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Revolutionizing Dye Sensitized Solar Cells – Impact of Silicon Dioxide Purity Derived from Coal Fly Ash for Enhanced Photoelectric Performance

Mitha Pratiwi[1*], Leila Kalsum[2], Rusdianasari[2]

[1] Applied Master Program of Renewable Energy Engineering, Politeknik Negeri Sriwijaya, Jl. Srijaya Negara Bukit Besar, Palembang, 30139, Indonesia

[2] Renewable Energy Engineering Department, Politeknik Negeri Sriwijaya, Jl. Srijaya Negara Bukit Besar, Palembang, 30139, Indonesia

* Corresponding author's e-mail: mithapratiwi025@gmail.com

ABSTRACT

Dye-sensitized solar cells (DSSCs) provide a promising alternative to traditional solar technologies, as they offer a unique mix of cost-effectiveness, adaptability, and the possibility of achieving high efficiency. This study aims to investigate the effect of SiO_2 purity obtained from coal fly ash on the photoelectric performance of DSSCs. The study focuses on varying the purity of extracted SiO_2 from coal fly ash as a counter-electrode material and examining its impact on the efficiency of DSSCs. The efficiency of DSSCs was assessed by evaluating the performance of several various SiO_2 purity materials for counter-electrode materials. The performance testing of DSSCs revealed that the counter electrode material, consisting of artificial SiO_2 with a purity of 99.9%, achieved the highest efficiency of 0.0113%. Subsequently, DSSCs were fabricated using counter-electrode materials derived from coal fly ash with a purity of 52.91%. These DSSCs demonstrated an efficiency of 0.0076%. DSSCs utilizing SiO_2 with a purity of 91.20% exhibited an efficiency of 0.0062%. Dye-sensitized solar cells utilizing a counter electrode material composed of SiO_2 with a purity level of 72.54% demonstrated an efficiency of 0.0061%. The results showed that the level of SiO_2 purity obtained from coal fly ash has a substantial impact on the photoelectric performance of DSSCs, since higher purity of SiO_2 is associated with improved efficiency.

Keywords: DSSCs, silicon dioxide, coal fly ash, counter electrode.

INTRODUCTION

During a global economic slowdown, the Indonesian economy grew by 5.05% in 2023. In line with the economic growth of Indonesia, the energy production in Indonesia increased 1.55 percent (y/y) compared to last year and reach a value of 1853 million BOE by the end of 2023 [Ministry of Energy and Mineral Resource., 2023]. This is the highest energy supply since six years ago. Similar to the energy supply, consumption in 2023 saw its highest volumes in six years. In 2023, the industrial sector had the highest energy consumption, with a global share of 45.60%. The transportation sector at 36.74%, households at 12.35%, commercial sectors at 4.44%, and other sectors at 0.87%. Coal and natural gas consumption dominated the industrial sector's energy demand in 2023. By 2023, coal would have taken up the largest proportion of energy consumption in the industrial sector, reaching 56.90%, followed by 21.41% of gas, while electricity accounted for 12.7% [Ministry of Energy and Mineral Resource., 2023].

Energy diversification and conservation are critical approaches to reducing dependence on fossil fuels and ensuring the nation's energy security [Bagaskara et al., 2024]. According to [Akrofi, 2021], diversifying energy sources can enhance energy security and reduce reliance on fossil fuels by incorporating multiple energy types, including renewables. This diversification mitigates the risk associated with supply disruptions and price volatility inherent in fossil fuel markets.

Indonesia has a lot of potential for renewable energy, which presents an opportunity to reduce dependence on fossil fuels and meet the nation's growing energy needs. Indonesia's abundant natural resources enable the development of a variety of renewable energy sources, including hydro, geothermal, bioenergy, solar, wind, and micro-hydro power. Specifically, projections indicate that Indonesia has 419 GW of potential renewable energy, with significant contributions from solar at 207.8 GW, hydro at 75 GW, wind at 60.6 GW, geothermal at 23.7 GW, and bioenergy at 32.6 GW [Pambudi et al., 2023].

Indonesia is an equatorial nation with a tropical environment and an abundance of solar thermal energy because the sun shines there all year round. It transforms into an energy source with potential for development. The solar cell is one instance of how solar energy is used to create electrical energy that is more widely recognized and frequently mentioned in public [Laksana et al., 2021].

