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# Dye-Sensitized Solar Cells Using Natural Dye Extracted from *Hibiscus acetosella* Leaves as Photosensitizer

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***Abstract:*** *As the human population continues to grow, the need for energy that is most of the time filled in from fossil sources increases as well. This is the reason why problems with climate change and others have risen. Recently, technologies were developed to keep within limits the effects of climate change due to excessive carbon emission; one of these is the development of dye-sensitized solar cells (DSSCs) that converts the energy of the sun into its usable form which is electricity using natural plant pigments. DSSCs based on natural dyes extracted from Hibiscus acetosella leaves were fabricated in this study. The extracts of leaves were prepared using different extracting solvents – water, ethanol, and acidified ethanol. The extracted dyes were subjected to a UV-Vis spectrophotometer and the fabricated DSSCs were subjected to photoelectrical measurements which include voltage, current, and power. Results have shown that the maximum optical absorption of the extracted natural dyes ranges from 400 nm to 540 nm which confirmed the presence of plant pigments anthocyanin, betacyanin, and betaxanthin. Photoelectrical measurement results showed that the DSSCs with water-extracted Hibiscus acetosella leaves dye performed the best in terms of voltage, current, and power and this performance has a significant difference when compared to the DSSCs with no dye. This result highlights the practicability of the use of natural pigments as photosensitizers in DSSCs especially since these natural plant pigments are non-toxic, easily disposed of, cheaper, and environmentally friendly which can contribute to a sustainable solution for future energy production.*

***Key Words:*** *Dye-sensitized solar cell, natural dye, photosensitizer.*

## 1. INTRODUCTION:

The need for energy is rising day by day due to the increase in the population and the usage of electronic gadgets as ever before (Kalyani and Dhoble, 2018). With the current trend that this need for energy is most of the time filled in from fossil sources, the problem of climate change and other environmental concerns are also increasing accordingly. Thus, there are clamors and call to shift from fossil-dependent energy source to renewable resources. In the global context, the growing abundance of cost-competitive renewable energy technologies (especially solar) is accelerating the transition toward decarbonized energy systems (World Energy Council, 2019). In the Philippines, the government has passed into law RA 9513, or the Renewable Energy Act of 2008 which aims to accelerate the exploration and development of renewable energy resources, achieve energy self-reliance, and reduce the country's dependence on fossil fuels (Guarin, 2013). The exploitation of sunlight and air as a substantial renewable energy source is an important research and development domain over the past few years (Jha, Bilalovic, Jha, Patel, Zhang, 2017) more especially with sunlight as researches using this energy source mimic how plants use the seemingly unlimited energy coming from the sun into a usable form of energy. One of the technologies which were fabricated following how nature converts energy coming from the sun is the dye-sensitized solar cell. Due to their low cost, straightforward preparation process, minimal toxicity, and ease of manufacture, dye-sensitized solar cells (DSSCs) are among the thin-film solar cells that have been the subject of extensive research for more than 20 years (Sharma, Sharma, and Sharma, 2018). Dye-Sensitized Solar Cell (DSSC) is a solar cell device that works using electrochemical principles in which sensitive dyes are absorbed in the $TiO_2$ photoelectrode layer (Trihutomo, Soeparman, Widhiyanuriyawan, and Yuliati, 2018). Venkatraman, Raju, Oikonomopoulos, and Alsberg (2018) explained that DSSCs are influenced by various components of the cell such as the dye, electrolyte, electrodes, and additives among others leading to varying experimental configurations. But among others, the performances of dye-sensitized solar cells are mainly based on dye used as a sensitizer.

Nowadays, the study of dyes extracted from natural resources is the main concern for researchers (Richhariya, Kumar, Tekasakul, and Gupta, 2017). A promising advancement in this technique is the use of natural dyes. Through a straightforward extraction technique, natural dyes are a substitute for expensive chemical synthesis methods and reduce the high cost of metal complex sensitizers. Natural dyes are widely available, simple to extract, and environmentally benign. These can be extracted from flowers' petals, leaves, roots, and bark in the form of anthocyanin, carotenoid, flavonoid, and chlorophyll pigments.

