

## DYE PHOTOSENSITIZERS AND THEIR INFLUENCE ON DSSC EFFICIENCY: A REVIEW

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**Abstract.** Since early 1990s dye-sensitized solar cells (DSSC) has been developed by many research groups all over the World. This paper presents a review of researches focusing on photosensitizer influence on DSSC efficiency. Variety of dye substance has been analyzed. The highest efficiency around 11.2% has been noted for ruthenium-based DSSC devices. Natural dyes allowed to reach 4.6%. The most metal-free organic dyes resulted in efficiency ranged from 5% to 9%, however, some of them (e.g. Y123) allowed to obtain devices with efficiencies equal to 10.3%. Co-sensitization is the new approach which results in efficiencies up to 14.3%.

**Keywords:** photovoltaic cells, renewable energy sources, energy conversion, DSSC

### SUBSTANCJE SENSYBILIZUJĄCE I ICH WPLYW NA SPRAWNOŚĆ BARWNIKOWYCH OGNIW SŁONECZNYCH (DSSC): PRZEGLĄD

**Streszczenie.** Od początku lat 90 XX wieku ogniwa barwnikowe przyciągają uwagę naukowców na całym świecie. Praca ta poświęcona jest przeglądowi badań dotyczących wpływu substancji sensybilizujących na sprawność barwnikowych ogniw słonecznych (DSSC). Największą sprawność uzyskują ogniwa sensybilizowane barwnikami na bazie rutenu, podczas gdy barwniki naturalne pozwalają na pracę z wydajnością 4,6%. Sprawność konwersji energii ogniw uczulanych barwnikami organicznymi wynosi 5-9%, jednakże niektóre z nich, np. Y123 pozwalają na uzyskanie wydajności rzędu 10,3%. Zastosowanie kilku barwników do sensybilizacji jest nowym podejściem, które przekłada się na wartości sprawności nawet do 14,3%.

**Słowa kluczowe:** ogniwa fotowoltaiczne, odnawialne źródła energii, konwersja energii, barwnikowe ogniwa słoneczne (DSSC)

### 1. Introduction

Nowadays renewable energy sources are becoming more important due to the intense development of society combined with the depletion of fossil fuels. It should be noticed that energy demand is still increasing. One of the reasons is a tremendously growing population and significant improvement of its living standard. Another reason is the industry enhancement. Electricity is a core resource for the development of human civilizations. It is anticipated that power demand increases by 90% from today to 2040, because of the use of electric mobility, heating, and common electricity access [46, 55]. However, the current scheme of supply and use of energy is unsustainable and it should be stabilized in the short-time perspective. What is more, global warming and the climate change force the present generation to search environmentally friendly energy technologies which will contribute to sustainable development for the subsequent generations. Electricity can be received from various resources such as coal, natural gas or biomass. It should be taken into account that fossil fuels combustion has a tremendous impact on the environment. Environment preservation during the energy production process is crucial. Therefore, the energy production method should be reoriented towards renewable resources. Greenhouse gases emission, such as CO<sub>2</sub>, SO<sub>x</sub>, NO<sub>x</sub>, to the atmosphere is limited by Directive 2009/28/EC of the European Parliament and of the Council of 23 April 2009 on the promotion of the use of energy from renewable sources which assumes the emission reduction. What is more, European Council has presented a variety of legislative proposals related to the reform of the EU energy market called "Clean Energy for All Europeans". On the basis of these documents, Poland should achieve 32% of renewable energy sources in final energy consumption by the end of 2030 [54]. The renewable energy sources are characterized by high temporal and spatial variability thus they need to be carefully balanced and used in the national energy plan in order to maintain a well-balanced supply. Solar photovoltaic technology has tremendous high potential energy contribution which can fulfil the energy demands of the entire world. Therefore, and having regard to this fact, harvesting and supplying photovoltaic (PV) technologies should be readily available [7]. Photovoltaic use the direct conversion technology of sunlight into high-quality electricity energy. Three main categories of photovoltaic solar cells, called generation, can be distinguished. Currently, the market is covered by the first two. The most-known, the first

