

Parametric Studies of Natural Dyes in Dye Sensitized Solar Cells

Rupesh Shelke^{1,*}, Pramod Walke², Vivek Khond³, Nilesh Nirwan⁴

Abstract

Natural dyes used in dye sensitized solar cells (DSSCs) are low cost viable substitute to high cost ruthenium dyes because of its easy availability and no environment threat. Parametric studies of natural dyes in dye-sensitized solar cells (DSSCs) are essential for optimizing the performance of these renewable energy devices. One kind of thin-film solar cell called a DSSC uses organic dyes to absorb sunlight and turn it into electrical energy. Natural dyes, derived from plant sources or other organic materials, have gained attention as potential replacements for synthetic dyes due to their environmental benefits and lower cost. This paper discusses the appearance and feasibility of natural dyes in DSSCs. Cobalt sulphate and graphite are the two counter electrodes and natural dyes were used in the DSSCs. The sensitizing dye is one of the crucial components that is essential to the device's functionality. Natural colors made from plants are becoming more and more popular due to their abundance, low toxicity, and environmental friendliness. These natural dyes are widely accessible and at reasonable price anywhere in the world. Due to their modest energy conversion efficiency and low cost compared to ruthenium dyes, natural dyes have been investigated for usage in DSSCs. Because of its affordability, ease of manufacturing, and promise for sustainability, DSSCs made from natural dyes have become a viable substitute for traditional silicon-based photovoltaic systems in future.

Keywords: Natural dyes, dye, sensitized solar cells, cobalt sulphate, graphite

INTRODUCTION

Dye sensitized solar cells (DSSC) have attracted much attention as low cost photovoltaic cells and have become a rapidly emerging area with prospective applications. In DSSCs, the dye as a sensitizer plays a crucial role for converting absorbing sunlight into electric energy. The bond between sensitizer (dye) and TiO₂ film is very important in enhancing the power output of DSSC. Till date, the use of high film TiO₂ electrodes with ruthenium dye and platinum as counter electrode leads to the maximum efficiency of 11.1% achieved by Yasuo Chiba et al [1]. Nowadays ruthenium based sensitizers have been commonly used due to their better efficiency and high stability. Although DSSCs using ruthenium dye gives comparatively high efficiency, there are some drawbacks of using it. Ruthenium is considered as noble metals and there is scarcity of noble metal in world since they are not available in adequate quantity. Furthermore, producing such metal comes at a very higher cost. Hence, it remains a technological challenge to explore low cost, easily accessible dyes as efficient sensitizers for DSSC. In view of this, there is always a need for alternative photosensitizers mainly due to the high cost of ruthenium metal and the unavailability of these noble metals in future. The use of natural dyes as a sensitizer to produce a power in DSSCs is appealing because of economical aspect and environmental point of view. Since the natural dyes are easily available in flowers, leaves and fruits, they can be easily used in DSSCs for converting solar energy

*Author for Correspondence

Rupesh Shelke

^{1,2}Professor, Department of Mechanical Engineering, G H Raison College of Engineering, Nagpur, Maharashtra, India

^{3,4}Assistant Professor, Department of Mechanical Engineering, G H Raison College of Engineering, Nagpur, Maharashtra, India

Received Date: November 12, 2024

Accepted Date: December 13, 2024

Published Date: April 08, 2025

Citation: Rupesh Shelke, Pramod Walke, Vivek Khond, Nilesh Nirwan. Parametric studies of natural dyes in dye sensitized solar cells. Journal of Polymer & Composites. 2025; 13(Special Issue 3): S121–S132p.

into electrical energy. Natural dyes have been the focus of DSSC technology research because of their low cost and non-toxicity. Numerous efforts have been done by several research groups all over the world to utilize these natural dyes as sensitizers in these devices. Using red sicilian orange juice dye as a sensitizer, Calogero et al. [2] created DSSCs in 2012 with a conversion efficiency of 0.66%. According to Khwanchit et al. [3], DSSC employing natural dyes derived from blue pea, rosella and a combination of extracts yields 0.37%, 0.05% and 0.15% efficiency respectively.

