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A Natural Dye-Sensitized From Pare (*Bitter Gourd*) Leaves Extracts For Dye-Sensitized Solar Cell (DSSC)

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Abstract: The availability of energy sources is dwindling so a renewable energy which has a potential chance to be developed, such as solar panels, is needed. The use of solar panels is still quite expensive in terms of manufacturing process. For this reason, a cheap solar panel based is developed, and it is called DSSC (Dye-Sensitized Solar Cell). The use of DSSC is developed in Indonesia, a country which is famous for its biological richness. In this study, pare leaves were used as photosensitizers obtained from the extraction process of maceration by various solvents. The DSSC test was done on four extracts including N-hexane extract, Ethyl Acetate, Methanol, and Combination of the three extracts. The highest value of efficiency obtained from each extract respectively are 0,03%, 0,04%, 0,14% and 0,30%. Characterization was done by examining the UV-Vis and FTIR spectral data. The result of UV-Vis analysis shows that wavelength for N-hexane, Ethyl Acetate, Methanol, and Combined extracts are 269,1 nm, 668,0 nm, 663,9 nm, and 6631 nm, respectively. FTIR results found that the chromophore and auxochrome groups were identified on all four tested extracts, namely; C = C, C = O, -OH, and -C-H groups.

Keywords: auxochrome, chromophore, DSSC, efficiency, Pare leaves

1. INTRODUCTION

The exploitation of renewable energy sources in Indonesia needs to be maximized. Indonesia has rich unlimited energy sources in its nature, such as wind, water, and the sun which has very potential chance to be developed. Solar energy can be exploited by converting it into electrical energy called solar cells (Puspitosari, Sumarno, and Budi, 2006). Solar cells are generally silicone-based which are relatively expensive. For this reason, new cheaper materials are needed instead of silicone, such as a dye. Dye-based solar cells are a type of solar cell firstly introduced by Oregan and Grätzel in 1991. Dye-based solar cells, known as DSSC (Dye-Sensitized Solar Cells), use dyes as a sensitizer to light and semiconductors as electron carriers (Gratzel, 2003). These solar cells are the third generation of solar cells. The advantages of using this solar cell type are the use of safe materials, abundant basic materials, and scalable production processes, so it produces good and high productivity data (Radwan, 2015). One of the important roles in dye-sensitized cells is how the ability of dyes to absorb the energy of sunlight that will be converted it to electrical energy (Ridwan et al., 2018).

The basic principle of DSSC is the occurrence of electron excitation from the valence of dye band to the conduction band of the semiconductor. The excitation of these electrons results from the interaction of HOMO or LUMO between titanium oxide with carboxyl groups in dyestuffs (Trianiza and Gatut, 2012). Electron excitation of the organic components in the form of the dye is injected by electrons to the oxide conduction band (Gratzel and O'Regan, 1991) in which the dye will be positively charged while the semiconductor TiO₂ is negatively charged by the process (Trianiza and Gatut, 2012). One of the DSSC parameters such as short-circuit current density and maximum power intensity decrease significantly after irradiation (Liu Min, 2016).

The advantage of TiO₂ used as a semiconductor is that it has high structural stability under sunlight, non-toxic and low cost (Radwan, 2015). The use of porous TiO₂ can increase the absorption of dye on the surface (Ren et al., 2011). TiO₂ sold in the market is usually in the form of *Degussa P25* which consists of a mixture of *anatase* and *rutile* crystalline forms (Jiputti, 2008). A TiO₂ nanowire

(NW) is also used as semiconductors in the DSSC circuit (Li et al., 2015). One of important in the TiO_2 is a form of nanostructures in determining the value of efficiency (Bhogaita et al., 2016).

Electrolytes have an important role in the DSSC circuit. It provides electrons in the dye (Gratzel, 2003). Liquid electrolytes typically consist of a redox couple and an additive (Al-Wani et al., 2016). According to Zahrok and Gotjang (2015: 31) in their research, the use of fluid electrolyte can only last for 10 minutes. There are many types of electrolyte use in the research done by Subramania A, et al (2013: 1649), these are ethylene carbonate (EC) and propylene carbonate (PC) and 1-dedocyl-3-ethyl imidazolium iodide ionic crystals (C12Elml) and ionic liquids 1, decyl-3-ethyl imidazolium iodide (C10Elml) which have been synthesized (Pan Xu, et al., 2013). In component of DSSCs the counter cathode regenerates the electrolyte. The platinum is best catalyst material for counter cathode results high effecicency of the cell although high cost increases overall cost og the cell (Richhariya et al., 2017).