generation is based on crystalline silicon material. The silicon wafers production and environmental costs are relatively high, but this technology is the most effective one. The research group of Yoshikawa [50] achieved efficiency equals 26.7%. The second generation of solar cells, which is called thin-film technology, is mainly based on cadmium sulfide (CdS), cadmium telluride (CdTe), amorphous silicon (a-Si) and copper-indium-diselenide/copper-indium-gallium-diselenide (CIS/CIGS). P. Jackson et al. [18] produced CIGS laboratory solar cell with the highest efficiency of 22.4%. It has created the opportunity to replaced high-cost conventional silicon solar cells with CIGS ones. However, the type of solar cells which focuses the attention of research groups all over the world are dye-sensitized solar cells (DSSC). They belong to the third generation of photovoltaic cells which is classified as a cost-effective photovoltaic device mainly due to inexpensive materials and simple fabrication process. After the breakthrough, in 1991, made by the group of Gratzel [32], DSSC are considered as one of the most potential renewable power sources. In contrast to conventional solar cells based on the p-n junction made of semiconductor, DSSC consist of four main components:

- 1) photoanode, called working electrode, with a mesoporous semiconductor layer (mostly made up of titanium dioxide (TiO<sub>2</sub>));
- 2) counter electrode covered by a catalyst which is typically platinum;
- 3) electrolyte containing the redox couple; its main role is to collect electrons at the counter electrode and dye sensitizer-regenerating;
- 4) dye sensitizer absorbed on the TiO<sub>2</sub> molecules.

The scheme of dye-sensitized solar cells is presented in Fig. 1.

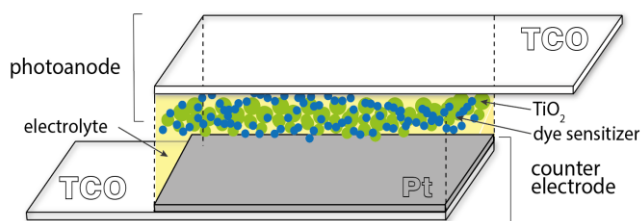


Fig. 1. Scheme of dye-sensitized solar cells

Dye-sensitizer solar cells are photoelectrochemical devices which use an electrolyte as the charge transport medium. Both electrodes are based on transparent conductive oxide (TCO) substrates. They are covered by several microns thick of mesoporous and nanostructured indium tin oxide (ITO) [38] or fluorine-doped tin oxide (FTO) [26]. In the process of manufacturing the DSSC, FTO material is more common, because is high temperature resistant which is important since the working electrode covered with TCO and semiconductor needs annealing treatment. At relatively high temperature degradation of electrical properties of ITO layers can appear, whilst layer based on fluorine-doped tin oxide is much more stable. ITO is characterized by the highest available optical transmittance of ~90% in visible spectrum light with the lowest electrical resistivity of  $\sim 10^{-4} \Omega \cdot \text{cm}$  [28]. Among all the components, the dye plays a crucial role in the conversion of sunlight into electricity [1, 19]. Its role is to extend the absorption of solar light by mesoporous  $\text{TiO}_2$  layer from visible to the near-infrared range (up to 920 nm) in the solar spectrum and thus increasing the efficiency of solar conversion [10]. It should be pointed out that DSSC use electron transfer reactions in a molecular scale. The initial efficiency that achieved by Gratzel equals 7% in 1991 [32], then it raised up to the value of 10% in 1993. Nowadays, the most efficient DSSC device works with an efficiency of 13% [30]. The attention on many research groups is focused on clarification of DSSC working principles, improvement of conversion efficiency, and commercialization.

Dye-sensitized solar cells have many components that have to be optimized not only as an individual but also as a part of the highly-advanced assembly. The efficiency of working cells absolutely depends on the type of substance used as dye sensitizer. This review article aims to represent recent studies of dyes that have provided important pathways towards better device performance.

## 2.1. Dye sensitizer

The efficiency of dye-sensitized solar cells is strongly determined by the molecular structure of the photosensitizer [43]. Roslan et al. [34] divided the photosensitizers into three groups: metal-complex, organic and natural photosensitizer. However, some research groups, such as [3, 17, 24] distinguish into natural and synthetic substances (metal complex dyes and metal-free organic dyes). The group of synthetic sensitizers is represented by e.g. polyphyrilal complexes of ruthenium and osmium, metal porphyrin, whereas organic one includes natural and synthetic substances. However, the principal criteria that are taking into account are an environmental influence, manufacturing, and origin of the dyes, ability of absorbance the wide spectra of the light and durability. The whole surface of the nanoparticles titanium dioxide should be covered with a single layer of molecules' dye which causes the effective electron injective into the semiconductor material directly. It would be low effective or even impossible if it was realized through the few layers of sensitizer.