Over the last ten years, solar cells or photovoltaic cells have grown as a significant player in the continuing energy transition [Pastuszak and Wegierek, 2022]. Solar cell technology has now developed to the 4th generation [Kant and Singh, 2022]. First generation of pv technology involves silicon based solar cells [Singh et al., 2021]. Thin-film solar cells are introduced in the second generation of PV technology [Pastuszak and Wegierek, 2022]. There are several innovative strategies in third-generation PV technologies, such as organic photovoltaics (OPVs), perovskite solar cells, and dye-sensitized solar cells (DSSCs) [Shah et al., 2023]. Introducing the fourth generation of PV cells, commonly referred to as hybrid in organic cells [Rehman et al., 2023]. Dye-Sensitized Solar Cells (DSSCs) are a notable development in the field of third-generation PV technologies [Ronado et al., 2024]. In 1991, Michael Gratzel and Brian O'Regan created DSSCs, which imitate the process of photosynthesis by utilizing a dye to capture sunlight and produce electricity. DSSCs consist of a porous layer of titanium dioxide (TiO_2) nanoparticles coated with a photosensitive dye, along with an electrolyte and a counter electrode [Sharma et al., 2018].

The problem with DSSC is that the resulting efficiency is relatively low when compared to silicon-based solar cells [Trihutomo et al., 2019]. As a result, numerous researchers are still working on DSSC-related projects, one of which is examining the counter electrode, one of the system's components. The counter electrode has a vital role in increasing the efficiency of DSSC. The counter electrode is essential to raising the DSSC's efficiency. Without the counter electrode, the DSSC cannot increase its efficiency. In the electrolyte of a dye-sensitized solar cell (DSSC), the counter electrode is crucial for speeding up oxidation and starting the redox process [Mithari et al., 2023]. This function is critical to the cell's continued operation because it makes it easier for electrons to move back into the electrolyte from the external circuit, completing the circuit and allowing the photovoltaic process to proceed [Soonmin et al., 2022]. Therefore, the selection of the type of material used as a counter electrode is very important because the material used must have high electrocatalytic activity, high conductivity, large surface area, high stability [Mithari et al., 2023; Ding et al., 2023] and of course have an affordable price.

Platinum (Pt) is widely known for having outstanding electrocatalytic activity and electrical conductivity [Mithari et al., 2023]. DSSCs commonly use platinum (Pt) as a counter electrode [Kumar et al., 2019]. However, the DSSCs frequently use the iodide-triiodide electrolyte, which presents difficulties due to its increased cost, restricted supply, and corrosion susceptibility [Kumar et al., 2019]. Subsequently, it is imperative to investigate platinum (Pt) substitutes. In order to satisfy the counter electrode requirement, it is critical to consider materials that are easy to synthesize, eco-friendly, and have an efficiency level comparable to platinum (Pt).

Coal fly ash is a solid waste produced from coal combustion in power plants for electricity production. Coal-fired power plant fly ash is a significant environmental concern due to its small particle size, heavy metal content, and increase emissions [Minh et al., 2023]. Fly ash contains various types of metal oxides, one of which is silica, which is the main mineral that makes up coal fly ash [Bhatt et al., 2019]. Silica is one of the by-products that will always be produced from the coal combustion process [Bhatt et al., 2019]. The high silica content in coal fly ash can be used as a support for TiO_2 photocatalysts, which aims to increase the effectiveness of TiO_2 photocatalysis [Jiang et al., 2020]. The presence of silica can enhance its process by providing a conducive environment for the electron hole pairs [Showman et al., 2024]. In photocatalysis, photoreduction and photooxidation processes will occur. Photocatalysis is a chemical reaction that occurs with the assistance of a catalyst, and the catalyst is active when irradiated with sunlight [Mohamadpour and Amani, 2024]. This study evaluated four different purity levels of silicon dioxide from coal fly ash as counter electrode materials in DSSC using TiO_2, red spinach dye, and SiO_2 configuration. The study examined the impact of SiO_2 purity on DSSC performance. It was observed in the efficiency of DSSCs.

METHODOLOGY

Dye extraction

The dye extraction was prepared following the procedure described in [Prabavathy et al., 2017]. The steps required to extract the red spinach are presented in Figure 1. Natural dyes were extracted from red spinach (*Amarantush tricolor* L.). 100 g of fresh red spinach were washed with distilled water to remove any dirt on the surface. Subsequently, all water molecules were eliminated from the red spinach by letting it air dry entirely. After drying, it was torn into fragments by hand to avoid the loss of dye when scissoring them. 20 g of leaf fragments were taken in airtight containers and added to 6 to 10 ml of citric acid (1 g/100 ml). The containers were stored in the dark for 24 hours. After 24 hours, the contents in the container were filtered, stored in an amber bottle, and refrigerated at 4 °C until use.

