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Characterization of Eight Natural Dyes as Synthesizer for Dye-Sensitized Solar Cells Technology

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ABSTRACT

In the present work, dye-sensitized solar cells (DSSCs) were fabricated by using natural dye extracted from eight different flowers namely: [(MN1) Turkish Hibiscus sabdariffa, (MN2) Iraqi Hibiscus sabdariffa, (MN3) Rosehips, (MN4) Lavandula, (MN5) Nerium oleander, (MN6) Red Bougainvillea, (MN7) Pomegranate flower, and (MN8) Pink Bougainvillea] extracts as a photosensitizer. The UV-Vis absorption spectra for all eight samples were recorded at room temperature by using a mixture of 0.1 mol.L $^{-1}$ HCl and ethanol, and also in different solvents. In addition, the effect of the pH value of these dyes on absorbance were investigated. Moreover, the Fourier transform infrared (FT-IR) spectra of all dyes were recorded. The lamp power used to evaluate the DSSCs performance is 1000 W/m². The power conversion efficiency (η) of extracted dyes was reached as follows: (MN1) =1.589 %, (MN2) =1.229 %, (MN3) =0.602 %, (MN4) = 0.927 %, (MN5) =0.684 %, (MN6) =1.317 %, (MN7) =1.059 %, and (MN8) =1.149 %.

GRAPHICAL ABSTRACT

Code	Fresh flowers	Dried flowers	Powder	Dyes
MN1		*****	-	
MN2				
MN3				Austra
MN4		也是是		
MN5		11.223		
MN6		1		as assessment
MN7	TO NE	453		
MN8		200		

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Introduction

Dye-sensitized solar cells (DSSCs) are a type of the third generation of solar energy that are discovered by Professor Michael Grätzel in 1991, also known as Grätzel cells. It works to convert the photons coming from the sun into the electrical energy [1]. A DSSCs device is composed of dye molecules, a nano-crystalline porous semiconductor layer (TiO₂), an electrolyte, and a counter electrode. The dye acts as a sensitizer, it absorbs the photons from the sun, and therefore the electrical energy is produced. Most compounds that have been applied as sensitizers in (DSSCs) are ruthenium complexes because of their strong charge-transfer absorption in the completely visible ranges and are very effective the transfer (metal-to-ligand) However, the manufacturing of these compounds is complex, expensive, and they have contained the heavy metals, which are not suitable for the environment [2]. After that, the organic dyes and (ruthenium-free complex metals) have been developed by researchers. The organic dyes have indicated the good conversion efficiencies when used as sensitizers in (DSSCs), but the synthesis of these dyes is difficult and time-consuming [3, 4]. Likewise, in recent years, the natural dyes as (flavonoid, betacyanin, anthocyanin, chlorophyll, tannin, and carotenes) extracted from flowers, leaves, and fruits have gained increased interest for use as sensitizers for (DSSCs) an alternative to synthesis dyes because their materials are available, cheap, non-toxic, environmentally friendly, and do not require the complex techniques for dye extraction [5, 6]. Most green plants' are rich in chlorophyll dye and this dye has been studied in several research as a potential (DSSCs) sensitizer including in the year 2013 by Chang et al. employed chlorophyll dye from wormwood and anthocyanin dye from red cabbage as sensitizer in (DSSC) and the conversion efficiency were reported of 0.9% and 1.47%, respectively [7]. Anthocyanin pigments are natural dyes that are giving color to the plants and fruits. In general, these pigments are

responsible for the blue and red-purple color of plants and they have been widely used in (DSSCs) [8, 9]. For an efficient dye, there are three main characteristics are required, (i) the dye should have a broad absorption spectrum in the visible region, (ii) the strong attachment to the semiconductor materials such as TiO₂, and (iii) the dye should has the capability to inject the electron into the semiconductor materials [10]. In this work, the performance of eight obtainable natural dyes that have been extracted from flowers from two countries Iraq and Turkey are reported. In addition, we present a study of the effect of different pH on the colors and the absorbance of these dyes.

Materials and Methods

Collecting dyes

Eight types of flowers were collected to extract their natural dyes and give a code to all these flowers. Hibiscus sabdariffa are collected from two regions (MN1) from Turkey and (MN2) from Kirkuk, Iraq. Rosehips (MN3), Lavandula (MN4), and Pomegranate flower (MN7) are collected from Turkey. Nerium oleander (MN5), Red (MN6), and Pink Bougainvillea (MN8) are obtained from Kirkuk City, Iraq.