In nature, anthocyanins are responsible for the red, purple, and blue colors of plants (Khoo, Azlan, Tang, & Lim, 2017). One of the many locally-available plants with the prospect of having this plant pigment is Hibiscus acetosella which is locally known as *labog*. Thungmungmee, Wisidsri, & Khobjai (2018) confirmed the presence of anthocyanin in the Hibiscus acetosella plant. With this chemical property of this abundant plant, this experimental research was conceptualized and explored the role of natural dyes extracted from *Hibiscus acetosella* plant in fabricating Dye-Sensitized Solar Cells (DSSCs). This study, specifically, aimed to look into the potential of the natural dye that was extracted from Hibiscus acetosella or *labog* leaves as a photosensitizer for dye-sensitized solar cells (DSSCs). Furthermore, it looked into the different extracting solvents as a key factor in the performance of the fabricated dye-sensitized solar cells.

Specifically, this research sought to answer the following research questions:

- What is the optical absorption of *Hibiscus acetosella* leaves dye extracts using different extracting solvents?
- What is the performance of the dye-sensitized solar cells with natural dye extracted from *Hibiscus acetosella* leaves as photosensitizer using different extracting solvents in terms of its (1) voltage (mV), (2) current (mA), and (3) power (mW)?
- Is there a significant difference in the performance of the dye-sensitized solar cells with natural dye extracted from *Hibiscus acetosella* leaves as photosensitizer using different extracting solvents in terms of its (1) voltage (mV), (2) current (mA), and (3) power (mW)?

Given the preceding problems, this null hypothesis was advanced:

- There is no significant difference in the performance of the dye-sensitized solar cells with natural dye extracted from *Hibiscus acetosella* leaves as photosensitizer using different extracting solvents in terms of its (1) voltage (mV), (2) current (mA), and (3) power (mW).

## 2. METHODOLOGY:

**Materials :** *Hibiscus acetosella* leaves were collected from Passi City, Iloilo, Philippines, and plant samples were identified and authenticated by agriculturists from Passi City Agriculture Office. Titanium Dioxide ($TiO_2$), Acetic Acid ($CH_3COOH$), Potassium Iodide (KI), Iodine Crystals, Acetonitrile, Ethylene Glycol, and Ethanol were purchased from Patagonian Enterprises. The Transparent Conductive Indium Tin Oxide ITO (6 Ω/ 20x20x1.1 mm) glasses were purchased through a legitimate online distributor.

**Preparation of Natural Dye Photosensitizer :** Collected *Hibiscus acetosella* leaves were rinsed with tap water followed by distilled water to remove dust and other impurities and were oven-dried for 30 minutes at 150°C. The oven-dried leaves were blended to become powder. The powdered leaves of Hibiscus acetosella were then sieved to attain uniformly-sized particles. A mass of 10 g of the powdered Hibiscus acetosella leaves was weighed and mixed with 100 mL solvent (water, ethanol, acidified ethanol) using a magnetic stirrer for 5 minutes to disperse the powder completely and then was kept in a dark chamber at room temperature for 24 hours, then it was filtered. The extracts were then stored in an Erlenmeyer flask fully covered with aluminum foil to prevent the dye from light exposure for later use.

**Preparation of Solar Cells :** The dye-sensitized solar cells (DSSCs) were fabricated following the work of Sullano and Sia (2018) with some modifications. The DSSCs were made using tin oxide ($TiO_2$) powder suspension prepared by making a watery shampoo-like consistency paste with 1 g $TiO_2$ powder and acetic acid. The TiO2 suspension was deposited on the conductive side of the Indium Tin Oxide (ITO) glass by doctor's blade's method and was sintered by heating the slide for 30 minutes through the use of a hot plate. After cooling, the $TiO_2$ electrode was submerged in the natural dye extract of *Hibiscus acetosella* extracts for 24 hours. A counter electrode was prepared by coating the conductive side of an ITO glass with carbon soot from a lit candle. Two (2) drops of the electrolyte were placed on the counter electrode. The $TiO_2$ electrode was then placed on top of the counter electrode while allowing each side of the glass to hang off for further testing. The solar cell was then sealed using two (2) binder clips.