Preparation of TiO_2 working electrode

The TiO_2 paste was prepared following the procedure described in [Nadhirah et al., 2020]. 0.5 g of polyvinyl alcohol (PVA) was added to 2.5 ml of distilled water. The mixture is then stirred with a magnetic stirrer at a temperature of 80 °C for approximately 30 minutes until the solution thickens and is homogeneous. The solution that has been made was added to 0.5 grams of TiO_2 powder with 15 drops of PVA solution to form a TiO_2 paste. Four pieces of FTO glasses (fluorine-doped tin oxide) had been used as the substrates. The substrate's dimensions of 2.5 × 2.5 cm are characterized by 6 Ω/sq sheet resistance. The working electrode was prepared following the procedure described in [Choi et al., 2022]. The FTO glass was sonicated using an ultrasonic cleaner for 30 minutes with ethanol 96%, acetone, and distilled water. A thin TiO_2 layer was cast using the doctor-blade method in a rectangular shape of 5.0 cm^2. The film thickness was adjusted with a sticky tape. In order to avoid cracking during calcination, the substrate was warmed at 80 °C for 1 hour. Then, the TiO_2 layer was calcined at 450 °C for 30 min. The TiO_2 film layer was sensitized for 3 hours in a dark environment using a red spinach solution in citric acid.

Preparation of electrolyte

A solution of iodine electrolyte was made using the methodology described in the studies by [Kabir

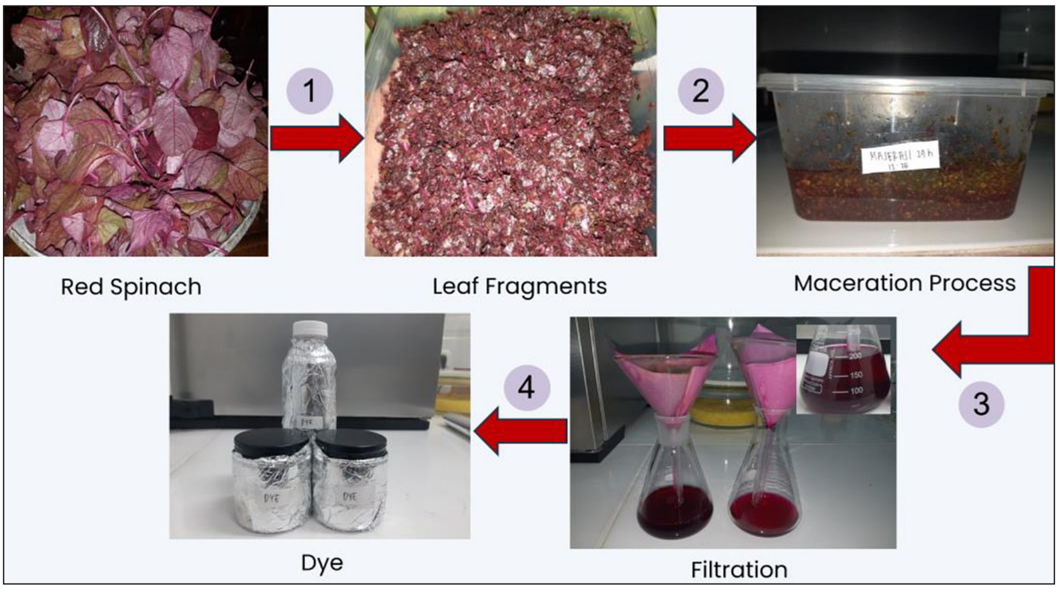

Figure 1. Red spinach extraction process

et al. (2019); Khan et al., 2023]. To prepare the electrolyte, 10 ml of ethylene glycol was placed in a 50 ml beaker, followed by the addition of 0.127 g of iodine. Subsequently, 0.83 grams of potassium iodide were introduced into the beaker. The beaker's components were agitated and gently heated using a hot plate until a transparent solution was achieved. Upon formation, the solution was placed in an opaque container, which was then covered with aluminum foil and stored in a lightless environment.

Preparation of counter electrode

The coal fly ash powder used in this study originated from PT Semen Baturaja in Ogan Komering Ulu Regency, South Sumatera (Table 1). Exactly 100 grams of coal fly ash were added to a beaker glass containing 500 ml of HCl 1 M. The mixture was mechanically agitated at 150 rpm and preheated to a temperature of 90 °C for 4 hours. The mixture was filtered after 4 hours of acidification, and the residue was repeatedly washed with hot distilled water before being dried for 12 hours at 110 °C. In order to extract SiO_2, 150 ml of NaOH 3 M was added to the dry coal fly ash, and the mixture was stirred magnetically at 150 rpm for 4 hours at 90°C (Figure 2). It was filtered after 4 hours to get a translucent sodium silicate filtrate. Sodium silicate was progressively neutralized with diluted HCl 1 M to pH 7 in order to precipitate silica. The precipitated silica was collected, cleaned several times with hot distilled water, and dried for 12 hours at 110 °C [Caroles, 2019]. Subsequently, the precipitated SiO_2 was purified by adding it to a beaker glass that had 20 ml of 37% HCl. The mixture was then maintained at 90 °C for 3 hours while being stirred magnetically at 150 rpm. After that, the solid was rinsed with hot distilled water and dried in an oven set to 110 °C for 3 hours [Andarini et al., 2018]. Coal fly ash and SiO_2 that has been made was added to 1.0 grams of powders with 15 drops of PVA solution to form a paste. Four pieces of FTO glasses (fluorine-doped tin oxide) had been used as the substrates. A coal fly ash and SiO_2 paste was cast using the doctor-blade process in a rectangular shape of 5.0 cm^2. A sticky tape was used to adjust the film thickness. The substrate was warmed for 1 hour at 80 °C.

