injected into a semiconductor, typically titanium dioxide (TiO_2), which acts as an electrode. The circuit is completed as an electron from the electrolyte replenishes the electron deficiency in the dye.*

Keywords: *Dye-sensitized solar cells (DSSCs), sensitizer, energy conversion.*

1. INTRODUCTION

Solar energy provides a multitude of benefits compared to other energy sources and it is a zero-emission, waste-free, and endlessly renewable energy source [1].

Solar cells convert light photons into electricity through the photovoltaic effect. There are two main types of solar cells: organic and inorganic cells. Traditional silicon-based solar cells are widely favored in the market for their long lifespan and high energy conversion efficiency [2-3]. While silicon solar cells offer significant advantages, they also have certain limitations. The manufacturing process is energy-intensive, their efficiency decreases at higher temperatures and the availability of high-purity silicon is becoming a growing concern [4]. In response to these challenges, the development of innovative materials, particularly organic materials, has emerged as a key focus of ongoing research in photovoltaics.

Organic solar cells can be primarily categorized into three types: dye-sensitized solar cells (DSSCs), polymer heterojunction solar cells (PSCs) and perovskite solar cells (PVSCs). DSSCs, in particular, offer significant potential due to their high efficiency and suitability for low-temperature fabrication processes [5].
Due to their high efficiency in low-light conditions, DSSCs are ideal for indoor use. Furthermore, they offer a broad spectrum of synthetic design possibilities, compatibility with diverse materials, and a low environmental footprint. One of the major cost factors in DSSCs is the use of synthetic dyes, particularly ruthenium complexes.
These dyes necessitate toxic raw materials and are involved in manufacturing processes.
Natural dyes, derived from plant sources like fruits, flowers, and leaves, present a promising alternative. However, their stability under light exposure must be carefully considered.

Natural dyes offer several advantages over synthetic dyes, including abundance, affordability, non-toxicity, and environmental friendliness. By utilizing co-sensitization strategies, the efficiency of DSSCs can be significantly enhanced.

2. SENSITIZERS (DYES) FOR DSSCs

A variety of photoactive sensitizers (dyes) can be used in DSSC cells, as follows:

2.1. Natural Dyes

Over the past two decades, research efforts to utilize natural dyes as sensitizers in DSSCs have yielded modest results. Most studies report efficiencies below 2% [6-8], with a few reaching 5% [9]. While natural dyes are abundant, environmentally friendly, and cost-effective, their low stability, tendency to aggregate, and weak

binding to semiconductor surfaces hinder their performance. The propensity of natural dyes to aggregate significantly hinders charge transport, limiting their overall performance [10]. The majority of natural dyes form weak bonds with the surface of the semiconductor oxide [11-12].

Betanin is a pigment that belongs to the betaxanthin group, which in turn belongs to the betalain class. The structure of betanin is presented in Figure 1.

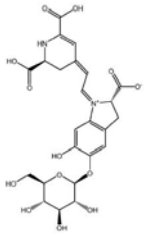

Figure 1. Chemical structure of betanin [13]

Betalains, a group of water-soluble pigments, possess an N-heterocyclic structure with a hydroxyl group. They are classified into two main types: betacyanins, responsible for red-violet coloration, and betaxanthins, which impart yellow hues. Betanin, a predominant betacyanin, can be sourced from various plants within the Caryophyllales order, including beetroot and carnation [14-15].
The spectral characteristics of betanin and related compounds make them promising candidates for DSSCs sensitizers. [16-18].

Several experimental studies have been conducted to explore the intramolecular electronic properties of betanin and to elucidate the mechanism by which electrons are injected from betanin onto the TiO_2 surface. Knorr et al. reported that free radicals are formed during the electron recombination process, and these radicals cause the oxidation of betanin [19]. Damayanti et al. demonstrated that one of the three carboxyl groups in the betanin molecule inhibits the deprotonation process, thereby hindering efficient electron injection from the dye to the semiconductor oxide surface [20].

Chlorophylls are a category of pigments also capable of absorbing light. These pigments absorb wavelengths corresponding to the colors violet, blue, and red and reflect green light.
Therefore, the presence of chlorophyll is responsible for the green color of plants.
Through the process of photosynthesis, chlorophylls are able to convert the radiant energy of the sun into the chemical energy of organic compounds in the cell.
These pigments are composed of a tetrapyrrolic ring structure with a central magnesium ion (Mg_{2+}).

Depending on the structure of the ligand, there are two main types of chlorophyll, namely: chlorophyll-a and chlorophyll-b, the chemical structure of which is presented in Figure 2.
Chlorophyll-b differs from chlorophyll-a in that it has an aldehyde group (-CHO) instead of a methyl group (-CH3).