**Preparation of Electrolyte :** The electrolyte used in the fabrication of dye-sensitized solar cells was made following the work of Gu, Yang, Zhu, Sun, Wangyang, Li, and Tian (2017) with some modifications. 1.08 g of Potassium Iodide

(KI) was weighed and dissolved in the mixed solvent of acetonitrile (20 mL) and ethylene glycol (5 mL) and obtained the solution, then put 0.5 g of iodine ($I_2$) crystals into this solution. After which, the mixture of all of the chemicals was mixed through a magnetic stirrer for 30 minutes. 2 drops of this electrolyte solution were added to each DSSC fabricated.

**Optical Properties of Natural Dyes :** The optical absorption of the as-prepared dye extracts of Hibiscus acetosella leaves was determined using the Shimadzu UV - 1280 UV-Vis spectrophotometer of Gregor Mendel Research Laboratory of the University of San Agustin in the wavelength range of 400-800 nm (Taya, El-Agezl, Abdel-Latif, El-Ghamri, Batniji, and El-Sheik, 2014). The effect of the different extracting solvents on the optical properties of dyes was studied.

**Photoelectrical Measurements :** The performance of a dye-sensitized solar cell was evaluated following the works of Sharma, Sharma, and Sharma (2018) with some modifications by using current (I, mA), voltage (V, mV), and power output (P, mW) at a constant light level exposure using a halogen lamp inside an enclosed wooden box. The voltage and current generated by each dye-sensitized solar cell were measured using ANENG AN8002 Digital Multitester.

**Data Analysis Procedure :** The data that was collected in this experimental research was processed and analyzed through Statistical Package for Social Sciences (SPSS) version 21. The following statistical tools were used.

- Mean was used to describe the photoelectrical performance of the dye-sensitized solar cells with *Hibiscus acetosella* leaves as photosensitizer using different extracting solvents in terms of (1) voltage (mV), (2) current (mA), and (3) power (mW).
- One-Way Analysis of Variance (ANOVA) was used to determine the existence of significant differences among different photoelectrical parameters of the fabricated DSSC with *Hibiscus acetosella* leaves as photosensitizer using different extracting solvents.
- Tukey's HSD Test was used to further determine which specific group had an existing significant difference in terms of the photoelectrical parameters of the fabricated DSSC.

## 3. RESULTS AND DISCUSSION:

This study was conducted primarily to determine the effect of *Hibiscus acetosella* natural dye extract as a photosensitizer in Dye-Sensitized Solar Cells (DSSCs) in terms of voltage (mV), current (mA), and power (mW). It also looked into the optical absorption of the dye extracted from *Hibiscus acetosella* leaves using different extracting solvents.

