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Nature Meets Technology: A Case Study of Gambier Leaf and N719 Hybrid Sensitizer in DSSCs Application

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Abstract

Dye-sensitized solar cells (DSSCs) offer a cost-effective and eco-friendly renewable energy solution, with dye as sensitizers playing a crucial role in their performance. While synthetic dyes are commonly used, they are toxic and expensive, making natural dyes such as gambier a sustainable and environmentally friendly alternative. This study explores the role of dye in the performance of DSSC based-TiO_2 as semiconductor, with a highly optimized surface, the dye is expected to absorb effectively, facilitating electron excitation. Two types of TiO_2 were utilized: synthesized TiO_2 and commercial P25. Morphological analysis confirmed that combining these materials enhances surface area and improves light scattering, boosting DSSC performance. To reduce synthetic dye usage and optimize the potential of natural gambier dye, a hybrid approach with the synthetic dye N719 was employed. Photovoltaic measurements revealed that this hybrid technique successfully optimized the natural dye's potential, achieving an efficiency of 2.764%. These findings demonstrate that natural dye can be effectively utilized, offering a sustainable and non-toxic alternative for DSSC applications.

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1. Introduction

Dye-sensitized solar cells (DSSCs), first developed by Brian O'Regan and Michael Gratzel in 1991 at the University of California, Berkeley, represent a breakthrough in solar cell technology due to their affordability and unique design [1]. As a type of third-generation photovoltaic technology, DSSCs offer several advantages over conventional silicon-based solar cells, including ease of fabrication, low production costs, and the use of flexible substrates [2], [3]. Unlike traditional cells that rely on p-n junctions, DSSCs operate via a photoelectrochemical mechanism, where a photo-sensitized anode absorbs sunlight and initiates electron flow through a redox electrolyte system [4]. Their basic structure comprising a transparent conducting oxide (TCO) glass, a mesoporous titanium dioxide (TiO_2) layer, a sensitizer dye, an electrolyte, and a counter electrode works synergistically to harvest light and convert it into electrical energy [3].

The working principle of DSSCs hinges on the efficient interplay between light absorption, electron excitation, charge injection, and regeneration. When sunlight strikes the sensitizer dye adsorbed on the TiO_2 surface, electrons in the dye are excited from the ground state to a higher energy level [5]. hese electrons are then injected into the conduction band of TiO_2, leaving behind oxidized dye molecules that are subsequently regenerated by redox reactions in the electrolyte. The electrons travel through the external circuit to reach the counter electrode, while the electrolyte transfers holes to complete the circuit. The success of this process relies heavily on the alignment of energy levels among the dye, semiconductor, and electrolyte, making the selection and modification of dyes a critical aspect in enhancing cell performance[6].

Among the key components in DSSCs, the sensitizer dye plays a pivotal role in determining light absorption efficiency and overall device performance [7]. Ideally, a dye must exhibit strong absorption in the visible spectrum, good photochemical stability, and appropriate alignment of the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels with respect to the TiO_2 conduction band and the redox electrolyte, respectively [8]. Sensitizers used in DSSCs can be broadly categorized into synthetic and natural dyes. Ruthenium-based complexes like N3 and N719 remain the benchmark in synthetic dyes due to their excellent photostability and broad absorption spectra [9], [10]. However, the high cost, limited availability, and potential environmental toxicity of these metal-based dyes have sparked increasing interest in natural dyes as green alternatives [11].

Natural dyes, derived from plant sources such as fruits, flowers, leaves, and bark, offer ecological and economic benefits due to their biodegradability, non-toxicity, and renewable nature. Nevertheless, their practical application is often hindered by poor light stability, low molar extinction coefficients, and weak chemical bonding to TiO_2 surfaces. To overcome these limitations, recent studies have explored various strategies including co-sensitization, dye modification, and molecular engineering [11]. Co-sensitization, in particular, involves combining two or more dyes often a synthetic dye with a natural dye to broaden the absorption spectrum and improve electron injection efficiency, leading to enhanced photovoltaic performance [12].

Uncaria gambir (gambier) has gained recognition as a promising natural dye for photovoltaic applications due to its high concentration of catechins and tannins. These polyphenolic compounds feature functional groups, such as hydroxyl and carboxyl, that enable strong binding to TiO_2 surfaces and promote effective absorption of visible light [13]. As an abundant and low-cost botanical resource cultivated extensively in Southeast Asia, gambier offers substantial potential for environmentally sustainable dye production. However, its practical application in dye-sensitized solar cells (DSSCs) is hindered by inherent drawbacks, including rapid photodegradation and oxidative instability under continuous illumination [14], [15], [16]. To overcome these limitations, various refinement techniques and hybrid sensitization approaches have been explored to improve its stability and photovoltaic efficiency [17].