Using rose bengal dye as a sensitizer, Roy et al. [4] discovered that DSSC yields J_{sc} and V_{oc} of 3.22 mA/cm² and 0.89 V respectively leading to a 2.09% conversion efficiency. Fruit dyes such as mulberry and others were investigated for DSSC by Garcia et al. [5]. Using natural dyes taken from mulberry, cabbage-palm, and chaste tree fruit as sensitizers, it was discovered that the I_{sc} and V_{oc} of DSSC are 1.06, 0.86 and 0.37 mA and 390, 422, and 442 mV, respectively. On dye sensitization of the aforementioned dyes, fill factor values of 0.40 to 0.61 have also been obtained. When Cherepy et al. [6] used flavonoid anthocyanin dyes that were derived from blackberries, they reported a conversion efficiency of 0.56% under full sun. For such a simple system, the V_{oc} of 0.4 to 0.5 V and the I_{sc} of 1.5 to 2.2 mA/cm² are noteworthy. Hao S. et al. [7] reported that they achieved I_{sc} between 1.142 and 0.225 mA, V_{oc} between 0.551 and 0.412 V, fill factor (FF) between 0.52 and 0.63, and P_{max} between 58 and 327 μ W. Because of the presence of anthocyanin molecule on black rice extract and the better TiO₂ film interaction, black rice extract had the best photosensitized impact among all of these natural dyes. It has been studied that the performance of extract from tropical flowers as possible sensitizers for DSSC. Photocurrent densities, photo voltages, conversion efficiency and fill factor was obtained in the range of 1.1 to 5.4 mA/cm², 390 to 410 mV, 0.2 to 1.1 % and 0.53 to 0.64 respectively. Table 1 shows a summarized review of performance of DSSCs made from natural dyes [8].

Table 1. Performance of DSSCs made from natural dyes

Type of natural dyes	V_{oc} (V)	FF	η (%)
Festuca ovina	0.548	0.699	0.46
Tagetes erecta	0.475	0.606	0.8
Rosella	0.40	0.57	0.37
Blue pea	0.37	0.33	0.05
Mixed rosella-blue pea	0.38	0.47	0.15
Black rice	0.55	0.52	-
Bixin	0.57	0.59	0.37
Crocetin	0.43	0.46	0.56
Fruit of Calafate	0.47	0.36	-
Syrup of Calafate	0.38	0.20	-
Red Sicilian orange	0.34	0.50	-
Purple egg plant extract	0.35	0.40	-
Dragon fruit	0.22	0.30	0.22
Bougainvillea	0.30	0.57	0.36
Chlorophyll	0.43	0.39	0.59
Begonia	0.54	0.72	0.24
Red Bougainvillea glabra	0.26	0.74	0.45
Violet Bougainvillea glabra	0.23	0.71	0.31
Spinach	0.55	0.51	0.13
Ipomoea	0.55	0.51	0.13
Bougainvillea brasiliensis	0.25	0.36	0.45
Garcinia suubelliptica	0.32	0.33	0.69

In this study, DSSCs are fabricated using ten natural dyes extracted from different sources, namely: hibiscus, black grapes, spinach, orange, pomegranate, sugarcane, papaya, lime, pineapple and sweet lime. Using these natural dyes as sensitizers, the photo-electrochemical characteristics of the DSSCs were examined [9]. The aim of this study is to fabricate DSSCs and to examine whether it is possible to produce the power with natural extract based dyes instead of the commonly used high cost and toxic ruthenium dyes. DSSCs are fabricated using natural dyes with two counter electrode viz. graphite and cobalt sulphate and its comparative performances is studied and reported [10].

PREPARATION OF DYE SENSITIZED SOLAR CELLS

Materials

We bought transparent conductive oxide coated glass from M/s. Man Solar in the Netherlands that was 2 cm x 4 cm in size and had a resistance of 15-20 Ohm/cm². Potassium iodide (KI) was employed as the electrolyte in the current study. The juice from the cut and crushed flowers and fruits was used to make the dye solution [11]. In this study, dye solution was utilised directly as dye (sensitizer) for the manufacture of solar cells after being filtered to remove any solid residue. TiO₂ was employed as the photo electrode (anode), and cobalt sulphate and graphite powder were used as the counter electrode (cathode) [12].

Preparation of Electrode

Preparing an electrode typically involves coating or modifying a conductive substrate with a specific material or catalyst to facilitate the desired electrochemical reaction. A substrate for the production of dye-sensitive solar cells was tin oxide conductive glass that has been doped with fluorine [13]. The glass's functional area is 6 cm².

Cathode (Graphite and Cobalt Sulphate Electrode)

Initial electrodes are cleaned with water and allowed to air dry. A continuous layer of graphite is created on the conductive glass acting as a cathode by coating graphite over a conducting surface. When creating cobalt sulphate as a counter electrode, diluted acetic acid is combined with tiny, fine cobalt sulphate particles to create a semi-liquid paste [14]. On the glass's conducting side, the paste is spread. To create a continuous fixed coating on the conductive glass, it is sintered at a temperature of 75–80 °C for 20 minutes, and then it is cooled to room temperature in the furnace itself [15].