**Optical Absorption of Natural Dye Extracted from *Hibiscus acetosella* leaves using different extracting solvents**

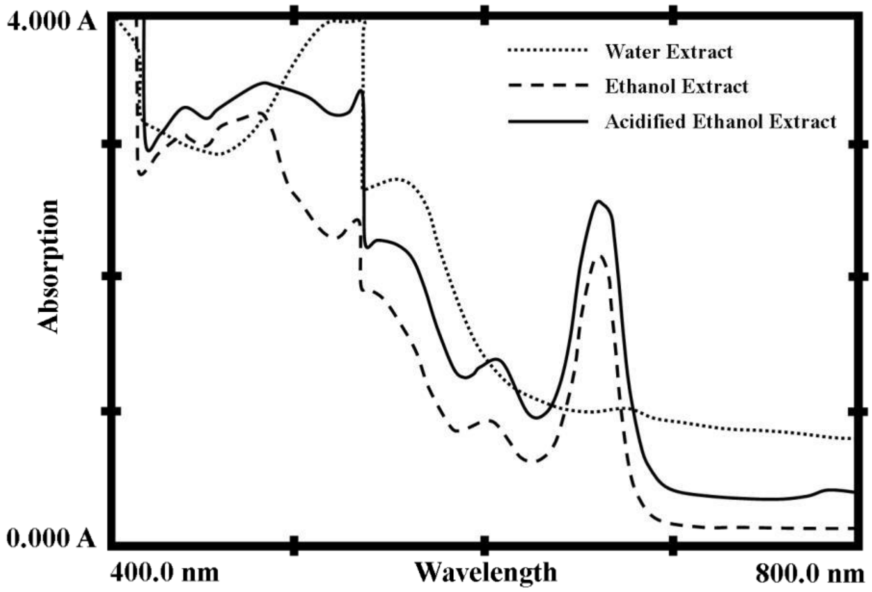


***Figure 2.*** The wavelength-absorbance graph of Hibiscus acetosella leaves dye extract using different extracting solvents

The UV-Vis absorption spectra of the natural dyes extracted from *Hibiscus acetosella* leaves using different extracting solvents – water, ethanol, and acidified ethanol – are shown below. As can be seen, the common maximum optical absorption among three different extracts was found to be at approximately 540 nm. It can be observed also from the wavelength-absorbance graph that there is a wide band of maximum absorption that is found between 400nm to 540 nm. These results concur with the reported absorption peaks of some natural pigments used as photosensitizers for DSSCs which includes anthocyanin with a peak absorbance at 520-540 nm (Merzlyak, Chivkunova, Solovchenko, & Naqvi, 2008; Woodall & Stewart, 1998). Aside from anthocyanin, this range of wavelengths of maximum optical absorbance also confirms the presence of betacyanin with a peak absorbance at 535-540 nm and betaxanthin with a peak absorbance at 463-494 nm (Cabiles, Caburnay, & Abulencia, 2011). This UV-VIS spectrophotometry result confirms the presence of some plant pigments which are considered the key factors in improving the performance of dye-sensitized solar cells.

**Performance of DSSCs with natural dye extracted from *Hibiscus acetosella* leaves as photosensitizer using different extracting solvents**

The graph shows the performance of the fabricated dye-sensitized solar cells in terms of the voltage generated in millivolts. It presents that in terms of voltage, the dye-sensitized solar cell with water-extracted *Hibiscus acetosella* natural dye had the highest mean voltage ($\bar{x}$ =369.2 mV) followed by the DSSC with ethanol-extracted *Hibiscus acetosella* natural dye ($\bar{x}$ =288.3 mV) and DSSC with acidified ethanol-extracted *Hibiscus acetosella* natural dye ($\bar{x}$ =214.61 mV). The DSSC with no dye had the lowest voltage generated with a mean of $\bar{x}$ =184.67 mV.