Assembly of DSSC

The working electrode and counter electrode were assembled in a sandwich configuration, and a liquid electrolyte solution (prepared as described in the section on electrolyte preparation) was injected between the two cell sides using a pipette. Two binder clips were used to hold the two cell sides together.

Performance test of DSSC

DSSC performance testing was carried out by measuring the DSSC voltage and current directly under sunlight from 09.00 to 15.00 WIB for 8 days from March 2, 2024, to March 9, 2024. The intensity of sunlight was measured using a solar power meter to ensure accurate testing conditions.

RESULTS AND DISCUSSION

Absorbance spectra analysis

In DSSC, dyes act as photosensitizers, absorbing photons from sunlight. Anthocyanins are chemical compounds contained in red spinach that can provide different colors at various pH levels.

Table 1. Properties of coal fly ash and extracted SiO_2

No.	Component	Percentage (%weight)			
		Coal fly ash (CFA)	Extracted SiO_2 before purification	Extracted SiO_2 after purification	SiO_2 artificial
1	SiO_2	52.91	72.54	91.20	99.90
2	Al_2O_3	18.21	13.51	1.03	-
3	Fe_2O_3	5.02	0.02	0.02	0.005
4	CaO	7.52	3.29	0.29	-
5	MgO	0.51	0.13	0.03	-
6	Na_2O	3.64	3.66	4.66	-
7	K_2O	0.33	0.19	0.19	-
8	TiO_2	0.88	0.33	0.33	-
9	SO_3	0.45	-	-	-

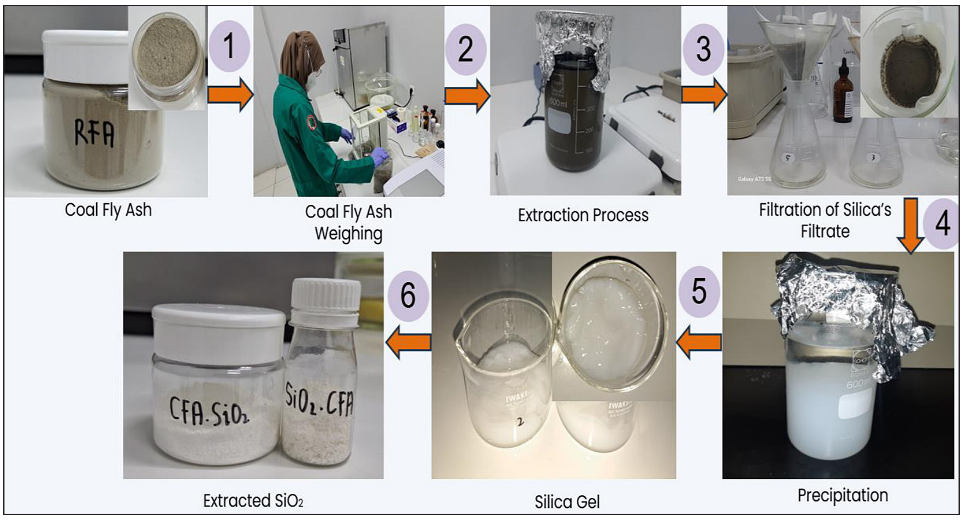

Figure 2. The extraction process of SiO_2 from coal fly ash

Anthocyanins turn red in acidic conditions and blue in alkaline conditions [Pramananda et al., 2021]. The maceration method isolates the dyes in red spinach under acidic conditions. During the maceration process, acidic conditions are used to create flavylium salts that are protonated. These salts will help keep the anthocyanins stable. In addition, in acidic conditions, more anthocyanin pigments are produced [Prabavathy et al., 2017].

To determine the absorption capacity of red spinach dye, UV-VIS spectroscopy testing was carried out using a Shimadzu UV-1780 UV-VIS spectrophotometer instrument. Measurement of light absorption was carried out in the ultraviolet to visible light region (270–700 nm). Anthocyanins have a maximum wavelength in the UV region of 270–290 nm and 465–560 nm for the visible region [Adam, 2015]. The results of measuring the red spinach extract are displayed in Figure 3. Spectroscopy testing was carried out in three stages, namely the first measurement of the UV-to-VIS wavelength at a wavelength of 270–700 nm. From the measurements carried out, three wave peaks were obtained in the VIS and UV regions. In the UV light region, the wavelengths obtained were 289.20 nm with an absorbance of 0.6316 and 322.30 nm with an absorbance of 0.6362. In the visible light region, the obtained wavelength was 535.10 nm with an absorbance of 0.1782. Referring to the literature [Adam, 2015], which states that anthocyanin compounds have maximum wavelengths in the 270–290 nm and 465–560 nm regions, spectroscopy tests