Anode (TiO₂ Electrode)

'Doctor-blade' technique is used to manufacture a photo electrode made of TiO₂. The resulting TiO₂ layer film has a thickness of 20 micrometres, while the individual TiO₂ particles measure roughly 30 nanometers in width. TiO₂ electrode was sintered at 450°C for 30 minutes to create a thin, mesoporous TiO₂ layer over conductive glass [16]. Depending on the concentration of the natural dyes and the thickness of the TiO₂ layer, the TiO₂ fired electrode (anode) is immersed in the natural dyes for sensitization for 45–60 minutes at room temperature. The TiO₂ electrode is positioned against the counter electrode cobalt sulphate layer on conductive glass, and the DSSCs are tightly clamped. To provide enough room for clamping, both electrodes are positioned somewhat off-center [17]. After being poured into the cell, the electrolyte KI solution uses capillary action to cover the whole inner surface of both electrodes. Two electrodes are held together using binder clips.

PHOTOGRAPHIC ELECTROCHEMICAL CHARACTERIZATION MEASUREMENTS

A design of the experimental setup used to evaluate the performance of DSSCs is shown in Figure 1. It is made up of a string of potentiometers with values between 0 and 10 K. The circuit is coupled with a sensitive digital voltmeter with a least count of 0.1 V and an ammeter with a least count of 0.001 A. Setting the potentiometer resistance to its maximum value yields the open circuit voltage Voc, while setting it to its lowest value yields the short circuit current Isc. The highest current and voltage that the cell can produce for a given value of resistance is known as I_{max} and V_{max}. This value of resistance is derived by adjusting the resistance from minimum to maximum so that their output is maximal when compared to outputs for other resistance values. The manufactured DSSCs are put to the test under a natural light source. With the assistance of a calibrated pyranometer, the sun intensity is determined [18].

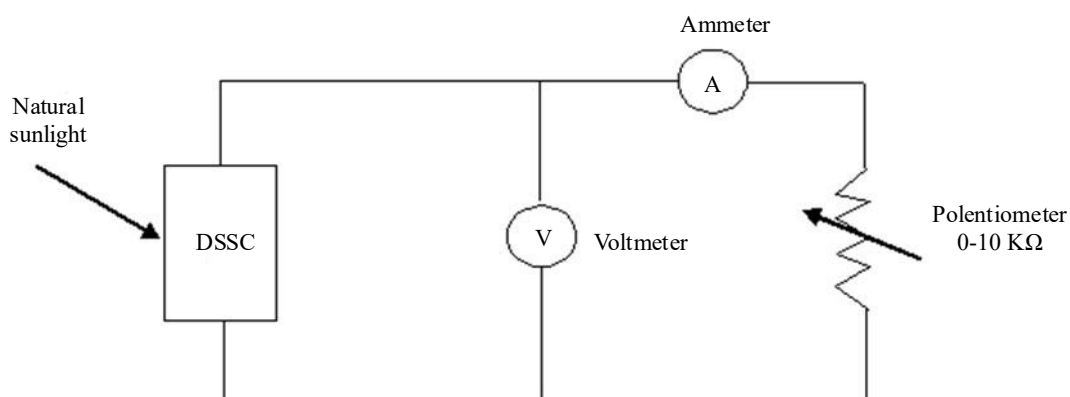


Figure 1. Experimental setup.

Short circuit current (I_{sc}), open circuit voltage (V_{oc}), current density (J_{sc}), power (P), fill factor (FF), and energy conversion efficiency (η) are used to assess the performance of DSSCs.

- i. The fill factor of the DSSC can be calculated using equation,

$$FF = (I_{max} \times V_{max}) / (I_{sc} \times V_{oc}),$$

Where, I_{max} - Current at P_{max} (A)
 V_{max} - Voltage at P_{max} (V)
 I_{sc} - Short-circuit current (A)
 V_{oc} - Open-circuit voltage (V)

- ii. Efficiency of the solar cell is defined as

$$\eta = I_{sc} \times V_{oc} \times FF / P_{in} \times \text{Area of DSSC}$$

Where P_{in} is the illumination input power at surface of the DSSC.

RESULTS AND DISCUSSIONS

Performance of Natural Dyes

The photoelectric parameters of DSSCs sensitized with the extract of all ten natural dyes were measured and calculated. Photovoltaic tests of DSSCs were conducted by evaluating the current-voltage (I-V) curves under natural sunlight. Open circuit voltage (V_{oc}), short circuit current density (J_{sc}), fill factor (FF) and conversion efficiency (η) were used to assess the performance of DSSCs [19]. These performance parameters of the DSSCs sensitized with natural dyes using cobalt sulphate as a counter electrode are listed in Table 2.