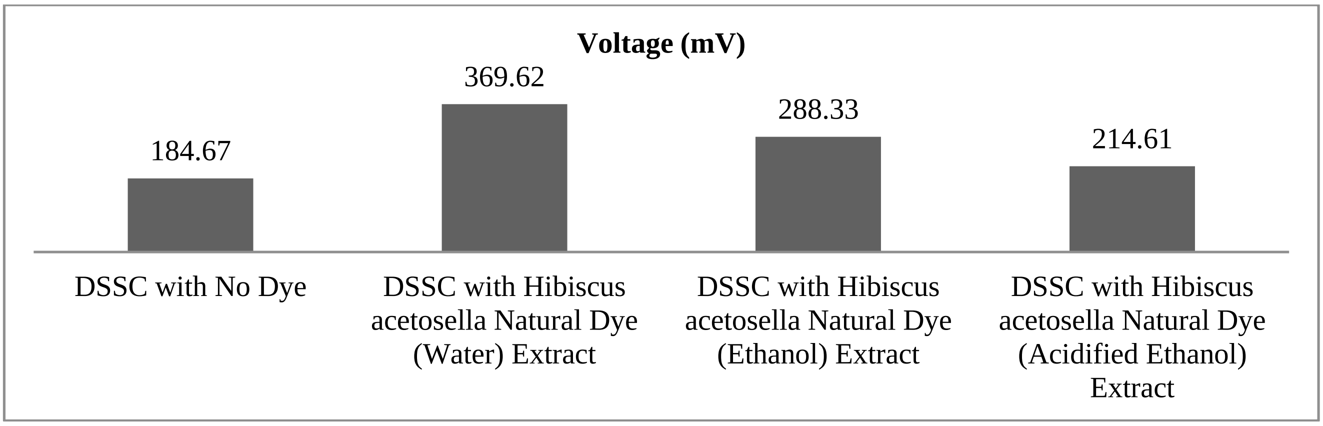


***Figure 3.*** Voltage (mV) of Dye-Sensitized Solar Cell with Hibiscus acetosella leaves extract using different extracting solvents

These derived values for voltage in the fabricated dye-sensitized solar cells were found to be higher when compared to other plant pigments used like in the work of Sullano and Sia (2018) where they used *mayana*, Malabar spinach, and nut grass leaves as a photosensitizer and had a range of voltage values of 6.2 mV to 108.1 mV and in the work of Ayalew and Ayele (2016) where the natural dyes were extracted from *Salvia spelendens* flower, *Jacaranda mimosifolia* flower, and lemon leaves and generated only 214 mV, 221 mV, and 225 mV, respectively. The highest generated value for voltage in DSSC with water-extracted *Hibiscus acetosella* leaves dye is also higher than in the work of Marco, Caramori, Argazzi, and Carlo (2010) where they used bougainvillea flowers as a source of dye and generated an average voltage value of 300 mV.

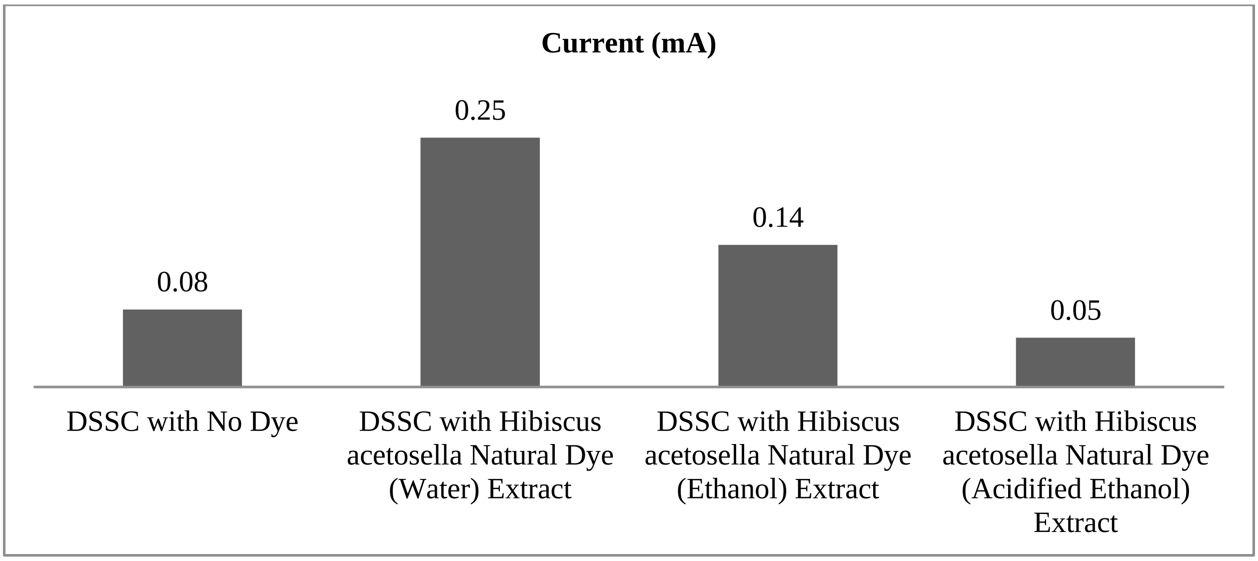


***Figure 4.*** Current (mA) of Dye-Sensitized Solar Cell with Hibiscus acetosella leaves extract using different extracting solvents