Besides the use of ITO glass components, TiO_2 semiconductors, and electrolytes, the dye has a very important role in this process. Many DSSCs have been developed based on natural dyes such as teak leaves which use the methanol fraction: n-hexane dye from teak leaves with efficiency by 0.05127% (Aminuddin et al., 2016). Meanwhile, *Biduri* leaves extract yield value of efficiency by 0,8596% (Suprianto, 2016) and extracted from petals of male flowers *Luffa cylindrica* L value of efficiency by 0,13% (Murya et al., 2016). One of the dyes that can be used as a photosensitizer is the dye of *Pare* (bitter gourd) leaves. *Pare* plants, commonly used as food sources and medicines, are also believed to be used as dye sensitizers given the presence of dyestuffs from tannins, polyphenols, and carotenoids (Mutiara and Ahmad Wildan 2014).

In this study, the preparation of the natural dye materials from *Pare* leaves as a photosensitizer is done and also has been characterized using UV-Vis and FTIR then preparing the DSSC and performing with the extracted of natural dyes.

2. RESEARCH METHOD

Materials

The materials used in this study are the samples of *pare* leaves (*Momordica charanti*) obtained from Bajeng District, Gowa. chemicals include aquadest (H_2O), sulfuric acid (H_2SO_4) pa, iron (III) chloride (FeCl_3), Ethanol ($\text{C}_2\text{H}_5\text{OH}$), ethyl acetate ($\text{C}_4\text{H}_8\text{O}_2$) (*Bratachem*), iodine (I_2), *pare* (*Momordica charantia*), potassium iodide (KI), ordinary filter paper, waxes, methanol (CH_3OH) (*Bratachem*) n-hexane (C_6H_{14}) (*Bratachem*), 10% sodium hydroxide (NaOH), Titanium Dioxide (TiO_2) catalog 7508, clear insulation, Dragendroff reagents, Mayer reagents, Wagner reagents, and Lieberman-Burchard reagents.

Experiment

1. Preparation of Natural dye as sensitizer and characterization

The sample of *Pare* (bitter gourd) leaves is firstly cut into small pieces and dried at room temperature and then blend to obtain *pare* leaves powder. The extraction is begun with an n-hexane solvent, followed by an ethyl acetate solvent and finally with a methanol solvent. The obtained filtrate is concentrated using a rotary vacuum evaporator at a temperature of 40-50 °C and 50 rpm to form a viscous extract.

2. Preparation of DSSC and performance with the extracted natural dye

TCO glass is cleaned with water in ultrasonic and dried. Next, the TCO glass is heated over a fire (wax) to form a black color on the TCO glass. Cleaned TCO glass covered with clear insulation on one side and coating using TiO_2 p.a then sintered for 30 minutes. TCO glass coated with TiO_2 paste is sprinkled with *pare* leaves extract and left for a while until absorbed into TiO_2 . The gel electrolyte liquid is added and closed by a comparative electrode. After the DSSC circuit is completed, it is connected to a multimeter tool and a light intensity meter. Finally, it is illuminated by sunlight and recorded the current and voltage generated by the DSSC circuit.

3. Characterization of TiO₂-dye pare leaves by SEM

3. RESULT AND DISCUSSION

1. Natural dye as sensitizer and characterization

The results on the experiment of combined extract, methanol, and ethyl acetate show that the presence of flavonoid content (phenolic compound) is indicated by the presence of light yellow due to the addition of 10% NaOH. For a positive result, the alkaloid compound is characterized by a change in yellow or white color with the addition of Mayer reagent and orange precipitate for the addition of *Dragendroff* reagent. This occurs in the combined extract, methanol, and ethyl acetate.

Table 1. Screening Fitokimia of dye pare leaves

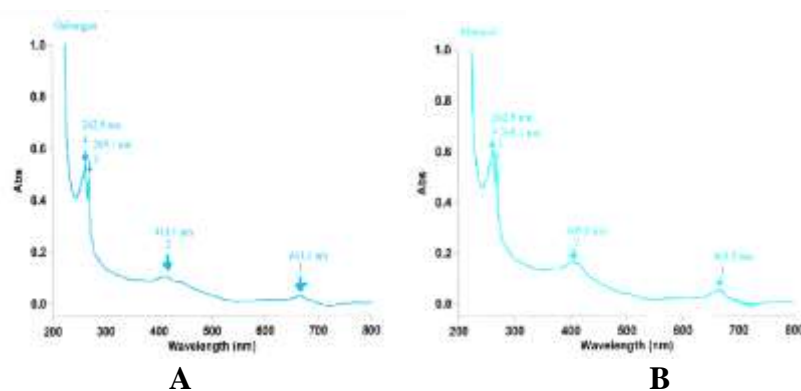
Secondary metabolic compound	Extract			
	Mixture	Methanol	Ethyl Acetate	Hexane
Terpene	-	-	-	-
Flavonoid	+	+	+	+
Alkaloid	+	+	+	-