Table 2. Photovoltaic performance of DSSCs for different natural dyes using Cobalt sulphate as a counter electrode

Name of dye	V_{oc} (V)	J_{sc} ($\mu\text{A}/\text{cm}^2$)	P_{max} (μW)	FF	η (%)
Hibiscus	0.442	235.43	404.51	0.647	0.08
Black grapes	0.449	188.70	336.84	0.661	0.066
Spinach	0.521	169.45	299.87	0.564	0.059
Orange	0.356	27.07	24.25	0.42	0.004
Pomegranate	0.365	26.7	23.56	0.406	0.004
Sugarcane	0.378	25.02	26.87	0.472	0.005
Papaya	0.388	21.7	21.31	0.418	0.004
Lime	0.346	18.39	19.41	0.507	0.003
Pineapple	0.348	16.14	10.18	0.30	0.002
Sweet lime	0.365	12.2	9.65	0.36	0.001

Figure 2 and 3 shows the typical I -V (Current-Voltage) curve and Power curve respectively of the DSSCs made from cobalt sulphate as a counter electrode using the sensitizers extracted from hibiscus, black grapes and spinach at irradiance (G) of 828 W/m². Since power produced by natural dyes viz. sugarcane, papaya, lime, pineapple and sweet lime was negligible hence IV curve and Power curve for these natural dyes were not generated. This is typical variation for all types of solar cells for all natural dyes and hence for representation in I-V curve and power curve of three natural dyes are considered. It is also observed that, lime, pineapple and sweet lime extract dyed cells showed marginal short lived photovoltaic activity due to rapid anthocyanin decay and consequently no current vs. voltage diagram is generated [20].

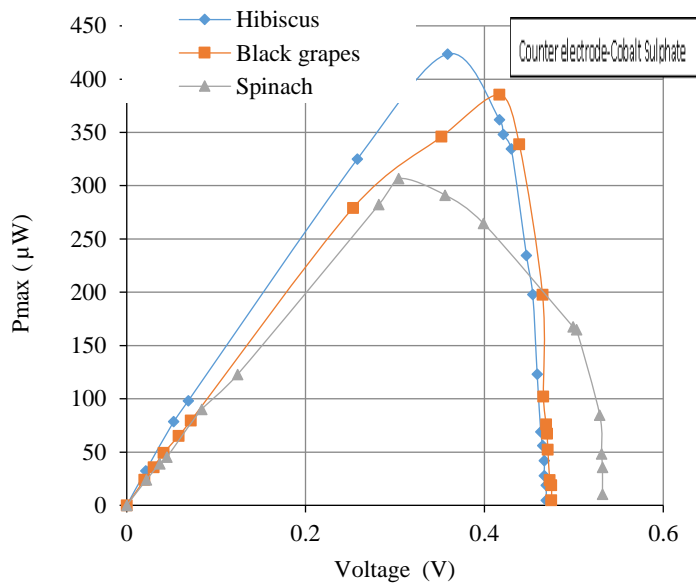


Figure 2. I-V curve for natural dyes with cobalt Sulphate as a counter electrode.

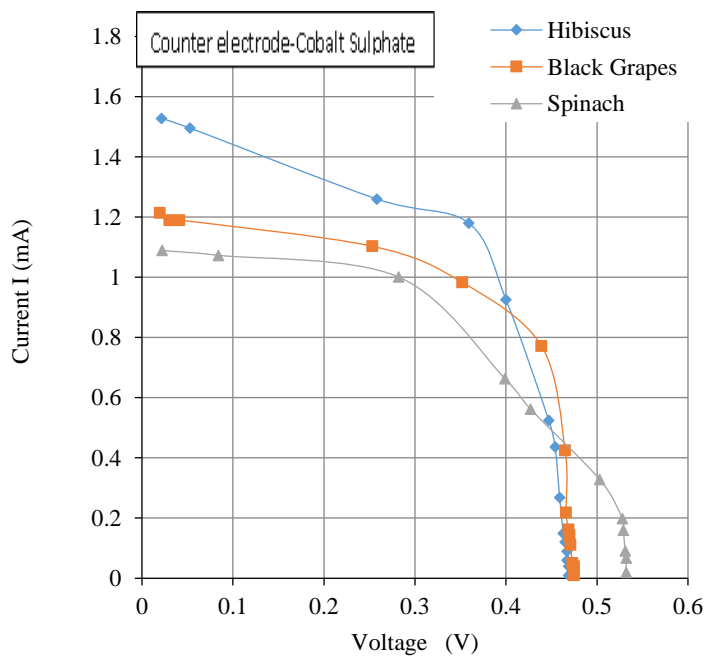


Figure 3. Power curve for natural dyes with cobalt sulphate as a counter electrode.

The following observations were made.

- i. For hibiscus dye, it is found that, as the V_{oc} increases, power increases steadily and becomes maximum at 0.359 V at a maximum power of 423.62 μW . After this point, as the V_{oc} increases, power produced by the solar cells drops down as shown in Figure 2. Same trends were observed for all natural dyes.
- ii. The power produced by the DSSCs increases steadily with the voltage becomes maximum and then drops down.
- iii. The maximum power produced by hibiscus is highest and is lowest for sweet lime amongst under all DSSCs under investigation [21].