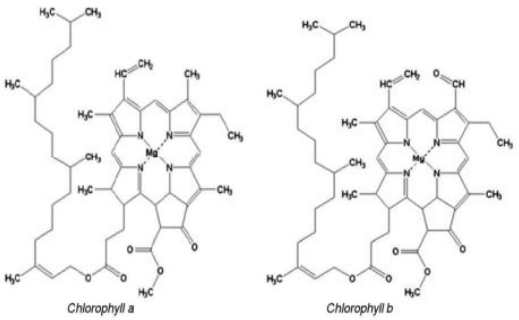

Figure 2. Chemical structure of chlorophyll [21]

Chlorophyll-containing leaf extracts were first used in DSSC cells by Kay et al. in 1993. Additionally, they investigated the performance of a modified DSSC where the central Mg_{2+} ion was substituted with a Cu_{2+} ion. The cell parameters were reported as follows: short-circuit current $I_{SC} = 9.4$ mA/cm², open-circuit voltage $V_{OC} = 0.52$ V and conversion efficiency = 2.6%.

This results was obtained by incorporating polyhydroxyl compounds, such as glucose, glycerol or sorbitol into the adsorption process. [22].
While Kay et al. were the first to demonstrate chlorophyll-based DSSCs, Kamat et al. preceded them by reporting TiO_2 sensitization with Cu-chlorophyll in 1986. They employed laser photolysis, tunable microwave absorption and spectrofluorometry to elucidate the underlying photoelectrochemical mechanisms in the sensitized TiO_2 system [23].
In the subsequent decade, Amao et al. continued the exploration of chlorophyll-based DSSC cells. They employed a chlorophyll derivative as a sensitizing dye, but the results were underwhelming. The short-circuit current density was 0.305 mA/cm², and the open-circuit voltage was 0.426 V [24].
Wang et al. [25] reported the development of chlorophyll-based DSSCs employing a modified chlorophyll photosensitizer. The introduction of a conjugated redox spacer minimized electron recombination with the oxidized dye, leading to a significant 30% improvement in device efficiency [26].
When comparing the performance of DSSC cells utilizing chlorophyll with TiO_2 and ZnO photoanodes, it was determined that the lower conversion efficiency of chlorophyll-ZnO-based cells is due to the significant distance between the anchoring site and the molecular orbitals responsible for electron excitation [27].
Although these cells exhibit lower efficiency, the crucial role of chlorophyll in natural photosynthesis has stimulated ongoing research efforts in the field of chlorophyll-based DSSCs.

Anthocyanins, another class of natural dyes found in various colored fruits and flowers, have also been explored for DSSC applications. Plants that are rich in anthocyanins include blueberries, blackberries, raspberries, blackcurrants, aronia, red cabbage, watercress, eggplant, etc. The chemical structure of anthocyanins is presented in Figure 3.

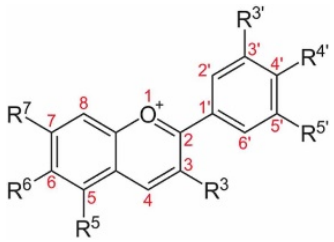

Figure 3. Chemical structure of anthocyanidins [28]

There are three fundamental types of natural anthocyanins: pelargonidin, cyanidin and peonidin [29], which differ in the number of hydroxyl groups in the phenyl ring, to these, three other methyl ethers are added, namely peonidin (a derivative of cyanidin with CH_3O in the 3' position), syringidin (a derivative of delphinidin with CH_3O groups in the 3' and 5' positions), and hirsutidin (a derivative of delphinidin with CH_3O groups in the 7', 3', and 5' positions).

Apigenidin, which is less common, is a derivative of pelargonidin without a hydroxyl group in the 3 position. The functional groups, particularly the carboxyl groups, can act as anchoring sites to facilitate the adsorption of the dye molecule onto the semiconductor surface. Catechol groups, in particular, exhibit enhanced anchoring ability compared to carboxyl groups [30].
The main published results regarding the use of anthocyanins as sensitizers for DSSCs highlight the fact that their performance is low compared to that of chlorophyll-derived dyes [31-32].

The integration of natural dyes into functional DSSC devices remains a significant challenge. Among the various natural dyes, chlorophyll derivatives have garnered the most attention owing to their superior performance.

2.2. Ruthenium-based Organic Dyes

Ruthenium-based organic dyes have emerged as promising photosensitizers for DSSCs owing to their favorable photoelectrochemical properties and enhanced stability in the oxidized state. These attributes render devices employing such dyes highly competitive in the solar cell market [33]. The ruthenium-based dye N3 was first used in DSSC cells by Nazeeruddin et al. in 1993. Thus, the obtained DSSC cells reported a conversion efficiency of 10.3%. This dye has only one ruthenium atom in its chemical structure (Figure 4).

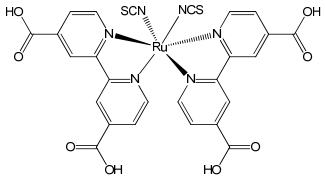

Figure 4. The chemical structure of the N3 ruthenium sensitizer [34]

In the following period, Nazeeruddin et al. used another ruthenium-based organic dye in DSSC cells, which they named "black dye," and the efficiency of the obtained cells was 10.4%. The chemical structure of this dye is presented in Figure 5.
This dye has an absorption spectrum that covers the entire visible spectrum, and its name was attributed to the fact that it has a green color in solution and a black color on the surface of TiO_2 nanoparticles [35].