What is more, an ideal dye sensitizer should fulfil important requirements, which are as follows [12, 49, 52]:

- 1) wide absorption spectrum; maximum photons harvesting from visible to the near-infrared region;
- 2) production of photo-excited electrons;
- 3) the ability of effective charge infusion into the conduction band of the mesoporous semiconductor layer;
- 4) appropriate levels of LUMO and HOMO providing effective electrons injection;
- 5) ability to dye regeneration; the oxidation potential of dye should be higher than the oxidation potential of redox couple contained in an electrolyte;
- 6) presence of chemical groups that are capable of permanent chemisorption of photosensitizer onto the  $\text{TiO}_2$  surface, such as  $-\text{COOH}$ ,  $-\text{H}_2\text{PO}_3$ ,  $-\text{SO}_3\text{H}$ ;
- 7) thermal and chemical stability, good solubility.

Metal complex dyes that have been widely investigated are based on ruthenium (Ru), iridium (Ir), osmium (Os). These elements are considered as heavy metals; complexes which they create are characterized by a long excited lifetime, high efficiency of DSSC assembly, and high redox properties.

## 2.2. Ruthenium (Ru) complex photosensitizers

Nowadays, the DSSC assemblies that occur the best efficiency are ruthenium-based. They are characterized by high efficiency, excellent chemical stability, and absorption of solar radiation in a wide range. Researchers developed many sensitizers based on ruthenium element. Among them, the most well-known and widely used are N3 ("red" dye), N749 ("black" dye), N719, Z907 complexes. N719 and N3 photosensitizers have almost the same structure; in the molecule of N3 is  $\text{TBA}^+$  instead of  $\text{H}^+$  at two carboxyl groups. The red and black dyes are often used as indicator ones. Many researchers [5, 11, 25] are working on N719-based dye-sensitized solar cell assemblies, but the highest efficiency equals 11.18% was obtained by [14]. The most important thing during the optimization of the photosensitizer for solar cells applications is the ratio of electrons injection rates and recombination (reverse and forward electron transport) [13]. High efficiency of the N3 and N719 dye is the result that the injection process is much faster than recombination due to the separation of the donor LUMO orbital and the acceptor HOMO. However, all of Ru-based DSSC obtain similar, excellent efficiency on the level of 10-11%, which is shown in Table 1. That is caused especially by wide absorption range from visible to near-IR light. In the last years, in the field of dye-sensitized assemblies based on ruthenium, the stagnation can be observed. It is caused by a disadvantageous impact on the environment by their chemical nature. In addition, the synthesis of ruthenium complexes is a sophisticated and expensive procedure, and these complexes have a strong tendency to degrade in the water environment [51].

Table 1. The efficiency of the Ru-based dye-sensitized solar cells

Dye	Full name	$\eta$ [%]	Ref.
N3	cis-bis(isothiocyanato)bis(2,2'-bipyridyl-4,4'-dicarboxylato)ruthenium(II)	10.00	[48]
N719	di-tetrabutylammonium cis-bis(isothiocyanato)bis(2,2'-bipyridyl-4,4'-dicarboxylato)ruthenium(II)	11.18	[14]
N749	tris(N,N,N-tributyl-1-butanaminium)[[2,2''6',2''-terpyridine]-4,4',4''-tricarboxylato(3-)-N1,N1',N1'']tris(thiocyanato-N)hydrogen ruthenate	10.40	[33]
Z907	cis-Bis(isothiocyanato)(2,2'-bipyridyl-4,4'-dicarboxylato)(4,4'-di-nonyl-2'-bipyridyl)ruthenium(II)	11.10	[6]

Mathew et al. [30] achieved high conversion efficiency for the metal complex-based dyes which was 13% with the following working parameters: open-circuit voltage  $V_{OC} = 0.91 \text{ V}$ , short-circuit current density  $J_{SC} = 18.1 \text{ mA/cm}^2$ , fill factor  $FF = 78\%$ .

## 2.3. Natural dyes

Natural dyes have received intense research interest in recent years. They are considered as promising photosensitizers in dye-sensitized solar cells field because of low cost, resource reproducibility, non-toxic environmental impact, and their abundance of prevalence. The biggest advantage of dyes based on natural substances is that they are never threatened with extinction, even with very high usage. Pigments in natural dyes can be easily extracted from flowers, leaves, and fruits of some plants [9]. Each part of the natural plant is characterized by different pigment colour. What is more, obtaining natural dyes and optimization of extraction methods are less sophisticated than in case of ruthenium complexes, and thus the negative impact on the natural environment can be easily reduced [4].