Extraction

After collecting flowers, the colorful petals for flowers are separated and wash it with distilled water, and then they are left to dry at (25 °C) room temperature for 5 days. After drying and crushing into a fine powder by using a mixer, (5 g) of the samples was measured by using a sensitive balance and immersed in (50 mL) of the appropriate solvent, as listed in Table 1, and then aluminum foil was used to protect the prepared solutions from exposure to light and they were left in darkness for 3 days at room temperature, after filtration, these solutions from solid residues to obtain clear dye solutions, that are evaporated under low pressure. Table 2 presents the flowers used in this study.

Table 1: The solvents used to extract dyes from flowers

Codes	Solvents
MN1	Methanol
MN2	Methanol
MN3	Ethanol
MN4	Distilled water
MN5	Ethanol+ (0.1 M) HCl
MN6	Distilled water + (0.1 M) HCl
MN7	Distilled water
MN8	Distilled water + (0.1 M) HCl

Table 2: Flowers used in this study; (MN1) Turkish Hibiscus sabdariffa, (MN2) Iraqi Hibiscus sabdariffa, (MN3) Rosehips, (MN4) Lavandula, (MN5) Nerium oleander, (MN6) Red Bougainvillea, (MN7) Pomegranate flower, and (MN8) Pink Bougainvillea

Code	Fresh flowers	Dried flowers	Powder	Dyes
MN1				
MN2				No transport
MN3				Resolution
MN4			Carl.	9
MN5				
MN6				Ad Grangasian Pe
MN7				and open the state of the state
MN8		数		nd designation

Fabrication of natural DSSCs

A detergent solution was used to clean the ITO conductive glass, and then rinsed with distilled water. The semiconductive layer (TiO_2) was prepared by adding (20 ml) of ethanol to the (3.5 gm) of TiO_2 nanopowder. The solution was stirred for 40 min by using a small magnetic bar to form a TiO_2 paste. Doctor blade techniques were used to deposit the prepared paste to the top of the ITO glass sheet. The glass sheet with

the TiO_2 layer was heated at 80 °C for 40 min, and then sintered at 350 °C for 1 hour. After cooling, the deposit TiO_2 was immersed in the dye solution for 5 hours. The counter electrode was made by sketching a pencil (graphite pencil) on the surface of another conductive glass (ITO). The counter electrode and TiO_2 thin layer were assembled with dye to form a DSSC by sandwiching with a redox electrolyte solution (I–/I3).

Results and Discussion

The UV-Vis absorption spectra analysis

The absorption spectra of eight dye solutions were recorded by using a UV–Vis spectrophotometer T92 in the spectral range from 350 nm to 800 nm by using 0.05 g of the prepared dyes and 10 ml of solvents. There are two techniques that have been used as follow to measure the absorbance of dyes.

In different solvents

Many solvents with different polarities were investigated to choose the common solvent to dissolve all the eight samples, a mixture of ethanol with (0.1 M) HCl (at room temperature) were detected as the best solvent to study the optical properties of these dyes. As displayed in Figure 1, eight samples possess different

absorption bands (narrow and broad) in the UV-Vis region, with the maximum absorbance between 447 nm to 542 nm.

The absorption spectra of MN1, MN2, MN4, MN5, and MN7 consist of one intense absorption narrow peak in the visible region, with the maximum absorption λ_{max} at 542 nm, 546 nm, 539 nm, 539 nm, and 516 nm, respectively, are listed in Table 3. Furthermore, the MN8 absorption spectra consist of one broad peak in the visible region with λ_{max} at 537 nm. Moreover, the MN6 absorption spectra show a broad absorption peak (m) with an absorption maximum at 499 nm and gave shoulder at 526 nm. In addition, the MN3 dye absorption spectrum exhibits the weakest absorption peak with the maximum absorption λ_{max} at 447 and gave two shoulders at (420 and 470) nm.