In terms of current, the dye-sensitized solar cells (DSSCs) with water-extracted *Hibiscus acetosella* leaves natural dye had the highest mean current value of $\bar{x}$ =83.54 mA. This was followed by the DSCCs with ethanol-extracted *Hibiscus acetosella* leaves natural dye ($\bar{x}$ =42.29 mA) and dye-sensitized solar with no dye at all ($\bar{x}$ =15.56 mA). The DSSCs with acidified ethanol-extracted *Hibiscus acetosella* leaves natural dye had the lowest current recorded ($\bar{x}$ =15.56 mA). When compared to other research, the recorded current from this present research is also higher than another natural dye extract just like in the work of Ayalew and Ayele (2016) where the natural dyes were extracted from *Salvia spelendens* flower, *Jacaranda mimosifolia* flower, and lemon leaves and generated only 0.071 mA, 0.034 mA, and 0.017 mA, respectively. Also, in the work of Sullano and Sia (2018) where they used mayana, Malabar spinach, and nut grass leaves as a photosensitizer, the generated current had a range of 0.004 to 0.015 by which the result of the use of *Hibiscus acetosella* leaves natural dye extract had a higher current recorded. In terms of electrical power (mW), again the DSSCs with water-extracted *Hibiscus acetosella* natural dye had the highest generated electrical power ($\bar{x}$ =85.54 mW). It was followed by the DSSCs with ethanol-extracted *Hibiscus acetosella* natural dye ($\bar{x}$ =42.29 mW), then by the DSSCs with no dye used ($\bar{x}$ =15.56 mW), and lastly by the DSSCs with acidified ethanol-extracted *Hibiscus acetosella* natural dye ($\bar{x}$ =10.48 mW). This result in current with the use of water-extracted *Hibiscus acetosella* natural dye extract was found to be higher than the natural dye extracts that were mentioned previously in voltage and current photoelectrical performance of the DSSCs.

