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Innovative bio-inspired solar cells using fly ash-based dye-sensitized cells with fruit extract enhancements and Averrhoa bilimbi electrolyte

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Abstract
This study responds to the urgent need for renewable energy in Indonesia, driven by climate change and the energy crisis, by developing dye-sensitized solar cells (DSSCs) using locally sourced, eco-friendly materials. Traditional silicon-based photovoltaic cells, which have plateaued at 27% efficiency, are costly and environmentally unfriendly, leading to the demand for alternatives like DSSCs, which offer lower production costs, flexibility, and effective performance in diffuse light. The research focuses on designing DSSCs with Fe and Mg extracted from fly ash as counter electrodes, dragon fruit peel as a natural dye sensitizer, and Averrhoa bilimbi as an electrolyte booster. UV-Vis spectroscopy demonstrated that dragon fruit dye absorbs light effectively in the 360-700 nm range, peaking at 550 nm, making it an ideal sensitizer for wide-band gap semiconductors. Voltage output tests showed that Fe-doped DSSCs consistently outperformed Mg-doped ones, with Fe-based cells generating a maximum voltage of 413 mV compared to 163 mV for Mg-based cells. Long-term testing over three months further demonstrated Fe-doped cells' superior performance, peaking at 454.6 mV, while Mg-doped cells reached 261.96 mV. These results highlight Fe's effectiveness as a doping material, improving DSSC efficiency and supporting the use of natural dyes and sustainable materials. The study aligns with prior research on the critical role of material properties and solar irradiance in DSSC performance, demonstrating the potential of using fly ash and natural dyes for efficient solar energy solutions in South Sumatra. Future research will focus on optimizing material composition for enhanced performance.

Keywords:
DSSC;
Fe and Mg Counter Electrodes;
Fly Ash;
Natural dye;
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INTRODUCTION

Climate change and the energy crisis have prompted Indonesia to shift and prioritize renewable energy [1][2]. Palembang and South Sumatra generally have sizeable solar energy potential [3], and several researchers utilize this advantage in the agricultural sector, such as Yurni et al.'s solar-powered automatic greenhouse in [4][5], and sprinkler systems by Mases et al. in [6] and Putra et al. in [7].

The most widely used photovoltaic (PV) technology today is silicon-based solar cells, known as the first generation, which achieve a maximum efficiency of 27% [7, 8, 9]. Thin-film technology represents the second generation and involves materials like Indium Gallium Selenide (CIGS), Cadmium Telluride (CdTe), and Gallium Arsenide (GaAs). However, thin-film solar cells are more expensive, the materials are harder to obtain, and their production processes are not environmentally friendly, contributing significantly to CO2 emissions. Consequently, there is a need for more sustainable and truly renewable PV technologies.

Dye-sensitized solar cells (DSSC) have emerged as a promising alternative, offering cost-effective and environmentally friendly solutions, as discussed by Jia et al [10, 11, 12]. DSSC uses organic dyes to absorb sunlight and convert it into electricity, making them versatile for various applications. DSSC's production expenses are relatively low [10, 11, 12]. Materials utilized in DSSC, such as titanium dioxide (TiO_2), are very inexpensive and readily available [13].

Nature-based dyes and electrolytes have contributed greatly to developing DSSC, which has a wide range of design possibilities, including color, flexibility, and appealing design. This includes setting the ability of the solar cell produced to facilitate low-light regions by adjusting the type of dye and electrolyte used [14, 15, 16, 17, 18].

Another environmental issue contributing to nature destruction is manufacturing waste such as Fly Ash as the product of coal combustion. However, when investigated further, the fly ash composition still has useful metals that can be utilized as the counter electrode, such as Fe, Mg, Si, Al, and Na. Hence, this abundance of waste could create an environmentally friendly electric source [19].

Dye choice is crucial in PV cell design due to the ability to capture photo energy from solar irradiance [19, 20, 21, 22]. Anthocyanin is a pigment that captures and binds photon energy to generate electricity. It can easily be found in reddish fruits such as dragon fruit and pomegranate, as investigated by Erande et al. in [23] Fereira et al. extracted dye from different flower petals [24].