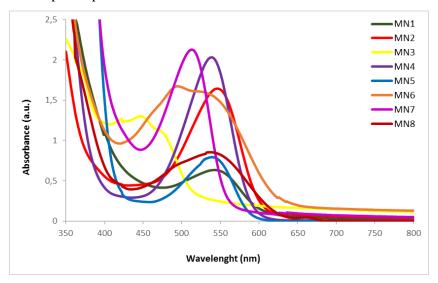


Figure 1: The UV-Vis absorption spectra of extracted dyes in a mixture of 0.1 mol.L⁻¹ HCl and Ethanol

Table 3: The characterization of the extracted dyes in a mixture of 0.1 mol.L⁻¹ HCl and ethanol

Dyes	Absorbance (a.u.)	λ_{max} (nm)
MN1	0.632	542
MN2	1.645	546
MN3	1.298	447
MN4	2.033	539
MN5	0.788	539
MN6	1.672	499
MN7	2.112	516
MN8	0.857	537

The main component of MN1, MN2, MN4, MN5, and MN7 dyes is anthocyanin [11-14]. The differences in the absorption properties between

these dyes are caused by the different types of functional groups on the anthocyanin pigments and the extracts' colors [8]. The most attractive

characteristic of anthocyanin dye (Scheme 1) is the wide absorption peak spectrum in the visible region. Furthermore, anthocyanin pigments have carbonyl and hydroxyl substituents as functional groups that can be attached easily to the surface of titanium dioxide substrate. This bonding is significant to transfer the excited electron of dye molecules to the semiconductor materials such as TiO₂ [15, 16]. The absorption peak of MN3 dye is due to dihydroflavonols [17]. In addition, the absorption spectra of MN6 and MN8 dyes are due to betalains dyes, the first peak of MN6 at 486 nm is due to indicaxanthin and the second peak at 536 nm is due to betanin, as is the peak of MN8 at 540 [18, 19].

Scheme 1: Chemical structure of anthocyanin

In different pH

Five different pH values were studied for these extracted dyes ranging from pH=2, pH=4, pH=6, pH=7, pH=8, and pH=10 by using (0.1 mol/L HCl) as acid and NaOH as a base to change the pH value.

Figure 2 shows the effect of different pH of the dye solutions by changing the pH values by using hydrochloric acid and sodium hydroxide, where the highest wavelength (λ_{max} nm) was recorded for MN1(582 nm), MN2 (591 nm), MN3 (508 nm), MN4 (617 nm), and MN7 (580 nm) when the pH =10, are listed in Table 4; however, it was recorded at pH =6for MN5 (534 nm), MN6 (536 nm), and MN8 (540 nm). Anthocyanin pigments are present in MN1, MN2, MN4, MN5, and MN7 that are red color under pH<7 and yellow-blue under PH>7 [20], with increased the pH value, the wavelength of dyes was increased. We have noted the redshift or bathochromic shifts appear when the pH value increasing of MN1, MN2, MN4, and MN7 dyes, but MN5 dye is unstable in alkaline conditions because of the anthocyanins present in this dye in the quinoidal form [21]. Likewise, MN3 dye is showing redshift at an increased pH. MN6 and MN8 dyes contain betacyanin pigments, these pigments were not stable in pH> 7.5 [22].

Photoelectrochemical measurements

The characterization of DSSC performance for the eight natural dyes was tested by using the current density-voltage (J-V) curves with a light source of $1000~(W/m^2)$ illumination lamps. The short circuit current (Isc), open-circuit voltage (Voc), fill factor (FF), and energy conversion efficiency (η) were calculated.

Figure 3 and Table 5 illustrate the photoelectrochemical performances of (DSSCs) fabricated from eight natural dyes samples based on three different dye pigments, as anthocyanin, betacyanin, and dihydroflavonols.

As presented in Table 5, the short circuit current density (Isc) assorted from 25.2546 mA/cm² to 10.5786 mA/cm². The highest short-circuit current density was obtained for the DSSCs sensitized with Turkish Hibiscus sabdariffa dye MN1. However, the lowest short-circuit current density was recorded for the (DSSC) sensitized with Rosehips dye (MN3). Likewise, the opencircuit voltage (Voc) exhibited a maximum value of 0.8237v with the MN1 and a minimum value of 0.7458v with the MN3. Moreover, another important parameter as the fill factor (FF) was investigated. The FF varied from 77.0571 for the (DSSC) sensitized with Lavandula (MN4) to 76.1827 for the DSSC sensitized with Nerium oleander (MN5).

The highest achieved energy conversion efficiency (η) was 1.5896% from the anthocyanin pigments extracted from Turkish Hibiscus sabdariffa dye MN1. Moreover, the anthocyanin pigments represented by MN2 and MN7 indicated the acceptable power conversion efficiency of 1.2297 and 1.0597, respectively.

This is because of the better bonding of the alcoholic groups represented in the anthocyanin extract with the titanium dioxide nanoparticle TiO_2 [23]. In addition, the betacyanin pigment represented by MN6 and MN8 also exhibited a good energy conversion efficiency of 1.3175, and 1.1498, respectively. The energy conversion

efficiencies obtained in this study are very acceptable. More importantly, the highest recorded data of energy conversion efficiency for

(DSSCs) based on natural dyes related to the betalain pigments extracted from purple wild sicilian prickly pear dye of 2.06 [24].