Table 3 shows the performance parameters of the DSSCs made from natural dyes using graphite as a counter electrode. Figure 4 and 5 shows the typical I -V (Current-Voltage) and power curves of the DSSCs made from graphite as a counter electrode using the sensitizers extracted from hibiscus, black grapes and spinach at irradiance (G) of 828 W/m^2 .

Table 3. Photovoltaic performance of DSSCs for different natural dyes using graphite as a counter electrode

Name of dye	V_{oc} (V)	J_{sc} ($\mu\text{A}/\text{cm}^2$)	P_{max} (μW)	FF	η (%)
Hibiscus	0.392	215.33	320.01	0.630	0.063
Black grapes	0.436	183.60	302.04	0.630	0.059
Spinach	0.459	113.33	106.15	0.323	0.021
Orange	0.365	35.37	22.21	0.283	0.004
Pomegranate	0.380	23.60	20.34	0.378	0.004
Sugarcane	0.364	20.47	13.33	0.286	0.002
Papaya	0.330	18.08	6.51	0.172	0.001
Lime	0.305	14.20	4.97	0.182	0.0009
Pineapple	0.241	8.97	2.52	0.181	0.0005
Sweet lime	0.247	5.10	1.19	0.148	0.0002

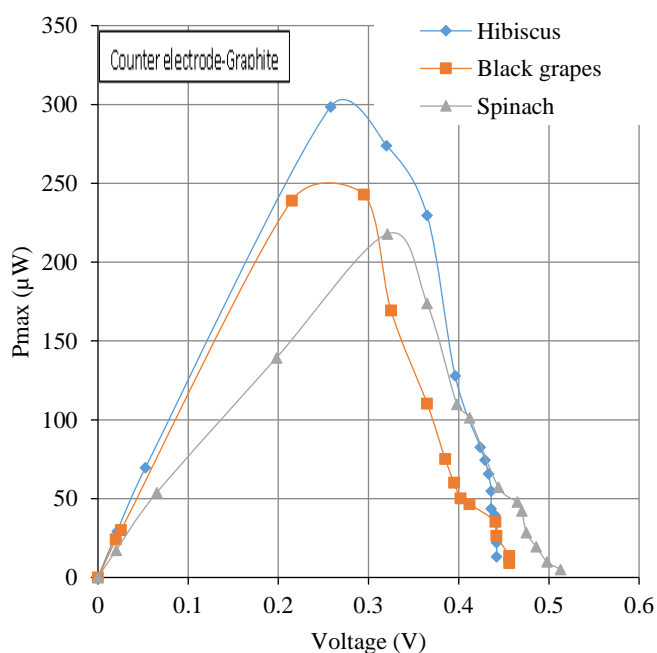


Figure 4. I-V curve for hibiscus, black grapes and spinach dyes with graphite as a counter electrode

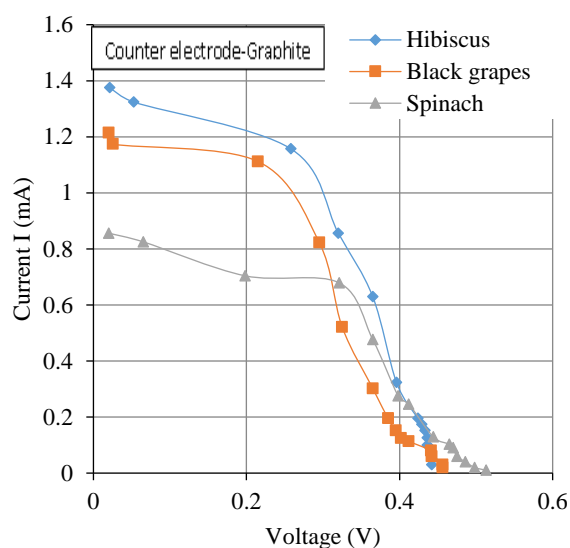


Figure 5. Power curve for hibiscus, black grapes and spinach dyes with graphite as a counter electrode

It is seen that, similar type of trend was also obtained as shown in Figure 4 and 5 for DSSCs made from graphite as a counter electrode except the power produced by DSSCs using cobalt sulphate as a counter electrode is more than that of graphite. For all DSSCs, as the voltage increases, power increases linearly. After some time, peak power is observed and power starts decreasing with respect to voltage. Power developed by solar cells for all natural dyes are different and is higher for hibiscus followed by black grapes, spinach, orange and pomegranate. The power produced by DSSC sensitized with the hibiscus is superior to those sensitized with black grapes and spinach. Solar cells sensitized with spinach shows less output as compared to those sensitized with hibiscus and black grapes. One of the reason for the same is the poor link between their dye molecule and the TiO_2 coating. Even though spinach's chlorophyll content is essential for the plant's capacity for photosynthesis, this capacity cannot produce a high photo-to-electric conversion in DSSC due to the absence of bonds between the spinach molecule and TiO_2 film, which allow electrons to move from excited dye molecules to the TiO_2 film [22].

