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REVIEW ARTICLE

Dye sensitized solar cells based on natural dye- A review

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Abstract

The third generation of solar cell is known as dye-sensitized solar cells (DSSCs), which is convert the visible light to electricity by using the photo-electrochemical reaction. This type has a sandwich structure that consists of a semiconductor formed between a photo-sensitized anode and an electrolyte and it's based on the width of the semiconductors. However, the natural dyes which are usually included the organic molecules have been considered to be more important due to cost-effective, water-soluble, availability, and degradable, such as fruit, leaves, and flowers. In this article, we will try to cover the most important information about the DSSCs which are usually based on the using of the natural dyes and will discuss and reviewing the most important methods that can be used for extraction of different types of dyes from natural.

1. Introduction

Basically, the principle of solar cell work is founded on the conversion of solar energy to useful energy, and that through directly convert of the sunlight to electricity. Dye-sensitized solar cells called Gratzel cells were first reported in 1991 [1, 2]. Since that time this subject has attracted considerable attention by many scientists to investigate and explore the principle of its working and find a new methods or tools to increase their conversion efficiency. However, DSSCs have so many advantages, for instance, it's consider as one of the energy source that can be generated with low cost, ease, effectiveness, flexibility, and transparency. The natural dyes are accessible, easily extracted, and environmentally friendly [3-8]. Generally, these types of dyes can be taking out by using different methods of extraction from the petals of flower, leaves, roots, etc. in the form of "anthocyanin, chlorophyll, flavonoids, and carotenoid pigments". Some compounds are used for dying textiles causing water pollution and no degradable and carcinogenic, instead of that and to avoid this problem, it can be used the natural dyes. [9]. However, dyes are classified into different forms as organic dye (natural dye), and organometallic dye.

On one hand, the organometallic dyes such as Ruthenium dyes are important for the manufacturing purpose of DSSCs with a good outcome of efficiency [10, 11].On the other hand, they are expensive and difficult to purification compared with natural dyes[12, 13]. Conversion of sunlight to the electricity is depended on the photo-sensitization and is formed via the absorption of a part of the sunlight by dyes then converts it into electrical energy [14, 15].

The DSSCs act as efficient photovoltaic cells made by using alternative nanostructured films of the metal oxide, such as the ZnO, SnO_2, SrTiO3, Ta_2O_5, WO_3 , etc. [11]. The DSSCs are consisted of four different parts which are included: photo-anode, a counter electrode (CE), electrolyte solution, and dye sensitizer. The working of DSSC involves many steps started from the electrode is set, firstly will need to deposit a thin layer of the oxide which should be a semiconductor, for instance, TiO2, ZnO, SnO_2 on the specific type of glass plate

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(transparent glass), which is usually made of (fluorine or indium) tin oxide, (FTO) (ITO), respectively. TiO_2 which is mostly used since it's consider a non-toxic, less expensive also due to the availability. After that, the film will immerse in the solution of the dye, which will lead to adsorb the dye onto the surface of the metal oxide under investigation such as the TiO_2 as in this case. However, surface area of the electrode increases via the highly porous structure and with increasing of dye molecules that would attach to the nano-crystalline TiO2 surface as a result, the sunlight that absorption on the surface of the semiconductor materials will increase. Dye "Photosensitizer" must be luminescent and cover the regions of both UV-vis and near-IR. Electrolyte solution such as I^-/I^-3 (redox couple), has an impact of the creation of an effective contact between the both electrodes (WE and CE) inside the cricket. And the last one (counter electrode) is usually prepared from using the carbon or platinum. After absorbed the incident light by the photosensitizer, this will lead to excited the electrons from the "ground state" to the "excited state" of the dye, after that, electrons will be injected to the surface of TiO_2. These injected electrons are transported from working to counter electrode, then Through the oxidation –reduction of the electrolyte solution I^- 3/ I^-, the electrons are go back again to the dye external circuit, [16, 17], as seen in Fig 1.

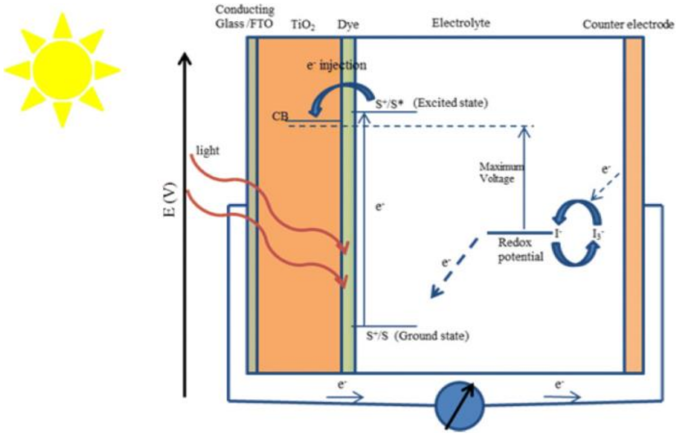


Figure 1: Construction of dye-sensitized nano-crystalline solar cell [16].