Electrolytes play a critical role in the operation of DSSCs by facilitating the movement of charge carriers between the dye-sensitized semiconductor [17] and the counter electrode [25]. Bilimbi (belimbing wuluh) is also known as vegetable starfruit or sour starfruit due to its sour taste. It is used as a cooking spice or herbal remedy and contains tannins, saponins, sulfur glucose, formic acid, peroxide, flavonoids, and triterpenoids [26]. Since bilimbi contains formic acid and high acidity, it can generate electricity.

Dragon fruit peel contains betacyanin pigments, which can absorb light in certain spectrum ranges, making it suitable as a dye sensitizer in DSSC. Using dragon fruit peel waste as a dye harnesses its natural pigment's light absorption to generate electricity efficiently [16].

This research proposes the PV cell design using Fe and Mg extracted from Fly Ash as the counter electrode [25], the natural binding extracted from dragon fruit, and the electrolyte booster from Averrhoa bilimbi. The novelty of this paper is combining the natural power of Averrhoa bilimbi and dragon fruit anthocyanin with Fe and Mg electrodes to create a nature-friendly solar cell as the way to contribute to the advancement of renewable energy technologies and pave the way towards greener and more sustainable future

MATERIAL AND METHOD

This study fabricates a DSSC using Fe and Mg from fly ash as counter electrodes, dragon fruit peel as a natural dye sensitizer, and *Averrhoa bilimbi* extract as the electrolyte. TiO_2 paste acts as the semiconductor, assembled on FTO glass through paste deposition and sensitization, creating an eco-friendly, efficient solar cell, as shown in Figure 1.

Material

As shown in Figure 2, the DSSC layers are stacked with TiO_2 coated in dragon fruit dye, Fe or Mg as the counter electrode, and *Averrhoa bilimbi* electrolyte, all enclosed between TCO glass layers.

Semiconductor Material

Semiconductors play a vital role in the creation of solar cells, producing electricity through the photovoltaic effect by promoting electrons from the valence band to the conduction band. This study utilizes Titanium Oxide (TiO2) as the semiconductor because of its favorable characteristics [14].

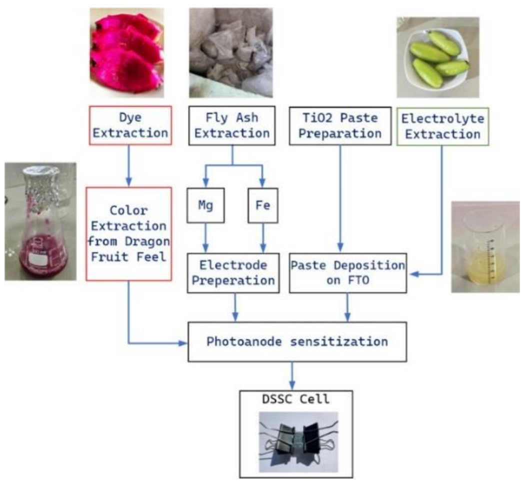


Figure 1. DSSC fabrication proposed in this study

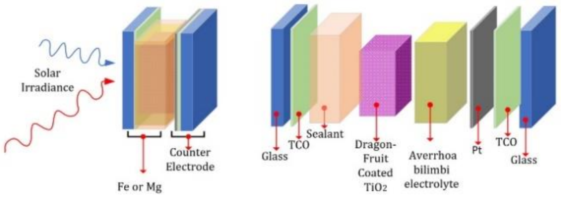


Figure 2. The layered structure of the DSSC

Dragon Fruit Peels as the Sensitizer

Dragon fruit peels, rich in anthocyanins, serve as an efficient natural sensitizer for DSSCs by absorbing a wide range of sunlight and converting it into electricity. This eco-friendly alternative reduces reliance on synthetic sensitizers, lowering both production costs and environmental impact.