It is observed that, out of ten natural dyes, hibiscus, black grapes and spinach have the best performance parameters. Hibiscus dye gives more output as compared to other dyes for both counter electrodes. Order of performance DSSCs using both counter electrodes and natural dyes is as follows. Hibiscus has highest output followed by black grapes, spinach, orange, pomegranate, sugarcane, papaya, lime, pineapple and sweet lime. For DSSCs with hibiscus dye and using a cobalt sulphate as a counter electrode, highest current density ($235.43 \mu\text{A}/\text{cm}^2$) is obtained with an open circuit voltage of 0.442 V, fill factor of 0.647 and efficiency of 0.08%. While the lowest current density ($12.2 \mu\text{A}/\text{cm}^2$) is obtained for sweet lime using a cobalt sulphate as a counter electrode with an open circuit voltage of 0.365 V and FF of 0.360. Similarly, for DSSCs with hibiscus dye and using graphite as a counter electrode, the highest current density ($215.33 \mu\text{A}/\text{cm}^2$) is obtained with an open circuit voltage of 0.392 V, fill factor of 0.630 and efficiency 0.06%. While, the lowest current density ($5.10 \mu\text{A}/\text{cm}^2$) is obtained for sweet lime using graphite as a counter electrode with an open circuit voltage of 0.247 V and fill factor of 0.172. DSSCs with natural dyes such as sugarcane, pineapple, lime, pineapple and sweet lime extract dyed cells showed marginal short lived photovoltaic activity due to rapid anthocyanin decay and consequently power produced by DSSCs using these dyes are less [23].

It is found that, DSSCs with lime dye using both counter electrodes shows extremely high initial current spikes, but dropped off drastically as time elapsed and hence overall performance of lime dye is less. It is also seen that, there is much variations in the current densities of the DSSCs made from the natural dyes using both counter electrode. This is also due to the contact between the natural dyes and the TiO_2 film which results in enhancing the conversion efficiency of DSSCs. The anthocyanin

concentration of natural dyes is another potential explanation for the exceptional performance. Voc and Jsc values increase with the amount of anthocyanin in the dye due to their capacity to absorb light and transform it into electrons. Hence, the relative merits of the anthocyanin content in natural dyes used in this study are examined. Blue, purple, and red pigments called anthocyanins are present in a variety of fruits, flowers, and plants. As can be seen from the above, the hibiscus based anthocyanin dye gives significantly higher output compared to the dye extracted from lime, pineapple and sweet lime [24]. This is due to the fact that content of anthocyanin in the hibiscus is more (456 mg/100 gram) as compared to pomegranate (6.902 mg/100 gram) and sugarcane (0.253 mg/100 gram). Spinach also had good photo electrochemical parameters due to the presence of chlorophyll (42.7 mg/100 gram) which also contains hydroxyl groups but a lesser amount of anthocyanin. Lime, pineapple and sweet lime dyes shows less performance due to the negligible content of anthocyanin [25].

Conversion efficiency of DSSC is strongly depends on bonds present between the dye molecules and TiO₂ particles. Content of anthocyanin & chlorophyll also plays an important role in natural dyes. However, in this work, DSSCs sensitized by natural dyes mainly composed of orange, pomegranate, sugarcane, papaya, lime, pineapple and sweet lime produces considerable power but did not offer high conversion efficiencies as compared to hibiscus, black grapes and spinach. The reason for this is that there is less interaction between the dye and the TiO₂ molecules which allows electrons to move [22].

VARIATION OF PERFORMANCE PARAMETERS OF DSSCS FOR NATURAL DYES

Figure 6 and 7 shows a comparative performance of Voc and Jsc of all ten natural dyes with cobalt sulphate and graphite as a counter electrode.

It is found that, Voc of DSSCs made from all ten natural dyes with cobalt sulphate and graphite as a counter electrode is more or less constant and lies in between the range of 0.36 V to 0.44 V. DSSCs made with natural dyes such as a hibiscus, black grapes and spinach using cobalt sulphate as a counter electrode have produced higher Jsc as compared to other natural dyes. Lime, pineapple and sweet lime has produced less values while the orange, pomegranate sugarcane and papaya have produced moderate values of Jsc.