Notes: + = Positive, - = Negative

Absorption wavelengths of dye pare leaves extracts

Tests on *Pare* (bitter gourd) leaves extracts on UV-Vis Spectrophotometer aim to know the absorption of the maximal wavelength which shows the absorption at Ultraviolet area (200-400 nm) and Visible or visible light (400-800 nm). Based on figure 1, the highest absorption for the combined extract, methanol, and ethyl acetate are 663.1 nm, 663.9 nm, and 668.0 nm. According to Harbone (1987), the main peak around 400 nm with a small peak in the 500 nm and one more peak above 625 nm, it indicates the presence of chlorophyll compounds. In the Combined extract, the peak shapeshifts to be shorter than methanol, due to the increase of the solvent polarity in the transition $n \pi^*$ so that the peak shapeshifts to a shorter wavelength (blue or hypochromic shift).

For n-hexane extracts indicated to contain flavonoids produce low-efficiency values because sunlight produces 5% of spectra under ultraviolet light and 45% invisible regions (Rofi'ah and Gotjang, 2013).



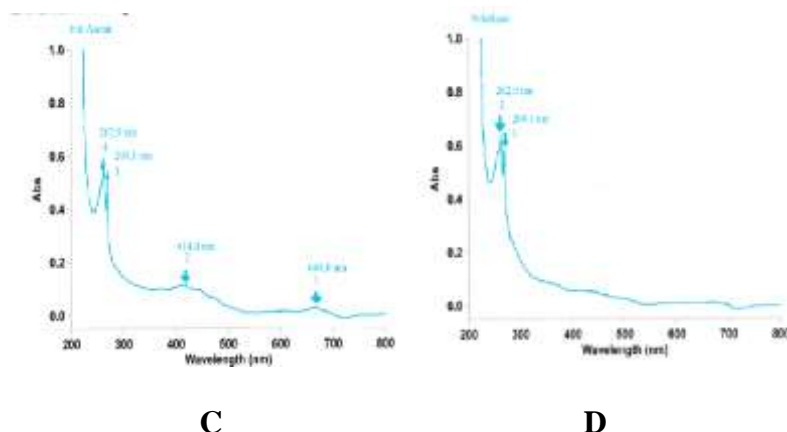
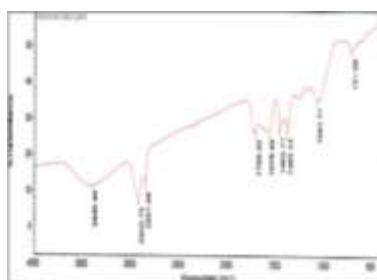


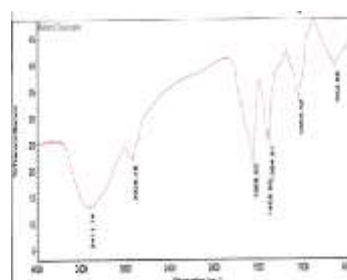
Figure 1. UV-Vis spectrum of a).dye mixture extract, b). dye methanol extract, c). dye ethyl acetate extract and d). dye n-hexane extract

FTIR Analysis of dye pare leaves extracts

FTIR is an instrument suitable to analyze functional groups in a compound. The FTIR spectrum has a wavelength of 40000-400 cm^{-1} (Silverstein, 2005). FTIR results in figure 2 show that the Combined extract, methanol, and ethyl acetate have a good efficiency average values because they are supported by clonal groups of chromospheres which absorb light in ultraviolet regions such as carbonyl and aromatic ($\text{C} = \text{C}$). Beside the chromosphere, there are also auxochrome groups such as OH and N-H causing the absorption of light previously placed in the ultraviolet region to become ultraviolet-visible in the presence of an auxochromic group to produce a good value of efficiency. As for the n-hexane extract, there is only a chlorophyll group that only absorbs light of the ultraviolet rays so that the efficiency value produced by the lower efficiency compared to the three extracts. It proves that the more polymorphic groups of chromophores and auxochromes contributed, the higher ability to absorb as much wavelength as possible and provide a strong bond with TiO_2 that will result in greater efficiency value.



a)



b)

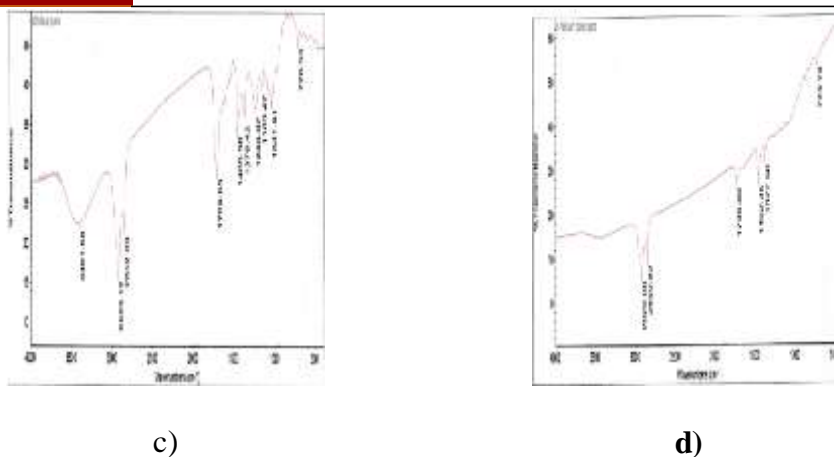


Figure 2. FTIR spectrum of a).dye mixture extract, b). dye methanol extract, c). dye ethyl acetate extract and d). dye n-hexane extract