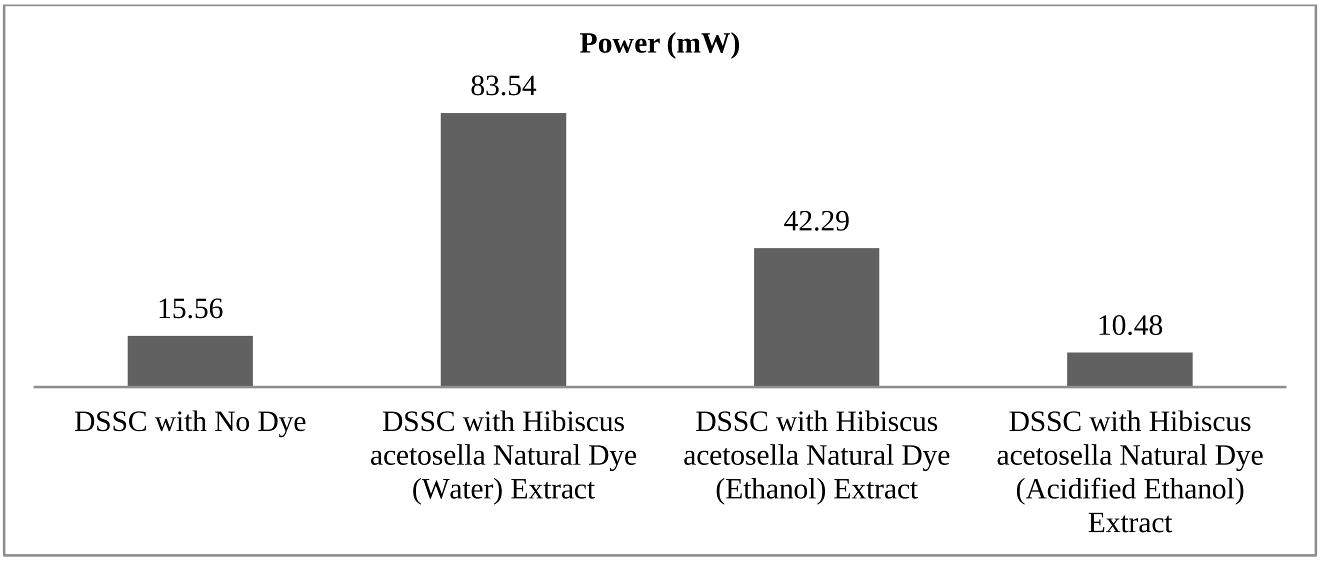


***Figure 4.*** Electric Power (mW) of Dye-Sensitized Solar Cell with Hibiscus acetosella leaves extract using different extracting solvents

**Significant difference in the performance of DSSCs with natural dye extracted from *Hibiscus acetosella* leaves as photosensitizer using different extracting solvents**

Based on table 1, shows that in terms of voltage, the p-value generated is p = .000 which is lesser than the standard α-value (α = 0.05), thus the null hypothesis which states there is no significant difference in the performance of the dye-sensitized solar cells with natural dye extracted from *Hibiscus acetosella* leaves as photosensitizer using different extracting solvents in terms of its voltage (mV) is rejected. Furthermore, the posthoc analysis shows that a significant difference in terms of voltage is found between the negative control (DSSCs with no dye) and DSSC with water-extracted and ethanol-extracted *Hibiscus acetosella* leaves dye but no significant difference is found in the voltage of DSSCs with no dye and DSSC with acidified ethanol-extracted *Hibiscus acetosella* leaves dye. Thus, the use of *Hibiscus acetosella* leaves dye extract as a photosensitizer for DSSCs is effective in terms of voltage when the extracting solvents to be used are water and ethanol. In terms of electric current, table 1 shows that the p-value is p = .002 which is again lesser than the prescribed α-value (α = 0.05), thus the null hypothesis stating that there is no significant difference in the performance of the dye-sensitized solar cells with natural dye extracted from *Hibiscus acetosella* leaves as a photosensitizer using different extracting solvents in terms of its current (mA) is rejected. Furthermore, the posthoc analysis (Tukey's HSD Test) shows that significant difference in terms of electric current is found between the negative control (DSSCs with no dye) and DSSC with water-extracted *Hibiscus acetosella* leaves dye but no significant difference is found in the electric current of DSSCs with no dye and DSSC with ethanol-extracted and acidified ethanol-extracted *Hibiscus acetosella* leaves dye. Thus, the use of *Hibiscus acetosella* leaves dye extract as a photosensitizer for DSSCs is effective in terms of electric current when the extracting solvent to be used is water.

Table 1. ANOVA of the different photoelectrical parameters (voltage, current, power) of the DSSCs with *Hibiscus acetosella* leaves dye extract in different extracting solvents

| | | Sum of Squares | df | Mean Square | F | Sig. |
|---|---|---|---|---|---|---|
| Voltage (mV) | Between Groups | 184327.423 | 3 | 61442.474 | 10.703 | .000 |
| | Within Groups | 183696.604 | 32 | 5740.519 | | |
| | Total | 368024.028 | 35 | | | |
| Current (mA) | Between Groups | .220 | 3 | .073 | 6.277 | .002 |
| | Within Groups | .374 | 32 | .012 | | |
| | Total | .593 | 35 | | | |
| Power (mW) | Between Groups | 30181.646 | 3 | 10060.549 | 10.375 | .000 |
| | Within Groups | 31031.529 | 32 | 969.735 | | |
| | Total | 61213.174 | 35 | | | |