This study examines hybrid dyes that combine the eco-friendly properties of natural dyes with synthetic dyes. The synthetic dye used in this study is the Ruthenium-based N719 dye, selected for its compatibility with TiO_2 semiconductor surfaces [18]. By addressing these challenges, this research advances sustainable innovation in dye technology, aligning closely with the focus on environmentally conscious solutions.

2. Methodology

2.1 Substrate Cleaning and Masking

Prior to conducting the experiments, the conductive glass used in this study, fluorine-doped tin oxide (FTO) from sigma-Aldrich company, underwent a thorough cleaning process. The cleaning procedure involved sequentially washing the FTO glass with deionized water, ethanol, and acetone. After the cleaning process was completed, the conductive side of the glass was inspected and prepared with masking areas in circular shapes (0.25 cm^2) and rectangular shapes (0.5 cm^2). This preparation was tailored to meet the specific requirements of subsequent characterizations. The masking area designs are illustrated in Fig 1.

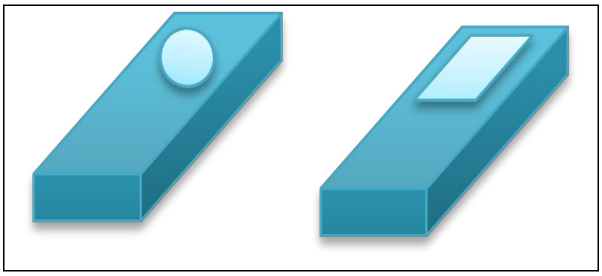

Fig. 1 *The masking area design on the conductive glass (FTO)*

2.2 Preparation and Deposition of TiO_2 Solution

In this study, the optimization of the semiconductor surface based on titanium dioxide (TiO_2) was conducted. Two types of TiO_2 materials were utilized: synthesized TiO_2, prepared using titanium isopropoxide (TTIP) as the precursor, and commercial TiO_2 (P25) from sigma-Aldrich company. These materials were mixed in varying ratios (0:1, 0.5:0.5, 1:0), as shown in Table 1.

Table 1 *Ratio between TiO_2 powder and P25 powder*

TiO_2 Powder (gram)	P25 Powder (gram)	Ratio
0	0.3	0:1
0.15	0.15	0.5:0.5
0.3	0	1:0

After mixing the two materials, acetic acid was added, followed by ethanol and 5 drops of Triton X-100. Once the solution was fully homogenized, it was sonicated for 30 minutes. The same procedure was repeated for the other mixing ratios. Once the TiO_2 solution was prepared, it was deposited onto the substrate using the spray pyrolysis method [18][1]. During this process, the masked samples were placed on a hot plate maintained at 150°C. After deposition, the sprayed samples were annealed in a furnace at 450°C for 1 hour.

2.3 Dye Extraction and Sensitization Process

The dyes used in this study consist of two types: the synthetic dye (N719) and the natural gambier dye. The N719 dye was prepared by dissolving 0.0178 grams of powder in a mixture of 25 ml of butyl alcohol and 25 ml of acetonitrile, followed by sonication for 30 minutes. In contrast, the gambier dye was extracted using maceration technique. The gambier dye was mixed with isopropyl alcohol (IPA) as a solvent and left for 24 hours. Afterward, the solution was filtered, and the final step involved separating the dye solution from the alcohol through evaporation. The evaporation was conducted manually by heating the dye solution on a hotplate, ensuring the temperature did not exceed 82.5°C, which is the boiling point of IPA.

Once both dyes were prepared, the next step involved immersing the semiconductor samples into the dye solutions. Three types of samples were used: sample 1 was immersed in the N719 dye, sample 2 in the pure gambier dye, and sample 3 in a hybrid solution of both of dyes. All samples were immersed simultaneously for 24 hours.

2.4 DSSC Assembly

The final step was the assembly of the DSSC. The dye-immersed sample (working electrode) was assembled into a sandwich structure to form the DSSC. Platinum (Pt) was used as the counter electrode, and a traditional iodide/triiodide electrolyte was applied. An illustration of the assembly process is presented in Figure 2.

