25th ABCM International Congress of Mechanical Engineering
October 20-25, 2019, Uberlândia, MG, Brazil

COBEM-2019-0916
PERFORMANCE OF DYE-SENSITIZED SOLAR CELLS USING LEUCANTHEMUM VULGARE FLOWERS AS NATURAL SENSITIZERS

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Abstract. *The growing demand for renewable energy, especially the solar energy, has been seeking new ways to enhance a new generation of solar cells, as the DSSC´s. This technology is based on a nanostructured semiconductor anode, an electrolyte and a photoelectrochemical cathode. DSSC's are considered attractive devices because they combine organic and inorganic materials, and are mechanically resistant. In this work, TiO_2 (Titanium Dioxide) was used for the cell anode, and for the cathode, platinum. Moreover, natural dyes containing anthocyanins were prepared using three specimens of daisy flowers (Leucanthemum vulgare). The photovoltaic performance was carried out to understand the effect of the different photo-anodes and the interaction with the dye molecules on the overall cell's efficiency. DSSC's sensitized with daisy flowers dye showed open-circuit voltage (Voc) of 520 mV and efficiency of 0.87 %. These results show that the DSSC's, using daisy flowers extracts as photosensitizers, are suitable for the fabrication of natural dye-based DSSC's.*

Keywords: *renewable energy, DSSC´s, anthocyanin.*

1. INTRODUCTION

The Sun is an obvious source of clean and cheap energy, and is already used by Nature to sustain almost all life on Earth. Therefore, harvesting the power of the Sun with these photovoltaic technologies works like a reasonable answer to the present energy challenge.

The DSSC´s bring an answer to this, working like a low-cost solution, using abundantly available raw materials. This is an important point that highlights the DSSC's, considering performance, cost efficiency and raw materials availability during the production of solar cells. Therefore, DSSC's have huge potential for the scientific community on the quest for the development of clean and sustainable energy (Atli et al., 2018).

The DSSC'S are composed of two conductive glasses of SnO_2 (tin oxide IV) doped with indium (ITO film), one of these glasses is deposited TiO_2 (titanium dioxide) along with natural dye forming the anode, the other glass is deposited the platinum solution forming the cathode the junction of these two electrodes plus the electrolyte it is possible to mount a DSSC.

In this work we have designed and assembled new DSSC, using the natural dye extracted from flowers petals of the family of *Leucanthemum vulgare*, which was optically characterized by Fourier Transform Infrared (FTIR) Spectroscopy. The morphological changes and depth profile were characterized by Atomic Force Microscope (AFM). The photoelectrochemical properties as open circuit voltage (Voc), short-circuit current density (Jsc), fill factor (FF), and conversion efficiency (η) of the fabricated DSSCs using this extract as a photosensitizer were also investigated.

2. WORKING PRINCIPLE

The working principle of a DSSC is based on the electrochemical interactions of the components. The anode, made of semiconductor material, absorbing the ultraviolet radiation allowing the absorption of the light visible for it is immersed in an anthocyanin-based dye, acting as a sensitizer for the DSSC. The occurrence of the photoelectric effect is guaranteed by the presence of anthocyanin in the dye, vegetable pigment responsible for the coloring of flowers and fruits.

When the anode receives energy coming from different wavelengths, referring to the visible light spectrum, electrons, unstable in their bonds, receive enough energy to reach an excited state. So, they pass from the highest occupied molecular orbital (HOMO) for the lower unoccupied molecular orbital (LUMO). If the LUMO level energy is higher than that of the oxide conduction band, it breaks the bandgap of the semiconductor material. The anode then carries the diffusion transport to the external circuit, arriving at the cathode.

The electrolyte has the function of making the replacement of the electrons and is also responsible for the closing of the cycle, helping the reduction of the photoanode and the oxidation of the photocathode. The electrolyte is composed of iodine / triiodine, and has the function of electron transport between the electrodes.

The iodine ions (I^-) fill the holes in the pigments and are converted to I_3^- on the nanoporous surface of the semiconductor oxide. When the electrons are injected into the oxide, the dye oxidizes, and the regeneration of the species is controlled by the electrolyte, which in turn is regenerated by the counter electrode.