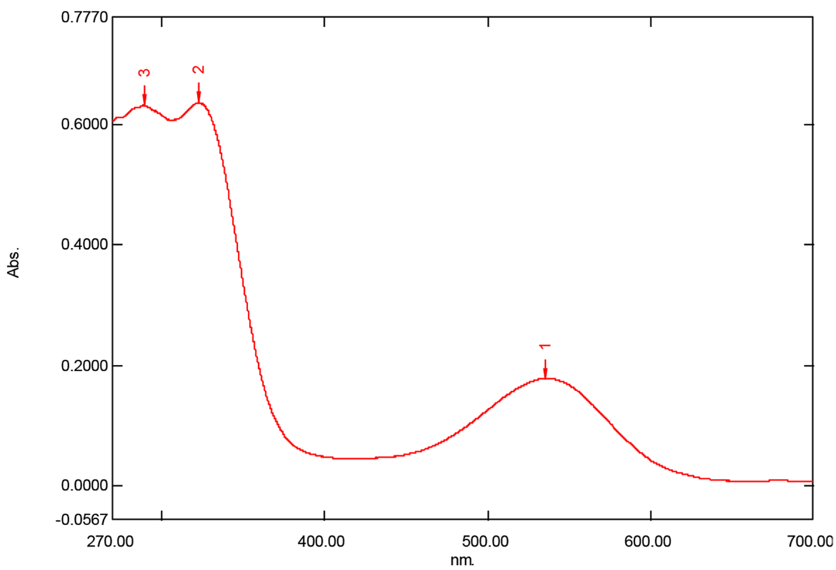

Figure 3. The absorbance spectrum of red spinach dye

were carried out separately for each light region. Figure 4 shows the spectroscopic measurements in each light region of anthocyanin. In the UV light region (270–290 nm), the maximum wavelength is obtained at 288.90 nm, and the maximum wavelength in the visible light region is obtained at 537.70 nm. When compared with previous literature [Adam, 2015], the results obtained are not much different; there is a shift of 1 nm for each light region, which can be influenced by various factors such as storage temperature and lighting in the dye storage room, which can affect the shift in the maximum wavelength of the dye.

Photoelectric properties of DSSC

Photoelectric measurements on DSSC (dye-sensitized solar cells) involve several key parameters that need to be measured to evaluate the performance of the solar cells, namely the I-V characteristics and efficiency of the DSSC. The current-voltage of the fabricated DSSC was measured directly under the sunlight (Figure 5). Each material tested in DSSC showed variations in voltage and current values during the observation period (Figure 6), with several peaks and dips seen on various dates. In general, the voltage generated is directly proportional to the current value produced. The higher the voltage value, the higher the current produced.

Current-Voltage (I-V) characteristics

Figure 6 shows the I–V measurement value from four different purity levels of SiO_2. The DSSC with SiO_2 artificial (purity 99.90%) produced the highest voltage and current was 602.74 mV and 60.78 μA as shown in Figure 6d. This