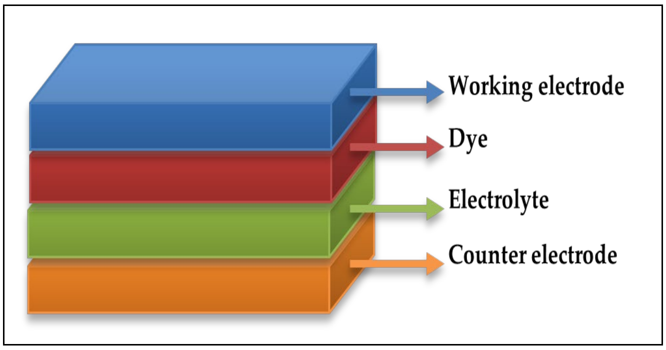

Fig. 2 *Illustration of the sandwich structure in DSSC application*

3. Result and Discussion

3.1 DSSC Performance for Optimization Semiconductor Surface

This study focused on optimizing the surface area of the semiconductor for dye immersion applications. To evaluate the optimization, the prepared samples were immersed in the synthetic dye N719, followed by current-voltage (I-V) measurements to assess their performance.

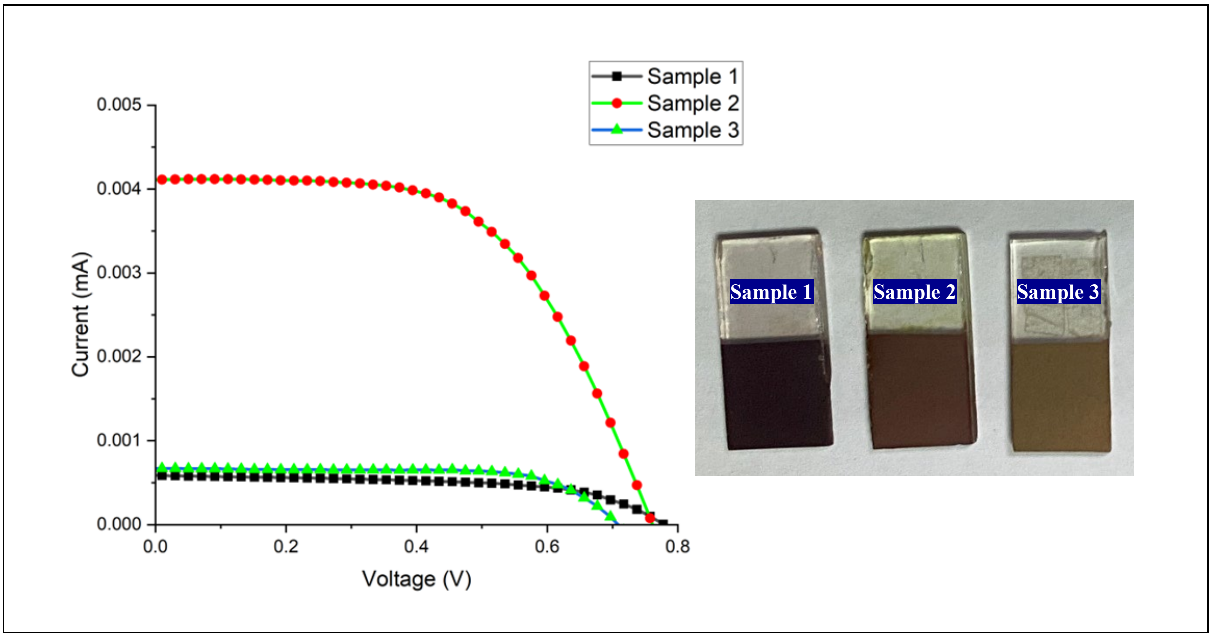

Fig. 3 *Current voltage (The J_{sc}-V) characteristics (left) and real sample (right) of DSSCs prepared with different TiO_2 ratios*

Figure 3 shows the current-voltage characteristics performance of three different mixing ratio which are sample 1 (ratio of TiO2 powder and P25 is 0:1), sample 2 (ratio of TiO2 powder and P25 is 0.5:0.5) and sample 3 (ratio TiO2 powder and P25 is 1:0). Table 2 summarizes the open circuit voltage, short circuit current, fill factor and the efficiency of three different samples. These DSSCs have almost the same V_oc value of 0.7V. It may occur due to the same thickness. V_oc (open-circuit voltage) is the voltage that measured at the solar cell's terminals when no current flowing. The higher V_oc values will indicate better cell quality. This V_oc value of 0.7V is for a standard solar cell with an absorption onset [19]. It makes sense that their values are almost the same. Besides, Sample 2 shows the highest J_sc value of 16.474. J_sc (short-circuit current) is the current measured at the solar cell's terminals when the voltage is zero. The higher J_sc value will indicate the better light absorption and charge generation. Sample 2 has the highest J_sc and efficiency which is the percentage of sunlight converted into electrical energy by the solar cell. This indicates that the sample is good at absorbing light and generating current because the higher efficiency values are desirable [20].