Fe and Mg as Counter Electrodes

Fe and Mg, extracted from fly ash, are used as counter electrodes due to their abundance, cost-effectiveness, and excellent electrical conductivity [19]. These materials facilitate efficient electron transfer and rapid dye regeneration, crucial for sustaining photocurrent generation. Environmentally friendly, Fe and Mg align with green chemistry principles, offering a sustainable alternative to noble metals like platinum.

Averrhoa bilimbi as an electrolyte

Averrhoa bilimbi, rich in organic acids such as oxalic and ascorbic acid, enhances ionic conductivity in DSSCs; hence, its natural antioxidant properties improve electrolyte stability and device longevity [26].

Methods

The DSSC fabrication process, shown in Figure 1, involves constructing a solar cell with 2.5 cm x 2.5 cm glass. The optimal composition includes 180 grams each of dragon fruit and *Averrhoa bilimbi* extracts. To prepare the dye, 21 ml of distilled water is used as a solvent, along with 25 ml of a 1% methanol-HCl mixture to extract and stabilize the pigments, while 4 ml of acetic acid adjusts the pH for dye stability. Polyvinyl alcohol (1.5 grams) acts as a binder, evenly applying the dye to 0.5 grams of titanium dioxide (TiO_2) nanoparticles, which function as the semiconductor. Additionally, 13.5 ml of distilled water ensures the proper consistency. These components work together to enhance light absorption, electron transport, and overall DSSC stability.

Extraction and purification procedures for Mg and Fe from Fly Ash

The preparation of fly ash begins with drying a 10-gram sample in an oven at 110°C for 24 hours to remove organic matter. The dried fly ash is then combined with 100 mL of HCl in a 1:10 ratio. This mixture is stirred using a magnetic stirrer at 250 rpm for 24 hours. After stirring, the solution is filtered through filter paper to separate the filtrate from the residue. The filtrate is subsequently analyzed using Atomic Absorption Spectroscopy (AAS) to determine the ferrous content dissolved during the HCl extraction process.

Creating Dye Extract from Red Dragon Fruit

The preparation begins with peeling and chopping the skin of the red dragon fruit into small fragments. These fragments are then crushed using a mortar to achieve a smoother consistency. The resulting material is transferred into an Erlenmeyer flask and combined with a solution of 21 ml distilled water, 25 ml of 1% methanol-HCl, and 4 ml acetic acid. This mixture is allowed to stand overnight at a cool temperature. The next day, the mixture is filtered using filter paper to separate the liquid portion (filtrate) from the solid residues.

Creating a Booster from Averrhoa Bilimbi Extract, Preparing TiO2 Paste, and Coating with TiO2 Paste

For the *Averrhoa bilimbi* booster, start by mashing small pieces of the fruit and mixing them with 21 ml of distilled water, 25 ml of 1% methanol-HCl, and 4 ml of acetic acid in an Erlenmeyer flask. Allow the mixture to sit overnight in a cold environment, then filter it the next day to separate the liquid (filtrate) from the solid residue. For the TiO_2 paste, dissolve 1.5 grams of polyvinyl alcohol (PVA) in 13.5 ml of distilled water at 40°C for 30 minutes until a thick mixture forms. Then, combine 7.5 ml of this solution with 0.5 grams of TiO_2 to form the paste. The paste is then applied to fluorine-doped tin oxide (FTO) glass, with scotch tape marking the area. Finally, the coated glass is dried in a furnace at 450°C for 1 hour to enhance bonding and create pores.

UV-Vis for Data Analysis Method

Ultraviolet-visible (UV-Vis) spectroscopy is a powerful analytical technique used to measure a sample's absorbance or transmittance of UV and visible light. It is widely utilized for analyzing the optical properties of materials, especially in DSSC, to gain valuable insights into the optical properties of natural dye extracts and their interactions with semiconductor materials.