3. EXPERIMENTAL PROCEDURE

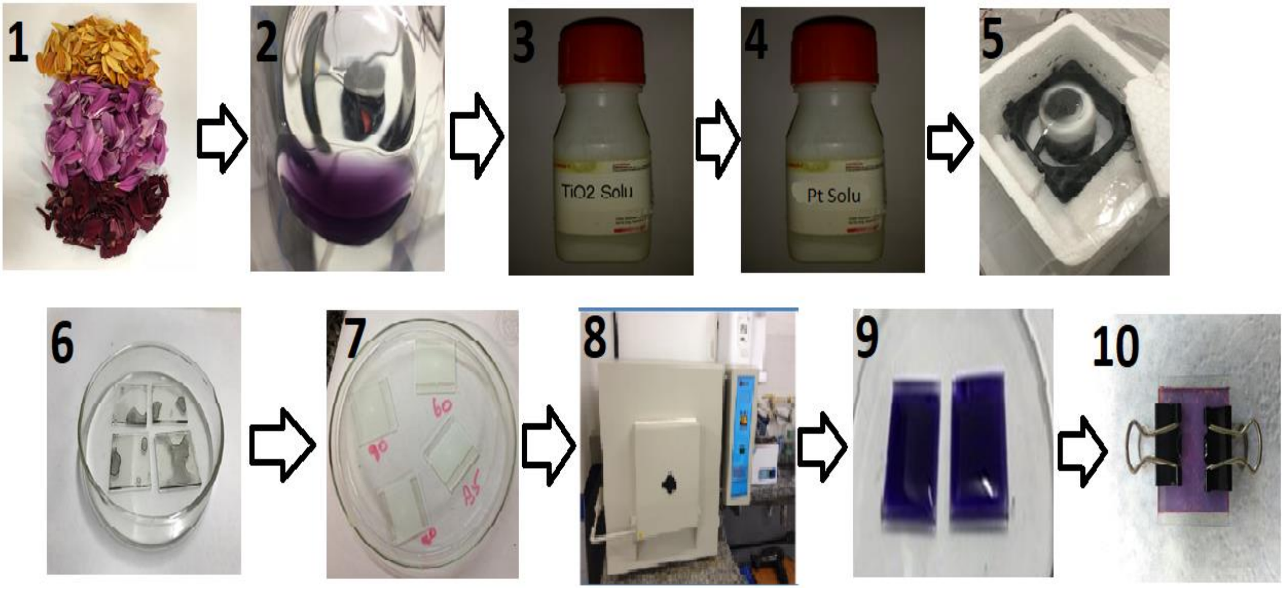

Figure 1. Flow chart of the full cell preparation.

Figure 1 summarizes the experimental procedure for this work:

1- *Leucanthemum vulgare* petals were selected
2- The dye was extracted from the petals
3- The anode solution was prepared
4- The chathode solution was prepared
5- The anode and cathode solutions were deposited on an ITO plate by a spin coater method
6- Cathode after spin coater deposition
7- Anode after spin coater deposition
8- The annealing was performed after the deposition on a muffle furnace
9- The dye extracted was deposited on the annealed anode
10- The cell was completed when the anode and cathode were connected by the electrolyte on the middle

3.1 Dye Preparation

The natural dye was prepared from daisy flowers (*Leucanthemum vulgare*), as they were bought in a flower shop in Rio de Janeiro. For the preparation, the flowers were washed with isopropyl alcohol and subsequently dried at room temperature (only the petals were used). Ethanol was used as solvent.

After drying, the flowers were dipped into ethanol and mixed for thirty minutes by the magnetic mixer. Finally, the solution was filtered and stored in a freezer at -20 ºC, which can be kept for months.

3.2 Anode Preparation

For assembling the cell, the TiO_2 thin film (photoanode) was deposited over the ITO glass substrate. This deposition was performed by spin coating technique at 1000 rpm for 10 seconds, using a self-built equipment designed and manufactured for this purpose and then, the samples were annealed at 450 °C for 30 minutes.

3.3 Cathode Preparation

The cathode is where the negative polarity is applied to drive the cell, as the basic operation of the DSSC's is performed by the interaction of a semiconductor material (anode) and a dye, which works as a sensitizer. The platinum solution was prepared by mixing 0.5 M of Platinum powder and 5 ml of ethanol, and then, the solution was mixed on an ultrasonic device for 30 minutes.

3.4 Electolyte Preparation

In this work, a new electrolyte was prepared, dissolving 2.075 g of KI and 0.19 g of I_2 in 25 ml of polyethylene glycol / acetonitrile mixture (4:1 V/V), and then, the solution was homogenized by an ultrasonic mixer for 30 minutes. The as prepared electrolyte was then injected in the space between the two electrodes of the cell.