With the use of graphite and cobalt sulphate as a counter electrode, figure 8 compares the power performance of all 10 natural dyes. It is observed that, power developed by DSSCs with hibiscus, black grapes, spinach, orange and pomegranate using cobalt sulphate as a counter electrode produced large power while a small amount of power is generated by DSSCs made from sugarcane, papaya, lime, pineapple and sweet lime dyes. This is due to the fact that current produced by these dyes are less as compared to hibiscus, black grapes and spinach [26].

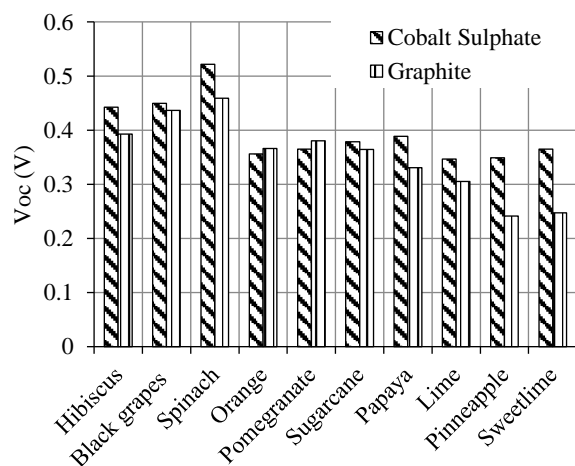


Figure 6. Voc of dyes using cobalt sulphate & graphite

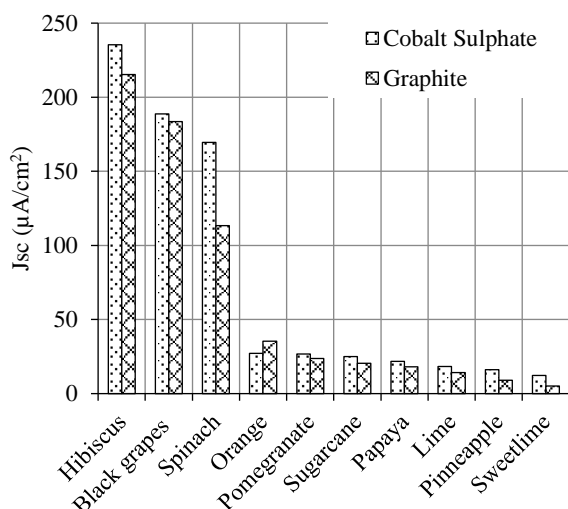


Figure 7. J_{sc} of dyes using cobalt sulphate and graphite.

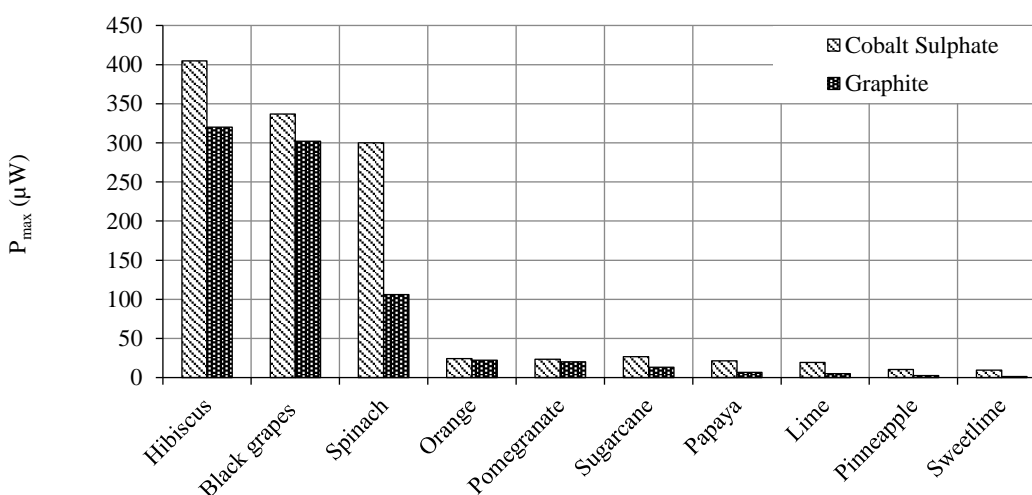


Figure 8. Power of dyes using cobalt sulphate and graphite.

The efficiency and power of cell sensitized by the hibiscus extract is significantly higher than that of dyes sensitized by the other extracts. This is because of the more content of anthocyanin in the hibiscus. The absorption of high intensity light through the hibiscus extract on TiO₂ also plays a significant role which could be accounted for a better performance. Other four extracts viz. black grapes, spinach, orange and pomegranate has also exhibit remarkable photo electrochemical performance parameters because they are helped by the electrolyte composed of KI. The natural dyes extracted from sugarcane, papaya, lime pineapple and sweet limes shows a comparatively less performance as shown in Table 2 and 3 due to the less amount of dye adsorbed onto the TiO₂ film in these extract.