DSSC Electric Efficiency Analysis

The efficiency of a solar cell can be calculated based on how much the cell area (A) exposed to solar irradiance (G) and generates electricity. The efficiency is given by:

$$\eta = \frac{P_{out}}{P_{in}} x100\%, = \frac{I_{mp} \cdot V_{mp}}{P_{in}} x\ 100\%, \quad (1)$$

where I_{mp} and V_{mp} are the maximum current and voltage, and $P_{in} = G \times A$ [27]. In this study the irradiance used to calculate efficiency is the maximum solar irradiance of the day.

RESULTS AND DISCUSSION

An experiment was conducted to evaluate the effectiveness of the proposed Fe-doped and Mg-doped dye-sensitized solar cells (DSSCs). The initial test involved UV-Vis spectroscopy to assess the natural dye binder extracted from dragon fruit peels. The UV-Vis results, presented in Figure 3, illustrate the absorption spectra of the dragon fruit dye, offering insights into the transitions between its ground and excited states and the range of solar energy it can absorb. Anthocyanins, the main components of the dye, typically show a broad absorption band within the visible spectrum, attributed to charge transfer transitions between the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO).

Anthocyanins, which give dragon fruit peels their vibrant colors, have absorption maxima around 520 nm. Figure 3 shows the absorption range of 360-700 nm, making it an effective sensitizer for wide-band gap semiconductors. The absorption peak is around 550 nm in the visible region. The binding of dragon fruit extracted dye on the TiO_2 surface increases solar irradiance absorption, facilitated by carbonyl and hydroxyl groups in the anthocyanins. The effectiveness of the solar cells produced by the proposed method in Figure 1 is demonstrated by the amount of electricity generated by the DSSC.

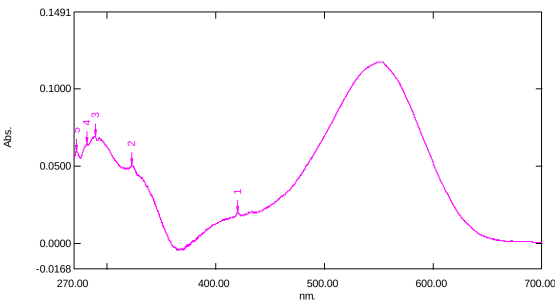


Figure 3. UV-Vis absorption spectra of dragon fruit dye considered in this study

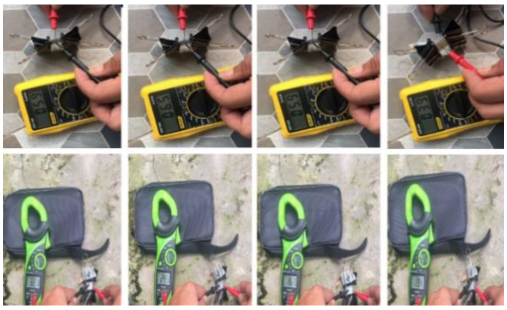


Figure 4. Measurement process

Figure 4 illustrates the process of measuring the voltage produced by the DSSC in this study. The early experiment was conducted over five days for both Fe-doped and Mg-doped DSSCs. During the five-day experiment, the weather conditions varied: the average irradiance on days 1 and 2 was cloudy (823 W/m² and 889 W/m²), day 3 was overcast (865 W/m²), day 4 was rainy (714 W/m²), and day 5 was very sunny with high solar irradiance (1039 W/m²).

Figure 5 illustrates the voltage output of a Fe-doped dye-sensitized solar cell (DSSC) over five days, measured at different times between 10:00 and 14:00, about solar irradiance. On Days 1 to 4, the irradiance fluctuates between 714 W/m² and 889 W/m², resulting in relatively low voltage outputs. However, on Day 5, when the irradiance peaks at 1039 W/m², the voltage output increases dramatically, exceeding 400 mV across all time intervals. This demonstrates the strong dependence of DSSC performance on sunlight intensity. These findings align with Shaikh et al. [18], who highlighted that higher irradiance directly improves DSSC efficiency by boosting energy conversion. Kumar et al. [15] also observed that natural dye-sensitized solar cells, such as the Fe-doped DSSC in this study, perform optimally under intense sunlight, leading to higher voltage outputs.