4. RESULTS AND DISCUSSION

4.1 Atomic Force Microscopy (AFM)

Atomic force microscopy (AFM) was used to characterize the surface's morphology of the photoanode sample. Topographic image of the photoanode surface was taken in the conventional non-contact mode before dye loading onto TiO_2 film (Ferreira et al., 2018). Analyzing the Fig. 2, it can be concluded that the sample has a desired morphology, a mesoporous structure that favors the dye absorption. Also, was observed a low rate of irregularities, proving that the deposition method was successful when compared to the literature (Polo et al., 2004).

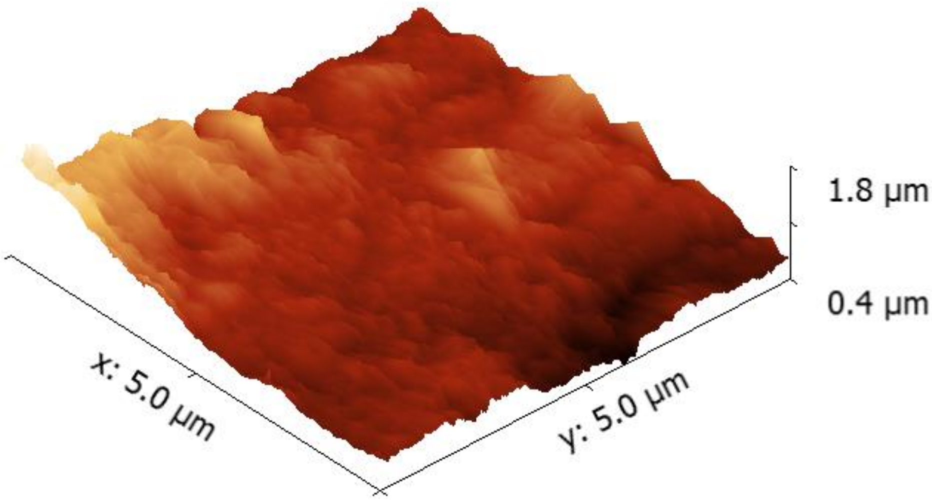

Figure 2. Three-dimensional AFM image of TiO_2 anode.

4.2 Fourier-Transform Infrared spectroscopy (FTIR)

With the FTIR analysis, it is possible to verify that the dye extracted contain high concentration of anthocyanin molecules and may exhibit better efficiency as a dye, once studies indicate that anthocyanin works as a good light absorbent (Sampaio et al., 2019). The FTIR is used to obtain an infrared spectrum of absorption which indicates presence of atom's bonds. As it can be seen in the Fig. 3, the region between 3600 and 3200 cm^{-1} shows a peak referent to the H-O-H bond, that indicates presence of air's water contamination and in the ethanol used as solvent as well. The peak 3000 to 2750 cm^{-1} is characteristic from the C-O-C bond, present in the anthocyanin's radical, and the region between 1800 and 1550 cm^{-1} refers to the C=C bond, present on the anthocyanins aromatic structure (Law et al., 2005).