1.1 Preparing and applying natural dye on the photo-electrode

In order to prepare the naturel dyes, in this case as mention before, will need to we to extract them from different natural sources such as from fruits, petals, leaves, and flowers. Then these collected samples should be washed and to remove the water molecules should be dried by hot air, and approximately of (20 to 50) gm of each sample will be chopped into small pieces and crushed. In order to collect the dye solution, the sample precipitate need to filtration. Then, the extracted dye should be stored in a clean and dark place. Later, the working electrode (cathode) which is labeled as (WE) will be immersed in the solution of dye for about six hours. All of these steps were happening at 25°C[18].

1.2 Preparing of photoelectrode

The TiO_2 paste was prepared by weight 1g of TiO_2 powder and mixed with drops of ethanol using a clean pestle. This paste was deposited on the conductive glass of FTO as a layer and dries for about a one hour at 450°C, then and at room temperature allowed for that slide to be cooled slowly. Then it immersed in the dye solutions for about one hour. The dye will adsorb on the TiO_2 surface and will bonded with Ti sites may be through the covalent bonds, while the residue dye (unabsorbed) was removed by rinsing with ethanol[19, 20].

1.3 Counter Electrode Preparation

In order to prepare the counter electrode which is labeled as (CE), in this case will use the conductive glass of FTO. Here, should be put or placed at the top of a burner or candle the conductive side of the glass, whereas, the light carbon layer should be place at the entire conductive side of the glass. However, the thin layer of carbon will act as a catalyst for the I^-/ I^-3.

1.4 Electrolyte Preparation

In order to prepare the electrolyte solution will need to mix (1.6g, 0.10M) of potassium iodide KI with (2.5g, 0.10M) of iodine I2 in a 1:1 ratio with (50 ml) of water and stirring until complete the dissolution. Then, the mixture was diluted with distilled water, giving 0.10 M I^-3 solution.

1.5 DSSC collecting

Both of photo and counter electrodes, which are the cathode and anode electrodes, respectively, were put together as (overlapping), and at the end of each different electrode (photo and counter) electrodes should leaving a space in order to added the electrolyte solution. However, these two electrodes are linked with used the double clip. In order to, close the secret between the two electrodes for the active layer of the (SCs) have been added a Few drops of iodine.

1.6 Evolution of dyes for DSCs

The preparations of four different types of triphenylamine have been reported by Liang et al., [21], and have been found that the design triphenylamine can help the progress of the DSSCs performance. Wang et al.,[22] synthesized two new tert-butyl benzene and dipropylfluorene dyes for DSSCs, to achieve the effect of the bridge and donor structure on the properties of photovoltaic cell. On the other hand, the Li et al., [23]were synthesized, cyclometalated ruthenium sensitizers coordinated with thienyl-pyridine ligands for DSSCs applications . L. Zhu et al.,[24] were synthesized new dyes of triphenylamine and dicyanovinylene, in this case, the ter-thiophene occurring as a bridge between the carboxylic acid group and the triphenylamine part will increase the power efficiency of solar cell . W. Lee et al.,[25] was designed and synthesized four new organic sensitizers, to examine the effect of cyanoacrylic acid as additional anchoring group also the effect of the N-substituent on the affiance of DSSCs.

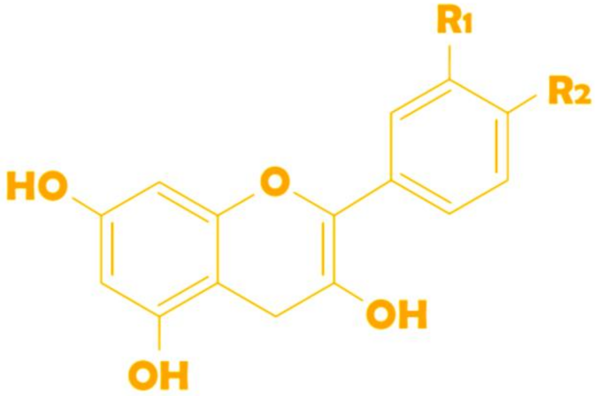
1.7 Extraction of different natural dyes