Additionally, Teja et al. [16] emphasized the importance of material properties and irradiance in determining DSSC performance, which is clearly reflected in the increased voltage output on Day 5. Overall, the graph strongly supports these findings, showcasing how critical solar irradiance is for enhancing DSSC efficiency and energy generation.

Figure 6 shows the voltage output of a Mg-doped DSSC under similar conditions. On Days 1 to 4, with irradiance levels between 714 W/m² and 889 W/m², the voltage remains relatively low, peaking at around 120 mV.

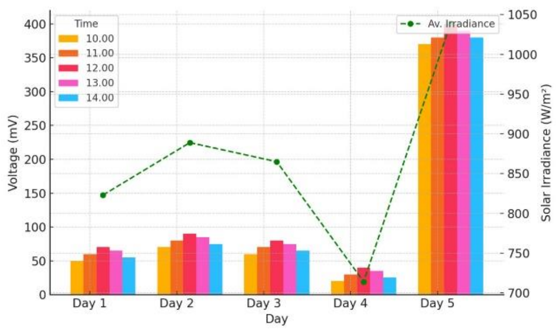


Figure 5. Fe-doped DSSC electricity generation test

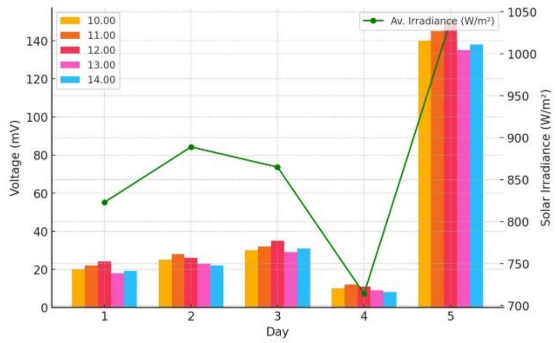
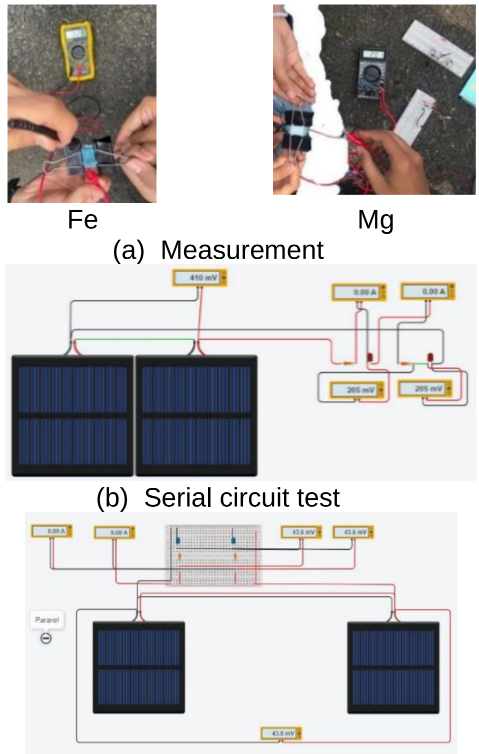