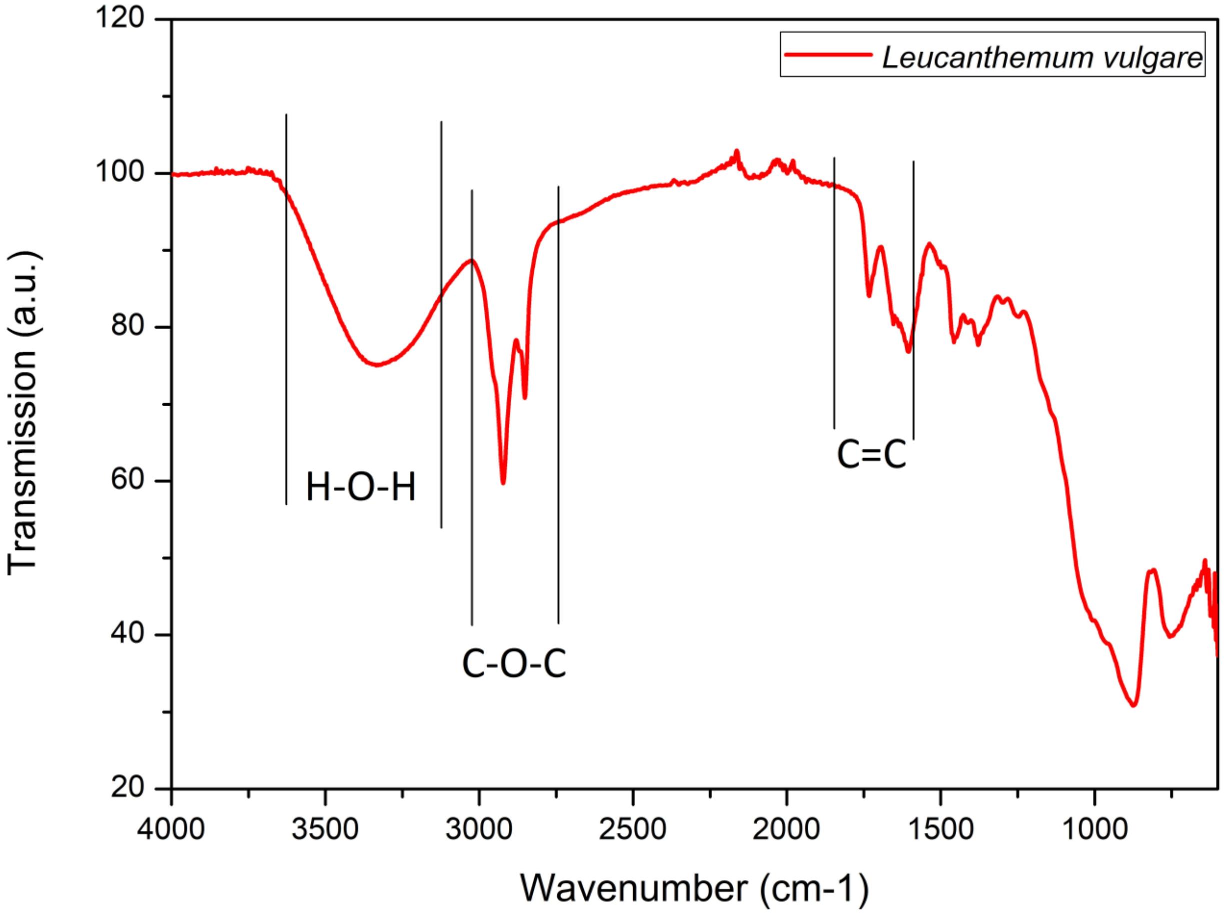

Figure 3. FTIR spectrum for Daisy Flower.

4.3 Photovoltaic Performance

To study the photovoltaic characteristics of daisy-based dyes, photovoltaic parameters such as short-circuit current density (Jsc), open circuit voltage (Voc), Fill Factor (FF), maximum power (Pmax) and efficiency (η) were determined. The efficiency (η) and FF were obtained from the following equations:

$$FF = (J_{max} \times V_{max})/J_{sc} \times V_{oc} \quad (1)$$

$$\eta\ (\%) = (J_{sc} \times V_{oc} \times FF/I_{Ins}) \times 100 \quad (2)$$

The open-circuit voltage (Voc) is the maximum voltage generated by the photovoltaic device, and can be obtained experimentally by measuring the voltage at the terminals of an uncharged cell under certain incident temperature and radiation conditions. The short-circuit current (Isc) is the maximum current that the solar cell can supply, and can be obtained by short-circuiting the terminals of the cell and measuring the current flowing through the device. Light Incident is the light energy emitted by the source that reaches the cell, also known as input energy. With these values we can find important parameters for the solar cells like efficiency and Pmax.

4.3.1 IV Graphics

An important parameter to evaluate the photovoltaic performance is the IV curve, which is the current *versus* voltage curve, and can give the idea of how efficient is the electrons transfer over the surface of the DSSC's (Grätzel, 2003). Additionally, is used to determine important factors as the short circuit current (Isc), open circuit voltage (Voc) and Fill Factor (FF). The IV curve for the *Leucanthemum vulgare* cell is represented in Fig. 4.

The results were promising, showing an Isc of 0.419 mA, a Voc of 0.52 V and a Fill Factor of 0.27. All ends on an efficiency conversion of 0.87 %, a good result when compared to others previous results, as can be seen on table 1.