Majorca C. et al.,[26] extracted the anthocyanin pigments as a natural dye from Malabar spinach fruits using three solvents: pure methanol solvent, or 50% methanol solvent, or 50% methanol + 1% HCl, these solvents were added to the crashed samples into a beaker avoid sunlight exposure the dye using aluminum foil, then the solvent extracts the pigments after 10 minutes. The extraction via pure methanol solvent has a good effective extracting. Abdul Hai Alami et al.,[27] was extracted a dye from the leaves of C. Gigantic, cut leaves, and mixed with 45 ml of ethanol. Then, add chenodeoxycholic acid was added; the beaker was cover with para-film and put at an ultrasonic bath for for about 30 min. And to to prevent oxidation the dye was stored in the dark palce. D. Sinha et al.,[28] are used the “chlorophyll” as a sensitizer for the production of DSSCs. Chlorophyll dye is extracted from “Spinaciaoleracea”. Spinach is cut and mashed, and then the sample after crashed it have been placed in an ultrasonic cleaner for about 15 min at 30°C and the coloring samples have been separated by using the centrifuge. While, Rajan, Aneesiya K., and Cindrella, L.[29] have collected the leaves of W. tinctoria, washed and dried. Here, there are two different extraction techniques have been employed, first method is called cold methanolic, when stirred for 2 hours about 0.5gm of the leaves powdered with 50 ml of methanol, then left that solution in the dark place during the first day. After that have been filtered the solution and collected the dye. In the second method which is called soxhlet extraction, 2 gm of powder are mixed with 100 ml of methanol and leave it for 5 hours at 65 °C. The remaining solution was collected, then centrifuged, and filtered. M. Nirmala et al.,[30]was prepared the blueberry dye through taken ten blueberries and washed with distilled water. Grinded and mixed it with ethanol. Put the mixture in a tube and separated it by a centrifuge. Filter the solid particles from the bottom of the tube and take the blueberry liquid dye. M. Manuel et al., [31] the Lees derive from the winemaking process of both red and white wines using soxhlet extraction technique at room temperature by different solvents such as ethanol, methanol, water, and mixtures of them, HCl 37% was added to acidify and maintain the pH less than 2. The dye solution was stored in a dark place at 5 ◦C after the extraction process. Taco-Ugsha et al., [32] extract the natural dyes from mortiño (Vaccinium floribundum) by maceration in a methanol acidified with HCl (0.01%), citric acid (2.5%) and trifluoroacetic acid (3%). Ratio 1:4 of Mortiño/methanol, and the mixture was filtered, the solvent was removed under reduced pressure [36] A. O. Boyo et al.,[33] take 100 grams of the powder sample (Ipomoea involucrata leaves and flowers) and submerged in 120 ml of the solvent system (distilled water, methanol, and HNO_3 in ratio 10: 9: 1) and then covered for 24 hours. Extraction occurred after 24 hours through the decantation and evaporation to obtain concentrated extract, then stored in dark beaker at room temperature[33] . D. B. Patil et al.,[34] extraction of dyes were carried out by four methods, aqueous method: 10 gram of the petals of red rose were boiled in 100 ml distilled water for 30 min. The decolorized petals were removed from the extraction solvent. Alkaline methods: 10 gram of petals were boiled in 1 % NaOH for 30 min.. Acidic methods: the petals (10 g) were treated with 1 % of acidic solution boil at 100° C for a few min. Alcoholic Extraction methods: 10 gram of the petals were boiled in 50 % alcohol for 30 min. the filtrate of all the above extraction methods was used for other studies. S.M.Amir-Al-Zumahi et al.,[35] cleaned the samples (petals, leaves) with distilled water and dried at room temperature and cut into small size (≈0.5cm). The extraction method of natural dyes using magnetic stirring and ultrasonic device, 1gram of samples put in a beaker with 50ml of solvent (water/ethanol) at 45°C for 60 min. In the second process used ultrasonic extraction process, 5g samples were treated with 60ml of ethanol. Then dyes were filtered and collected. The purification was done by column chromatographyو separate from solvent by vacuum evaporation process and finally found the raw dyes, the percentage yield and % improvement due to ultrasound process from the following equation (1).

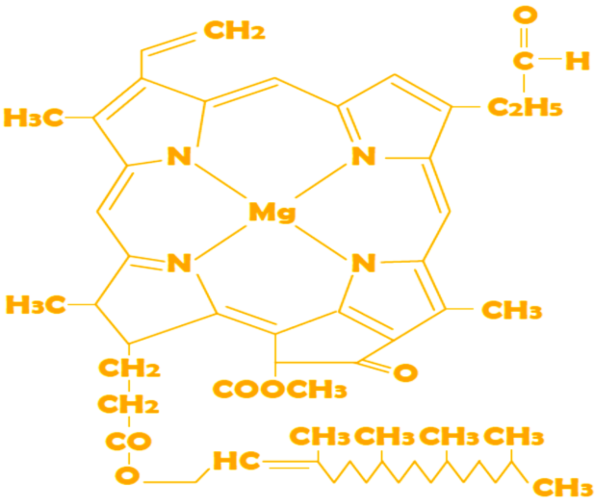
$$\%\text{ of yields} = \frac{\text{obtained Natural Dyes(g)}}{\text{Raw Material of Natural Dyes(g)}} \text{ x } 100\%$$

$$\begin{aligned}&\%\text{ improvement due to Ultrasound}\\&= \frac{\%\text{ yield of (Ultrasound} - \text{Magnetic stirring)}}{\%\text{ yield of Magnetic stirring}} \text{ x } 100\%\end{aligned}$$