Figure 6. Mg-doped DSSC electricity generation test



(a) Measurement

(b) Serial circuit test

(c) Parallel circuit test

Figure 7. Serial and parallel connection test

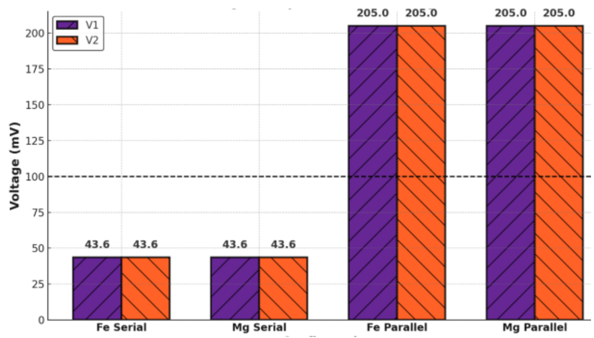


Figure 8. Serial and parallel test results

On Day 5, when irradiance reaches 1039 W/m², the voltage output rises, but only to around 160 mV. Compared to the Fe-doped DSSC, the Mg-doped version generates lower voltage outputs under the same irradiance conditions. This supports the findings of Shaikh et al. [18], who reported that Fe-based cells tend to outperform Mg-based ones due to better conductivity. Kumar et al. [15] also noted the critical role of material properties in DSSC efficiency, and the relatively lower performance of Mg-doped cells seen here highlights that material choice can significantly impact output. Teja et al. [16] further stressed that while solar irradiance is key, the type of doping material is equally important in optimizing DSSC performance, as demonstrated by the differences between the Fe and Mg-doped cells in this experiment.

Figure 5 and Figure 6 show that solar irradiance greatly affects both Fe-doped and Mg-doped DSSCs. On Day 5, with irradiance at 1039 W/m², the Fe-doped cell generates over 400 mV, while the Mg-doped cell only reaches around 160 mV, emphasizing Fe's superior performance as a doping material. This comparison underscores the critical role of both irradiance and material choice in enhancing DSSC efficiency.

Further tests assessed the performance of Fe and Mg-doped DSSCs in both series and parallel configurations, as shown in Figure 7. The DSSCs powered two LEDs with resistors added to the circuit. Figure 7b illustrates the series connection, while Figure 7c depicts the parallel setup. Figure 8 presents the results, showing the voltages generated by DSSCs 1 (V1) and 2 (V2) under each configuration.

Figure 8 highlights the results of serial and parallel tests on Fe and Mg-doped DSSCs, showing consistent voltage outputs in both setups. In the serial configuration, both DSSCs generated a uniform 43.6 mV per cell, while the parallel setup boosted the voltage to 205 mV per cell.

Unlike the no-load voltages in Figure 5 and Figure 6, Figure 8 displays load voltages, assessing the DSSCs' efficiency and stability across different electrical configurations. These findings indicate that Fe and Mg doping enhance DSSC performance, with serial and parallel setups offering flexibility to optimize voltage and current outputs for various energy needs.

An extended durability assessment of the Fe and Mg-doped DSSCs was conducted over a three-month period (April 1 to June 2), as shown in Figure 9. Given that the typical lifespan of DSSCs is approximately three months, this monitoring aimed to evaluate the cells' stability and longevity. Sustained electricity generation beyond this timeframe would indicate improved performance, validating both the design and material choices. These findings would not only demonstrate the study's success but also highlight the potential for further advancements in DSSC technology.

In comparison, studies such as Shaikh et al. [18] emphasize the importance of consistent voltage performance in DSSCs, particularly under varying electrical configurations, which aligns with the consistent voltage outputs observed in both Fe and Mg-based cells in serial and parallel setups. Kumar et al. [15] also highlighted the role of material properties in enhancing long-term stability and energy generation, paralleling the three-month performance evaluation conducted in this study. The potential for DSSCs to maintain performance beyond their typical lifespan could mirror the advancements discussed by Teja et al. [16], who focused on the stability of natural dye-based DSSCs, showing the critical role of material optimization in extending the operational life of these solar cells.

Figure 10 compares the efficiency of Fe- and Mg-based DSSCs across several dates, with efficiencies below 0.001%. Fe-based DSSCs generally outperformed Mg-based ones, peaking at 0.000634% on June 1.