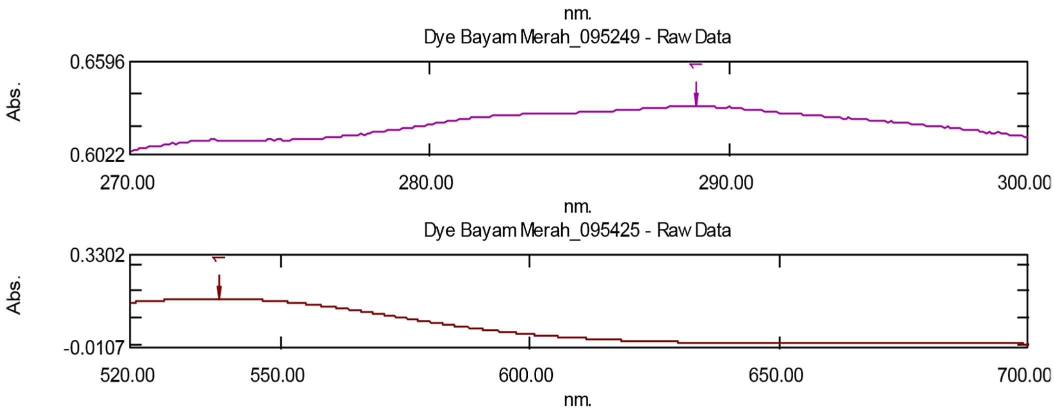

Figure 4. The absorbance spectrum of anthocyanin in red spinach dye

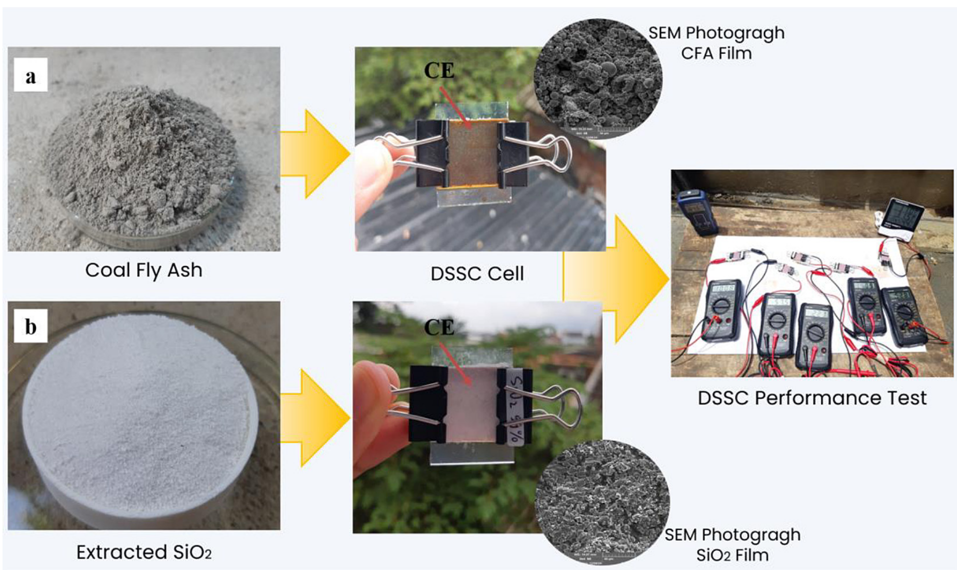

Figure 5. Schematic representation process and performance test of assembled DSSC

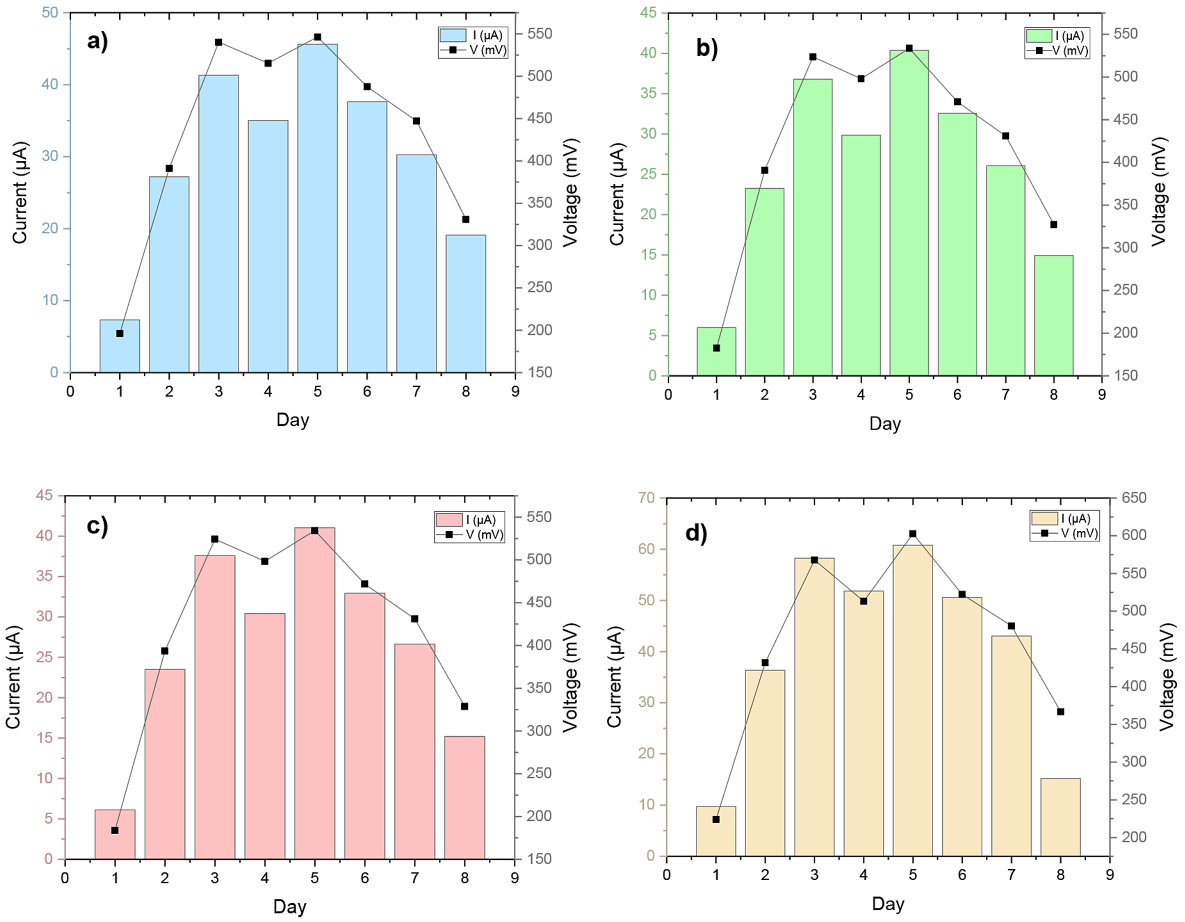

Figure 6. The I-V graph obtained at different purity of SiO_2; (a) CFA, (b) SiO_2 before purification, (c) SiO_2 after purification, (d) SiO_2 artificial