Natural dyes are classified into three types: plants, animals, and minerals. Plant dyes such as nuts, roots, and flowers are sources of coloring pigments stem, also to extract the natural color it can be from leave, fruits, and seeds. While the animal dyes such as Levantine sea snails and the mollusks may have the royal purple pigment. Ahmed M. Ammar et al.,[36] 6gm of onion pee was mixed with 250ml distilled water, and 147gram of pieces red cabbage was covered with 400ml of distilled-water. Dispersion solutions have been heated for 24 hrs at 90°C. Then cool to room temperature, and filtered to extract the anthocyanin dye. Another dye was extracted from spinach via using the acetone as a solvent. Here about 11gm of spinach have been crushed to have a fine powder then mixed with acetone. After that have been filtered the solution, to extract the dye. All dyes were placed in a dark room, Fig 2(a-b) shown examples of some natural dye.



(a)



(b)

Figure 2: (a-b) are the molecular structures of anthocyanin and chlorophyll dye, respectively.

1.8 Applications

One of the useful applications of natural dyes can be used as a sensitization in solar cells, in photo-electrode, TiO_2 act as semiconductors and the dye enhance the efficiency of this electrode through electron injection and slow reverse reactions to convert sunlight to electricity. Another application of dyes is used for the coloration of hair, skin, and textile materials such as henna, carrot, Plumeria hibiscus, Delonix, and combretum[37].

1.9 Comparison between the natural dye and synthetic dye solar cells

The comparison is based on many different parameters such as the cost, stability, absorbance, environment, the resources and the efficiency of the cell, as well as, the manufacturing method that have been used. The natural sensitizers are usually prepared by one the extraction methods from the roots or leaves, and flowers, via using the ethanol, water, or methanol as a solvent thus less costly comparing with synthetic dyes. [38, 39], see Table 1.

1.10 Efficiency of DSSCs:

To further enhance the energy conversion efficiency of the dye-sensitized solar cell should focus on the following points:

- To decrease the charge recombination between the oxidized sensitizer and the injected electrons into TiO_2, this can be by preventing the coordination that occurred between the sensitizer and iodide. Also the electron donor should be separated from the TiO_2.
- To increase the efficiency of electron injection this can be by using a good overlap of LUMO with the TiO_2 surface.

Table 1: *General dissimilarities between sensitizer synthetic and sensitizer natural dye.*

Case	Parameter	Sensitizer synthetic dye	Sensitizer natural dye
1	Cost	The preparation of complex material done by different chemical reaction So its high cost	The extraction process from plants, flowers, fruits, roots. So its low cost
2	Environment	Bad effects because its chemical nature	Fewer effects on the environment because of its natural occurrence
3	Stability	Slowly degradation through exposure sunlight, therefore long life of the cell	Degradation of natural dye in the presence of sunlight
4	Absorbance	N3 dye has an absorption band of over 800 nm	The dyes have absorption bands in the visible range (400–700 nm).
5	The efficiency of the cell	Higher efficiency	Low efficiency because of degradation of natural dye
6	The manufacturing method of the cells	Using multi procedures, plus a variety of solvents, take a long time for purification methods so that the dye production is very expensive	Using simple and direct chemical procedures, so that the natural dye production is less expensive
7	Resources	A noble material, so that less availability	Natural availability100%

1.11 Absorption spectra of extracting natural dyes

The absorption spectra of extracting dye solutions from Cytisus, Alcea rosea, and Roselle and dye materials covered the TiO_2 films in the range of (400 – 700) nm are shown in with maximum wavelength absorption (360, 540, 520) nm for from the Cytisus, Alcea rosea, and Roselle, respectively. Absorption intensity of extracting dye solutions was measured at 400 nm, because of charge transfer transitions from the HOMO to the LUMO. Therefore, the colors of the extracted dye were red and bluish red [40]. The absorption of the extracted dye adsorbed on the films of TiO_2 was shifted to a longer wavelength if compared with the absorption spectra for the dye solutions. This is due to the occurring of hydrogen bonding that formed between hydroxyl group of TiO_2 and the hydroxyl or methoxyl groups of colorant molecules [41].

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