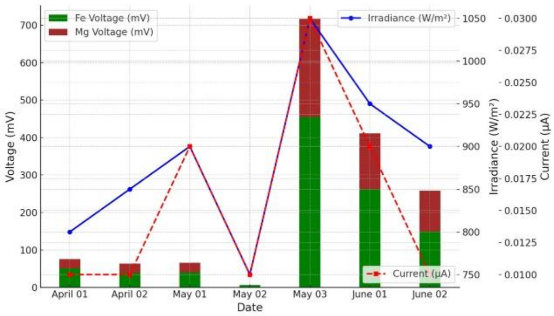


Figure 9. Voltage, irradiance, and current comparison for Fe and Mg-doped DSSCs over time

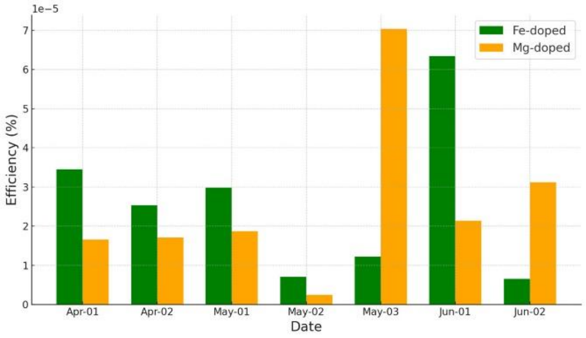


Figure 10. Efficiency Comparison of Fe- and Mg-doped DSSC

However, Mg-based cells reached their highest efficiency of 0.000704% on May 3, possibly due to optimal conditions. On April 1, Fe-based cells recorded 0.000345%, while Mg-based cells had 0.000166%, with slight declines observed by April 2. Although Fe-based cells maintained higher performance through May, Mg-based cells briefly surpassed them on May 3. Both DSSCs showed improved performance on June 2, with Fe-based cells at 0.000065% and Mg-based cells at 0.000312%. These results emphasize how environmental factors, like irradiance, significantly impact DSSC efficiency and solar energy conversion.

This study's findings align with Teja et al. [16] and Shaikh et al. [18], both of whom emphasize the impact of environmental factors like light intensity on DSSC performance. The observed fluctuations in efficiency highlight DSSCs' sensitivity to changing conditions, underscoring the critical role irradiance plays in solar cell output, as discussed by Kumar et al. [15]. The performance of Fe-doped and Mg-doped DSSCs in this research reaffirms the importance of irradiance and material properties. As noted by Shaikh et al. [18], both cells showed increased voltage under peak sunlight, with Fe-doped cells outperforming Mg-doped ones. Kumar et al. [15] also emphasized material selection, with Fe proving more effective than Mg for enhancing DSSC efficiency. The use of dragon fruit dye supports the findings of Prakash et al. [14] and Teja et al. [16], which highlighted strong light absorption in natural dyes. Additionally, Mahajan et al. [17] and Wu et al. [25] stressed optimizing dye and material properties, validated by this study. The proposed use of dragon fruit dye and Averrhoa bilimbi electrolyte, coupled with Fe from fly ash, shows potential for sustainable DSSC development in Palembang, South Sumatra. The results confirm that combining natural dyes with optimal materials like Fe significantly enhances DSSC performance, offering a promising renewable energy solution.

CONCLUSION

Solar energy is an environmentally friendly energy source that has a high potential to be developed in Palembang, Indonesia. The current silicon-based PV cell has reached its maximum efficiency of 27%. Thin film, which is still using less abundant material than silicon, is not purely renewable. DSSC offer unique advantages in low production costs, flexibility, performance in diffuse light, and aesthetic versatility. This paper presents the DSSC design utilizing the Fe and Mg extracted from fly ash for counter electrode with natural binder from dragon fruit peels and electrolyte booster extracted from Averrhoa bilimbi. The UV-Vis test shows the binder absorption range of 360-700 nm, making it an effective sensitizer for wide band gap semiconductors, and the absorption peak around 550 nm in the visible region. The experiment for electricity generation testing shows that Fe-doped DSSC generates more voltage (V_{max} = 413 mV) than Mg-doped DSSC (163 mV). This result is due to the fact that Fe is a better conductor than Mg. The series and parallel connection circuit test was conducted to show the possibility of proposed DSSC cell to power a load (LED) and to show the prolong life of the proposed DSSC the experiment was conducted for 3 months with the highest produced electricity is 454.6 V for Fe-doped and 261.96 mV for Mg doped DSSC. Future research will investigate the best composition of DSSC material design.

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