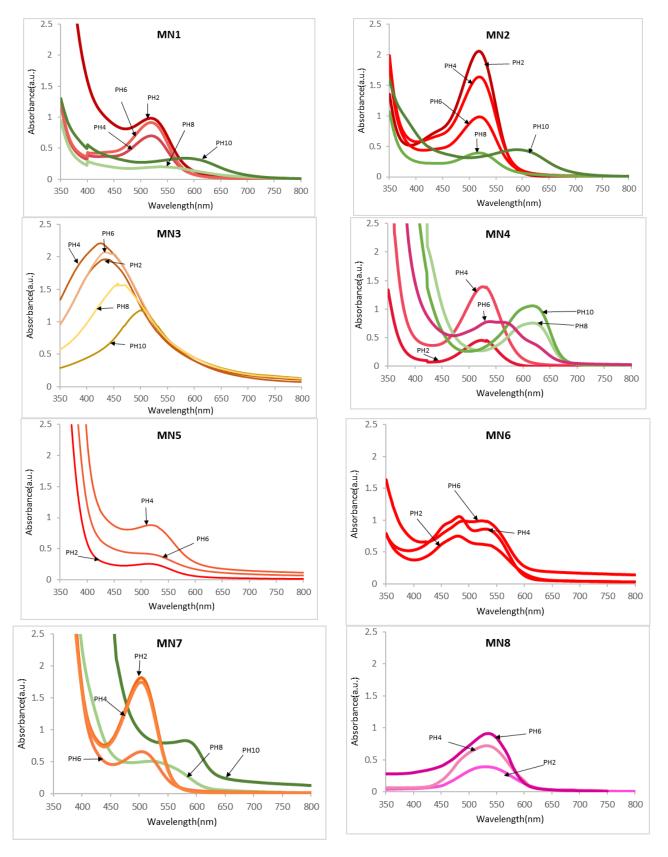


Figure 2: The UV-Vis absorption spectrum of dye solution at different pH

Table 4. The maximum v	vavelenght λ _{may} (nm) of natural	dves extracted from	flowers at different pH values

	MN8	MN7	MN6	MN5	MN4	MN3	MN2	MN1
pH=2	518	518	437	523	514	480	502	532
pH=4	519	519	427	525	516	490	503	532
pH=6	519	518	442	554	534	536	503	540
pH=8	536	529	471	617	-	-	519	-
pH=10	582	591	508	617	-	-	580	-

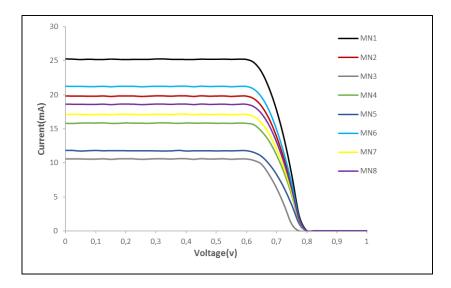


Figure 3: The J-V curve of fabricated DSSCs for all the eight extracted dyes

Table 5: Photovoltaic parameters of the DSSCs are sensitized by eight types of natural dyes extracted from flowers

Code	Isc(mA)	Voc(v)	FF%	η %
MN1	25.2546	0.8237	76.4176	1.5896
MN2	19.8296	0.8106	76.5072	1.2297
MN3	10.5786	0.7458	76.3732	0.6025
MN4	15.7986	0.7619	77.0571	0.9275
MN5	11.8397	0.7591	76.1827	0.6849
MN6	21.2341	0.8109	76.5173	1.3175
MN7	17.1027	0.8102	76.4827	1.0597
MN8	18.5955	0.8083	76.497	1.1498

Conclusion

Eight natural dyes extracted from different types of flowers based on three different dye pigments; anthocyanin, betacyanin, and dihydroflavonols were used as photosensitizers for DSSCs and fabricated with TiO_2 as a semiconductor layer. The highest conversion efficiency was 1.5896% for the DSSC sensitized with Turkish Hibiscus sabdariffa dye (MN1). However, the Rosehips dye (MN3) exhibited the lowest conversion efficiency by 0.6025%.

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Authors' contributions

All authors contributed to data analysis, drafting, and revising of the paper and agreed to be responsible for all the aspects of this work.

Conflict of Interest

The author declared that they have no conflict of interest.

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