was followed by CFA (purity 52.91%) (Fig. 6a), which produced 546.4 mV and 45.62 μA SiO_2, and SiO_2 after purification (purity 91.20%) (Figure 6c), which produced 534.41 mV and 41.04 μA. The DSSC with SiO_2 material without purification (purity 72.54%) produced the lowest voltage and current was 533.66 mV and 40.37 μA as shown in Figure 6b.

Efficiency of DSSC

The relationship between the electrical energy generated and the light energy received by a dye-sensitized solar cell (DSSC) is used to calculate the cell's efficiency. The type of material used and the amount of light received have a significant impact on the efficiency of dye-sensitized solar cells, or DSSCs. Optimizing the performance and design of solar cells in this setting requires an understanding of how differences in irradiance and material type affect DSSC efficiency. As shown in Figure 7, SiO_2 artificial (purity 99.9%) achieve the maximum was 0.0113%. CFA (purity 52.91%) was 0.0076%, CFA DSSC produces the second-highest efficiency. SiO_2 after purification (purity 91.2%), produces the next-highest efficiency was 0.0062%. The last material SiO_2 before purification (72.54%), was 0.0061%.

Based on this study, a higher SiO_2 purity in the counter electrode material would result in improved performance. This is supported by the observation that SiO_2 artificial (purity 99.9%) exhibits the highest efficiency of 0.0113%. However, the results, as shown in Figure 7, show that the performance of the DSSC does not increase as the SiO_2 content in the counter electrode material does. In fact, there is a decrease in performance in DSSC with a purity of 72.54% compared to CFA, which has a purity of 52.91%. The presence of contaminants in the form of NaCl can cause this issue in the SiO_2 pre and post purification. Sodium chloride (NaCl) is formed as a byproduct when silicon dioxide (SiO_2) is extracted from coal fly ash. This occurs through a reaction involving sodium-based chemicals in the extraction

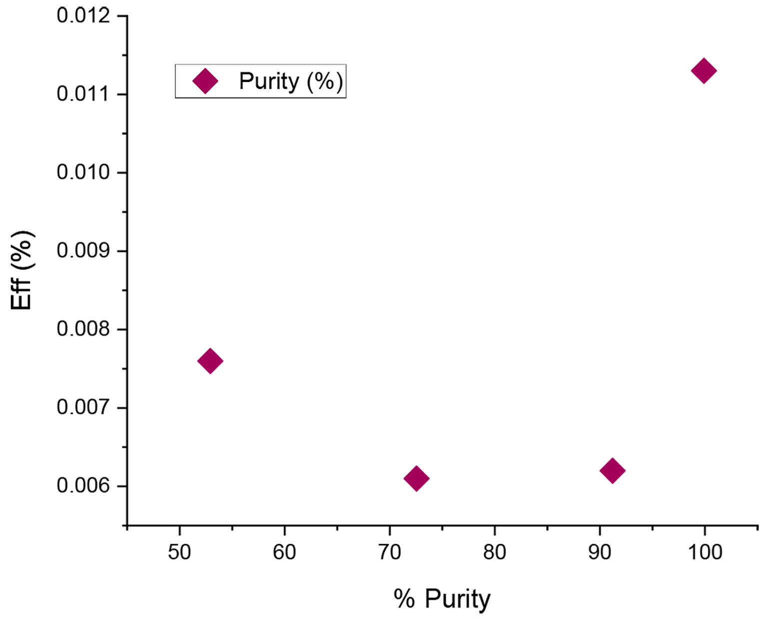

Figure 7. The effect of SiO_2 purity on DSSC efficiency

process. According to this research, the highest efficiency was achieved by using SiO_2 artificial with a purity of 99.9%, is 0.0113%, which is better than the study conducted by Musiana et al., 2020, using carbon as the counter electrode. The highest efficiency was achieved by using carbon with yellow sweet potato extract, which is 0.00818%. However, when using SiO_2 derived from coal fly ash, the efficiency was slightly lower, which is 0.0076%. Therefore, the approach using high-purity SiO_2 shows better potential to achieve higher efficiency.

In DSSC, NaCl has the potential to degrade the dye's quality and interfere with the electrode. Since NaCl is corrosive, it can lessen the capacity of cells to transmit electricity. Furthermore, it is possible that NaCl will interact with dye molecules, degrading them and lowering their capacity to absorb light and generate electrons.