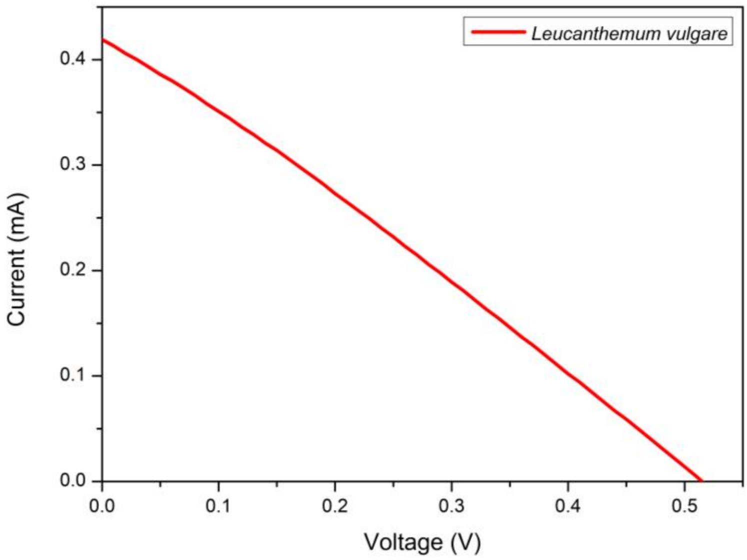

Figure 4. *Leucanthemum vulgare* IV results.

Table 1. Photovoltaic Performance Measurements of DSSC's.

Dye	I_{sc} (mA)	V_{oc} (V)	*FF*	η (%)	Ref.
Leucanthemum vulgare	0.42	0.52	0.49	0.87	This work
Thunbergia erecta	0.27	0.55	0.40	0.37	B. C. Ferreira et al. (2018)
Lithospermum	0.38	0.41	0.29	0.13	D. M. Sampaio et al. (2019)

4.3.2 PV Graphics

Another important analysis is the maximum output power (Pmax) which is obtained by choosing the point on the I-V curve corresponding to which the current (Imax) and potential product (Vmax) gives the maximum value. Figure 5 shows the power *versus* potential curve, and the corresponding power (Pmax) obtained was 58,5 μWcm $^{-2}$.

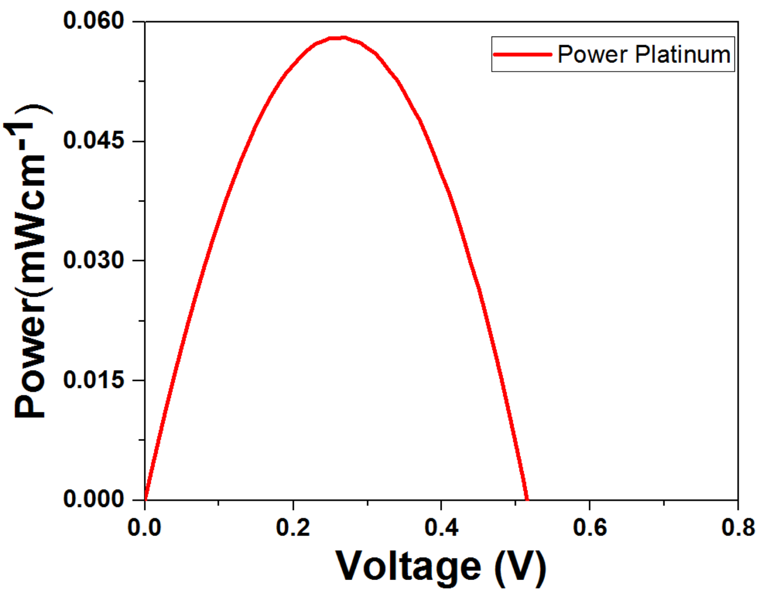

Figure 5. *Leucanthemum vulgare* PV graphics.

5. CONCLUSION

This work successfully presented Leucanthemum vulgare (daisy flowers) dyes as natural sensitizers for DSSCs, and show good overall efficiency, achieving 0.87 %, using TiO_2 as anode and platinum as cathode. As shown, there was a great advantage of the daisy dye in relation to other similar dyes, because of its high values of efficiency, Voc and Isc. In fact, daisy flowers as sensitizers appear to be a reasonable choice for DSSC's, due to its high efficiency and great availability. To improve the current density, and consequently, the fill factor values of the samples, the electrolyte solution can be replaced for room temperature ionic liquids-based I_-/I_3^- redox electrolytes. Therefore, is hoped that this work will serve as a research source for the use of natural dyes as light harvesting constituents in DSSC's.

6. ACKNOWLEDGEMENTS

This work was supported by the Brazilian agencies CAPES (BEX 5383/15-3), CNPq (301868/2017-4) and FAPERJ (E-26/110.087/2014, /213.577/2015 and /216.730/2015).

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8. RESPONSIBILITY NOTICE