2. DSSC and performance with the extracted natural dye

In table 2, the value of DSSC efficiency and n-hexane extract is 0.03%, ethyl acetate extract is 0.04%, methanol extract is 0.14%, and for the combined extract is 0.30%. The value is still relatively low compared to the value of DSSC efficiency in general by 2-3% (Grätzel, 2001). The increase of the value due to the indication of the compound contained in most pare leaves is polar compounds such as polyphenols, and other compounds such as chlorophyll (Mutiarra and Ahmad Wildan, 2014). It results in ease bonding process with TiO₂ as well as the more conjugation of the structure of the compound occurs and will absorb a greater wavelength.

Table 2. Measurement of DSSC Efficiency Value on Pare Leaves Extract

No.	Extract	Efficiency (η) (%)
1	Mixture extract	0.30
2	Methanol extract	0.14
3	Ethyl Acetate extract	0.04
4	N-Hexane extract	0.03

3. Morphology and structure of TiO₂-dye pare leaves

As the mixture extract gained the highest efficiency value among the other, its morphology was subsequently analyzed by SEM. The results of the morphological characteristics of TiO₂ of Pare leaves dye can be seen in Figure 3. It shows that the dispersion of Pare leaves on the surface of TiO₂ is uneven. It is clear that the dye is adsorbed uneven and forms some clots that indicate an unabsorbed dye. However, the presence of carbonyl groups (C = O) and hydroxyl groups (-OH)

indicates that in general there are strong bonds formed with TiO₂ molecules. This causes the dyestuff adsorbed well on TiO₂ resulting in a greater efficiency value.

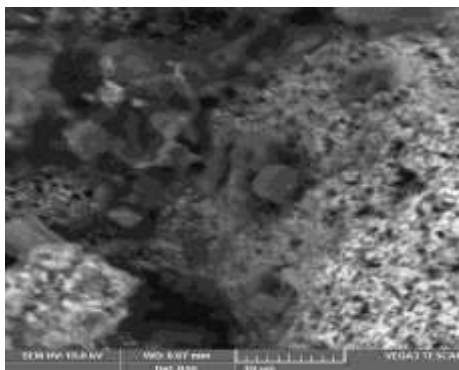
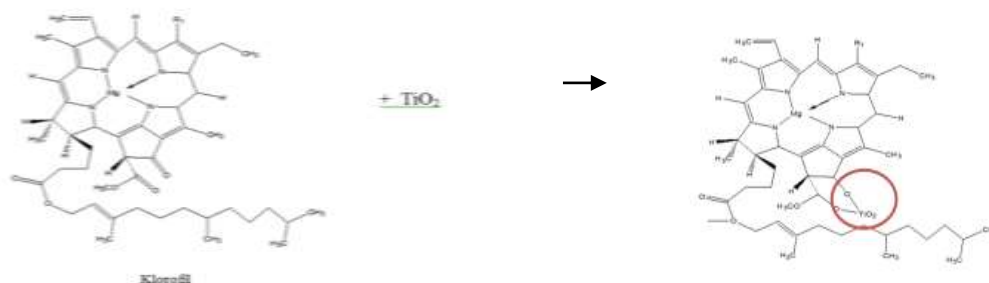


Figure 3. SEM image of TiO₂-dye pare leaves

Regarding its phytochemical test, this extract assumes to contain flavonoids and alkaloids compound which obviously have carbonyl, hydroxyl and amine groups. The functional groups of these compounds are responsible for the bindings to TiO₂. Chlorophyll is a dye that mostly contained in leaves and is a class of alkaloid compounds. The presence of adjacent carbonyl groups allows binding to TiO₂ as can be seen in Figure 4. To this end, the more bindings to TiO₂, the higher the DSSC efficiency value which records to this mixture extract.



Gambar 4. Structure of TiO₂-Klorofil pigment

4. CONCLUSION

The values of DSSC efficiency resulting from n-hexane, ethyl acetate, methanol, and *pare* leaves extracts are 0.03%, 0.04%, 0.14%, and 0.30%, respectively. The compound contained in the *pare* leaves extracts give a significant impact on the value of DSSC efficiency.

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