Lastly, in terms of electric power, table 1 shows that the p-value is p = .000 which is lesser than the standard α-value (α = 0.05), thus the null hypothesis which states there is no significant difference in the performance of the dye-sensitized solar cells with natural dye extracted from *Hibiscus acetosella* leaves as photosensitizer using different extracting solvents in terms of its power (mW) is rejected. Furthermore, the posthoc analysis (Tukey's HSD Test) shows that significant difference in terms of electric power is found between the negative control (DSSCs with no dye) and DSSC with water-extracted *Hibiscus acetosella* leaves dye but no significant difference is found in the electric power of DSSCs with no dye and DSSC with ethanol-extracted and acidified ethanol-extracted *Hibiscus acetosella* leaves dye. Thus, the use of *Hibiscus acetosella* leaves dye extract as a photosensitizer for DSSCs is effective in terms of electric power when the extracting solvent to be used is water. Overall, the result of the experiments conducted showed the potential of *Hibiscus acetosella* leaves dye extract with water as extracting solvent in fabricating dye-sensitized solar cells. This is in consonance with the work of Yin (2016) who highlighted that dye sensitizer is an important factor in the performance of dye sensitized solar cells (DSSC). Moreover, the effectiveness in terms of voltage, current, and power of the DSSC with *Hibiscus acetosella* leaves dye extract with water as extracting solvent over ethanol and acidified ethanol is congruent to the work of Mohammed, Kasim, Yabagi, and Taufiq (2015) which showed that the performance of DSSC with *Hyphaene thebaica* as photosensitizer was better when the extracting solvent was water than when the used solvent is ethanol. The same observations were found in the work of Aduloju, Shitta, and Simiyu (2010) with their research noting that ethanol could be considered not suitable as a solvent for the extraction of natural dye used as a sensitizer.

## 4. CONCLUSIONS AND RECOMMENDATIONS:

This study was conducted to determine the performance of dye-sensitized solar cells (DSSC) with *Hibiscus acetosella* leaves dye extract in different extracting solvents as a photosensitizer. The following are the conclusions derived after the conduct of the experiments:

- The extracted natural dye from *Hibiscus acetosella leaves* using water, ethanol, and acidified ethanol as extract solvents has a wide band of maximum absorption with wavelengths ranging from 400 nm to 540 nm. This confirms the occurrence of plant pigments anthocyanin, betacyanin, and betaxanthin, among others.
- The dye-sensitized solar cell with water-extracted *Hibiscus acetosella* leaves dye performed the best in terms of voltage, current, and power.
- A significant difference is present between the photoelectrical performance (V, I, P) of the dye-sensitized solar cell with water-extracted *Hibiscus acetosella* leaves dye and the solar cell with no dye used. Thus, the use of *Hibiscus acetosella* leaves dye extracted using water as extracting solvent as a photosensitizer is effective in improving the photoelectrical performance (V, I, P) of the dye-sensitized solar cell fabricated.
- Inconsistent differences or none at all, are observed between the photoelectrical performance (V, I, P) of the dye-sensitized solar cell with ethanol-extracted and acidified ethanol-extracted *Hibiscus acetosella* leaves dye and the solar cell with no dye used. Thus, the use of ethanol and acidified ethanol as a solvent in extracting *Hibiscus acetosella* leaves dye as a photosensitizer for dye-sensitized solar cells is not reliable in improving the photoelectrical performance of the dye-sensitized solar cell fabricated.
- The positive result in using water-extracted *Hibiscus acetosella* leaves dye as a photosensitizer for dye-sensitized solar cells shows the practicability of the use of natural pigments as photosensitizers in DSSCs especially since these natural plant pigments are non-toxic, easily-disposed, cheaper, and environmentally-friendly which can contribute to a sustainable solution for the future energy production.

To further improve this study and harness the full potential of *Hibiscus acetosella* leaves dye extract as a photosensitizer for dye-sensitized solar cells, the following recommendations are given:

- Future researchers should look into the effect of sensitization time on the overall performance of dye-sensitized solar cells.
- Researchers should also look into the performance of DSSC through co-sensitizing the solar cells with *Hibiscus acetosella* leaves dye extract and other plant extracts which are locally available.
- Researchers can also look into the performance of DSSC with photosensitizers extracted from other parts of *Hibiscus acetosella* plant like its stem and flower.

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