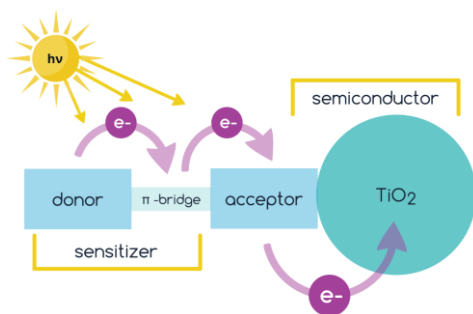


Fig. 4. Scheme of donor- $\pi$ -acceptor organic dye structure

The main factor which can limit the efficiency of working DSSC sensitized with organic dyes is the occurrence of dye aggregates formation on the surface of  $\text{TiO}_2$  nanoparticles which caused suppressing of the excited state of the sensitizer molecules.

It can be distinguished many metal-free organic dyes classes which are the subject of investigation. Among them, alizarin, coumarin, cyanine, triphenylamine, carbazole-based sensitizers which obtained the efficiency up to 5–9% [37, 52] can be underlined.

Alizarin is one of the metal-free organic dyes which results in red colour in DSSC. This dye is characterized by rapid injection electrons into the semiconductor conduction band with the indirect mechanism. The dye has wide absorption (400–600 nm) with the strongest peak at 432.5 nm [22]. The molecular structure of alizarin is shown in Fig. 5.

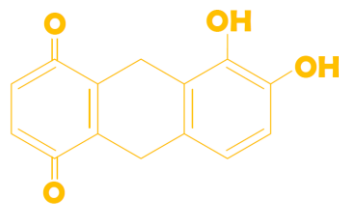


Fig 5. Basic molecular structure of alizarin pigment

In the paper by [35], the working parameters of alizarin-based DSSC with four different redox couple electrolytes e.g. (KI),  $[(\text{CH}_3)_4\text{NI}]$ ,  $[(\text{CH}_3\text{CH}_2)_4\text{NI}]$  and  $[(\text{CH}_3\text{CH}_2\text{CH}_2)_4\text{NI}]$  were examined. They achieved the dye-sensitized assemblies worked with the efficiency of 0.635%, 0.383%, 0.78%, and 0.915%.

Whereas, the research group of Manmeeta [29] fabricated two types of assemblies:  $\text{TiCl}_4$  treated and untreated  $\text{TiO}_2$  material with the use of oxidized alizarin photosensitizer. In the first group, they obtained  $J_{SC}$ ,  $V_{OC}$ ,  $FF$  and efficiency parameter equals to  $9 \text{ mA/cm}^2$ ,  $750 \text{ mV}$ ,  $0.53$  and  $3.57\%$  respectively. However, the research has shown that  $\text{TiCl}_4$  treatment significantly improves all working parameters, especially energy conversion efficiency (5.12%).

Tsao et al. [45] reported the power conversion efficiency of 10.3% by use of organic dye (Y123) and cobalt base electrolyte. Nevertheless, a very good effect can be observed by co-sensitization with two metal-free organic sensitizers. Kakiage et al. [21] demonstrated sensitized DSSC with ADEKA-1 and a carboxy-anchor organic dye of LEG4 collaboratively which show an excellent record of efficiency, up to 14.30%.

### 3. Conclusion

Dye-sensitized solar cells are a generation of photovoltaic devices which is intense explored and thus developed over the three decades. However, still many components need to be improved in order to enhance working parameters ( $V_{OC}$ ,  $I_{SC}$ ,  $J_{SC}$ ,  $FF$ ) and efficiency of DSSC assemblies. Dyes presented in this review article are only a small part of a broad group of photosensitizers. Molecular structure of dyes can be easily modified which resulted in nearly infinite types of

photosensitizers. Nevertheless, there is a strong need for high-performance dyes which increase light adsorption by the semiconductor material. The very high efficiencies are obtained by ruthenium-based dye-sensitized solar cells by now (up to 11%), however, they should be replaced with metal-free dyes with the nontoxic influence of environment. Natural dyes extracted from plants seem to be a promising alternative as well as organic dyes with efficiencies reaching 9%. However, it should be clarified that not only the photosensitizer but also other components and their mutual adjustment is of high importance. Wherefore, a new approach to co-sensitization results in the value of efficiencies which can be compared with those for Ru sensitizers. The comprehensive discussion allows to suggest that utilization of natural and metal-free dyes in DSSC result in high performances in the foreseeable future.

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