Compared to using platinum as a counter electrode, the efficiency gained from using silicon dioxide (SiO_2) derived from coal fly ash is still quite small. Consequently, a number of steps will still need to be taken in the future to enhance the performance of DSSC. First, improving the purity of SiO_2 derived from coal fly ash may enhancement the efficiency. This can be carrying out extra purification procedures to get rid of contaminants that could obstruct electron flow. Second, The morphology and particle size of SiO_2. The SiO_2 particle size and morphology might affect the surface area that is existing for dyes absorption, an essential process for electron transport. The efficiency can be increased by optimizing these factors. Furthermore, improving the absorption of dye. Enhancing the SiO_2 surface's dye adsorption capabilities may effect in improved light harvesting and a rise in the efficiency of the DSSC.

In addition to material use, the efficiency of dye-sensitized solar cells (DSSCs) is strongly influenced by the intensity of light received, or what is called irradiance. DSSC efficiency is measured as the ratio between the electrical energy produced by the cell and the light energy received by the cell. Based on the research, at low irradiance levels (< 600 W/m^2), the current (Figure 8a) and voltage (Figure 8b) produced tend to increase linearly with increasing light intensity, which will also affect the efficiency of the DSSC. At higher light intensities (> 600 W/m^2), more electrons are generated. However, this also increases the chance of electron-hole recombination, which is the return of excited electrons to the ground state before they can be contributed to the external circuit. This higher recombination reduces the number of electrons contributing to the electric voltage, thus reducing the efficiency. This can be seen in Figure 8, which shows an increase in current and decrease in voltage at irradiance conditions above 600 W/m^2. It occurs due to several factors related to the behavior of the PV cells under different levels of sunlight intensity. At high irradiance,

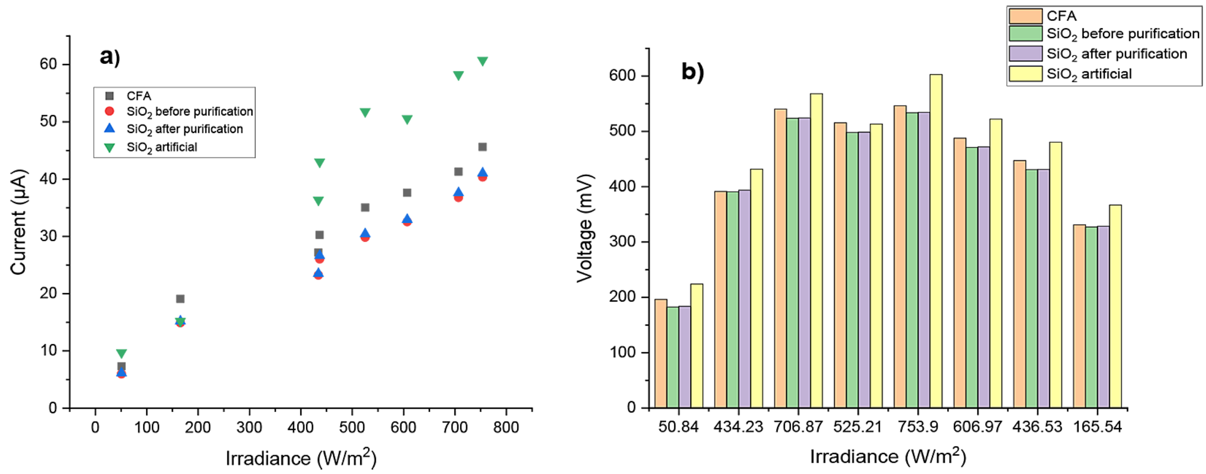

Figure 8. Current-Voltage in different irradiance

the photocurrent in a solar cell increases because more photons are available to excite electrons. However, this increased current can lead to a reduction in the voltage output due to the internal resistive losses within the cell. High irradiance often leads to an increase in the cell temperature. As the temperature of the DSSC rises, the open-circuit voltage decreases due to the temperature-dependent nature of the semiconductor material. This combination can lead to a decrease in the overall efficiency of the solar cell under very high light intensity conditions.

CONCLUSION

This study aims to investigate the effect of SiO_2 purity obtained from coal fly ash (CFA) on the photoelectric performance of dye-sensitized solar cells (DSSCs). Dye-sensitized dye cells have been created and sealed in accordance with the procedure to ensure proper operation. The study found that the purity of silicon dioxide obtained from coal fly ash (CFA) significantly influences the photoelectric performance of DSSCs. Higher purity levels of SiO_2 used as counter electrode materials were correlated with increased efficiency in the DSSCs. The study demonstrated that DSSCs utilizing SiO_2 with a purity of 99.9% achieved the highest efficiency at 0.0113%, whereas DSSCs with SiO_2 purities of 91.20%, 72.54%, and 52.91% exhibited progressively lower efficiencies of 0.0062%, 0.0061%, and 0.0076%, respectively. These findings highlight the importance of SiO_2 purity in optimizing the performance of DSSCs.

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