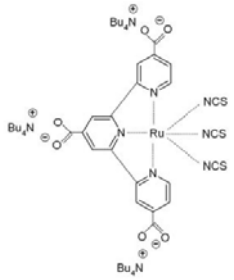

Figure 5. The chemical structure of the 'black dye' (Bu4N - tetrabutylammonium) [36]

Nazeeruzzin et al. also synthesized another dye from the same family, N179 (Figure 6).
DSSC cells using this dye achieved a conversion efficiency of 11.2%, surpassing previous records for ruthenium-based dyes. [37].

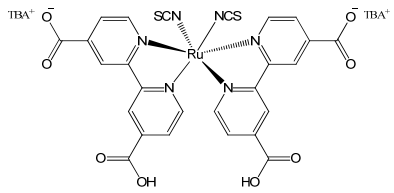

Figure 6. The chemical structure of the N179 dye, including the tetrabutylammonium cation (TBA+) [38]

Currently, the most advanced DSSCs, which utilize ruthenium(II) polypyridyl complexes as sensitizers, typically exhibit energy conversion efficiencies surpassing 13% under standard testing conditions [39]. This high efficiency can be attributed to their broad absorption spectrum, which encompasses the visible and near-infrared regions.
The presence of carboxylate groups on the bipyridyl moiety results in a red-shift of the ligand's π-π* absorption band, leading to a lower energy gap and facilitating electron injection into the semiconductor conduction band.

2.3. Porphyrin-Based Dyes with Metal Cations

The word „porphyrin" has Greek origins, deriving from the ancient Greek word „porphura", used to describe the purple color. Porphyrins represent a broad class of compounds, both natural and synthetic, sharing a common aromatic substituent consisting of a tetrapyrrolic macrocycle linked by four methine bridges [40].
They are crystalline compounds, often intensely red or

purple in color, frequently exhibiting fluorescence, and are used as pigments.

The structure of porphyrins was first proposed by Kuster et al. in 1912 and later by Hans Fisher et al. According to Fisher nomenclature, each position of the pyrrole fragments where a substituent could be attached was labeled β-carbon and assigned a number from 1 to 8.
The carbon atoms serving to unite these pyrrole rings are called meso-carbons, and they were assigned Greek letter notation, α-β [41].
The porphyrin macrocycle consists of two pyrrole units and two pyrroline units.

As in any aromatic molecule, there is a number of delocalized π electrons within the porphyrin macrocycle, in this case, 22 π electrons that follow Hückel's rule of aromaticity (4n+2), (n=5). However, only 18 π electrons are effectively part of a delocalized electron cycle, so Hückel's rule (4n+2) leads to an aromatic system with n=4. The other 4 π electrons are excluded from the count of delocalized electrons [42].
Figure 7 shows the chemical structure of the porphyrin macrocycle.

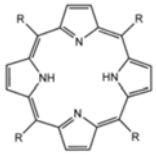

Figure 7. The chemical structure of the porphyrin macrocycle [43]

Absorption at long wavelengths of these chromophores allows the exploration of potential applications of porphyrins in various fields involving photochemical stimulation. Thus, porphyrin dyes are very often used in the DSSC cells manufacture.

2.4. Organic dyes without metal cations

This category of dyes is very diverse and includes dyes such as: phenothiazine, coumarin, indole, hemicyanine dyes and others. Some promising results have already been obtained for these dyes, many of them determining efficiency values greater than 5%.
Thus, the reported conversion efficiency values for DSSC cells are presented in Table 1.

Table 1. Efficiency of DSSC cells incorporating organic dyes without metal cations

Dye type	The reported efficiency η [%] [44]
Fluorene dye	5.23
Hemicyanine dye	5.1
Phenothiazine dye	5.5
Coumarin dye	8.2
Indole dye	9.03

3. CONCLUSIONS

To optimize the performance of a dye-sensitized solar cell, the dye must exhibit the following essential properties:
Strong visible spectrum absorption: A dye's light-harvesting capacity is a critical factor determining the overall efficiency of a solar cell.
Rapid electron injection: The excited electron must be promptly transferred from the dye molecule to the semiconductor to ensure efficient energy conversion.
Rapid regeneration: The electron hole in the dye must be quickly filled by an electron from the electrolyte.
Stability: The dye must be stable under both light and the operating conditions of the solar cell.
Low cost: A low cost of the dye is essential for the widespread commercialization of DSSC technology.
Researchers worldwide are currently working to develop new dyes with improved performance.
Some of the most important research directions include:
Broad absorption spectrum dyes: These dyes would improve the solar cell's spectral response by extending its absorption capabilities.

Improved dyes stability: Greater stability would lead to a longer lifetime of solar cells.
Low-cost dyes: Developing cheaper dyes is essential to make DSSC technology competitive.
Natural dyes: Researchers are exploring the potential of using natural dyes to reduce costs and environmental impact.

Dyes are essential to dye-sensitized solar cells. By understanding the requirements for an ideal dye and developing new materials, researchers are moving closer to the widespread commercialization of this promising technology.

In conclusion, the interdisciplinary nature of organic solar cell research presents ongoing challenges. Ongoing research in this field is essential for the development of the next generation of renewable energy cells.

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