Figure 9 and 10 shows an effect of FF and efficiency of DSSCs all natural dyes using cobalt sulphate and graphite as a counter electrode. It has been noted that, the relationship between FF and natural dyes is not linear. The efficiency of DSSCs with hibiscus, spinach and black grapes dyes is higher and goes on decreasing for other dyes. It is seen that, there is variation in power produced by DSSCs with different natural dyes. From the literature review, it is found that, the efficiencies of DSSCs made from natural dyes are not more than 1% [27]. Performance of these dyes in terms of efficiency using graphite as a counter electrode is found out to be 0.03% to 0.07% which is in good agreement with the efficiencies of 0.06% reported by Abel F. Ole [28].

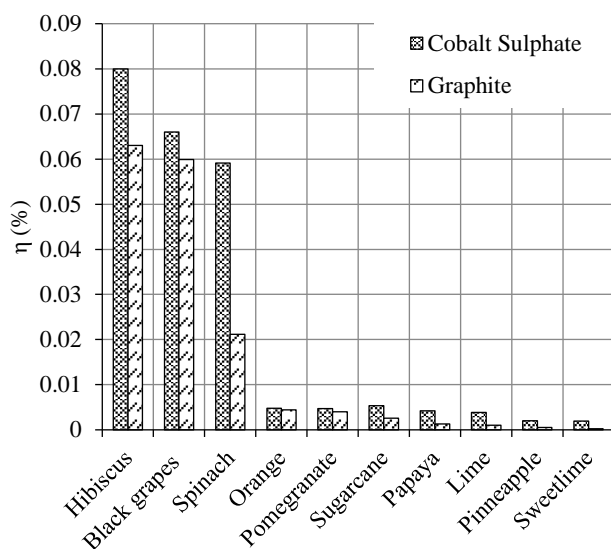


Figure 9. FF for dyes using Cobalt Sulphate and Graphite

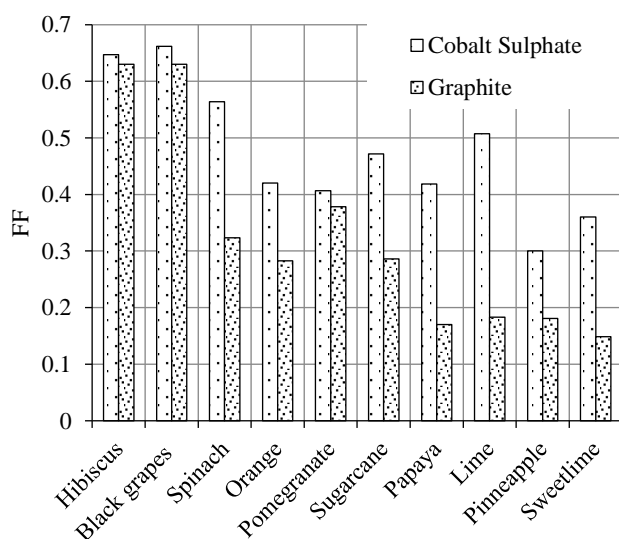


Figure 10. η of dyes using cobalt sulphate and graphite.

In general, the performance of the DSSCs is affected by many factors. Some of them are content of anthocyanin & chlorophyll, light intensity, size of DSSC and type of electrolyte. In order to produce efficient solar cell, there should be i) rapid charge injection in the solar cells. ii) no obstruction for the natural dyes to form bond with the oxide surface of the TiO_2 iii) strong bond between dye molecule and the TiO_2 film. This in fact justifies why dyes from lime, pineapple and sweet lime are not good sensitizer, while the hibiscus are excellent source of dye for sensitization. Low power output was reported for the DSSCs' made from lime, pineapple and sweet lime extracts. This suggests that their dye molecule and TiO_2 films have a weak connection which is consistent with the above discussions. Normally, natural dyes suffer from low performance. This is due to degradation of content in natural dyes. Resistance occurs in the TiO_2 /dye/electrolyte interface also leads to a decrease in J_{sc} . Some complication such as resistance due to more TiO_2 film thickness layer causes poor physical contact between the electrolyte and TiO_2 film surface which results in no electron injection, reducing reaction and slowed down dye absorption.

CONCLUSIONS

Ten natural dyes from nature were successfully used to produce a power and their photo responses are studied for possible application as sensitizers in DSSC. Although the power output and efficiencies of DSSCs using the natural dyes comparatively gives less efficiency which is not sufficient enough for large scale production, the results are encouraging for reducing the cost of DSSCs and to explore new dyes over existing natural dyes. In the near future, natural dyes used in this work are capable of replacing some of the manmade ruthenium dyes used in DSSC and can be possible alternative for the remote areas.

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