Table 2 *I-V performance of three different samples*

Type of samples	V_{oc}	J_{sc} (mA/cm^2)	FF	ŋ%
Sample 1	0.771	2.352	58.208	1.055
Sample 2	0.757	16.474	58.523	7.301
Sample 3	0.704	2.683	70.303	1.329

3.2 Structural Properties

The primary purpose of XRD is to identify and qualify the samples based on their diffraction patterns [21]. Other than that, it also can be utilized to look into the sample's crystal structure [22]. Figure 4 shows the XRD pattern for a) TiO_2 b) N719 c) gambier d) hybrid of gambier and N719. It was a crystal structure since it has a peak which are anatase and rutile. This peak proof that the crystal structure of P25 and synthesized TiO_2 are anatase and rutile. This phase contributes to a high surface area and enhanced light scattering, which facilitates more efficient light harvesting. These features are essential in areas like photocatalysis and solar energy, where capturing and using light efficiently is key. A larger surface area means more chances for light and other substances to interact. In contrast, better light scattering helps gather energy more effectively, contributing to better performance in different technologies. [23].

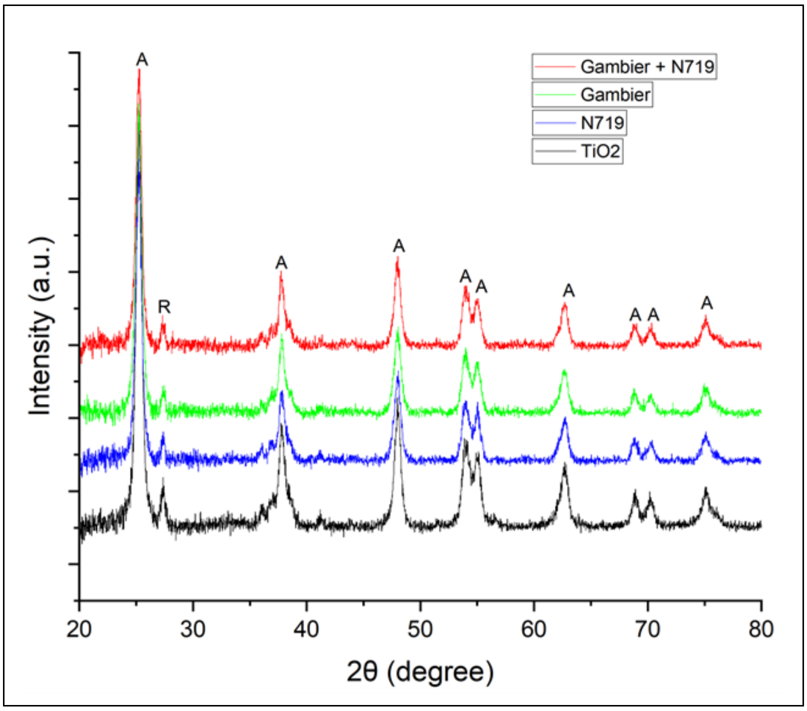

Fig. 4 *XRD pattern of film (a) TiO_2 (b) /N719 (c) TiO_2/gambier (d) TiO_2/hybrid (N719+gambier)*

3.3 Morphology Analysis

An advanced technology called FESEM is utilized to study the nanostructure image of the materials [1]. It is also used to see the structure in terms of shape, or size. Figure 5 shows the FESEM results of a) TiO_2 b) Dye N719 c) Dye gambier and d) Hybrid of dye gambier and N719 for Samples 2 (the optimize semiconductor).

The FESEM images were enlarged 50,000 times and showed different results of the surface morphology. In previous research [24], [25] stated that the crystals for P25 are anatase and rutile. Figure 5 (a) showed the shape of the surface is round like pores and not uniform in size. The big size of the particles was from P25 because the particle size for P25 is 25um [25]. While the smaller particles are from the TiO_2 powder. The TiO_2 powder crystals are anatase and the size of particles is around 14um – 17um [26]. This has proven that the mixing ratio of 1:1 is complete and has been proven through this FESEM characterization. The figure also illustrates that all samples exhibit consistent particle morphology, specifically in the form of nanoparticles.

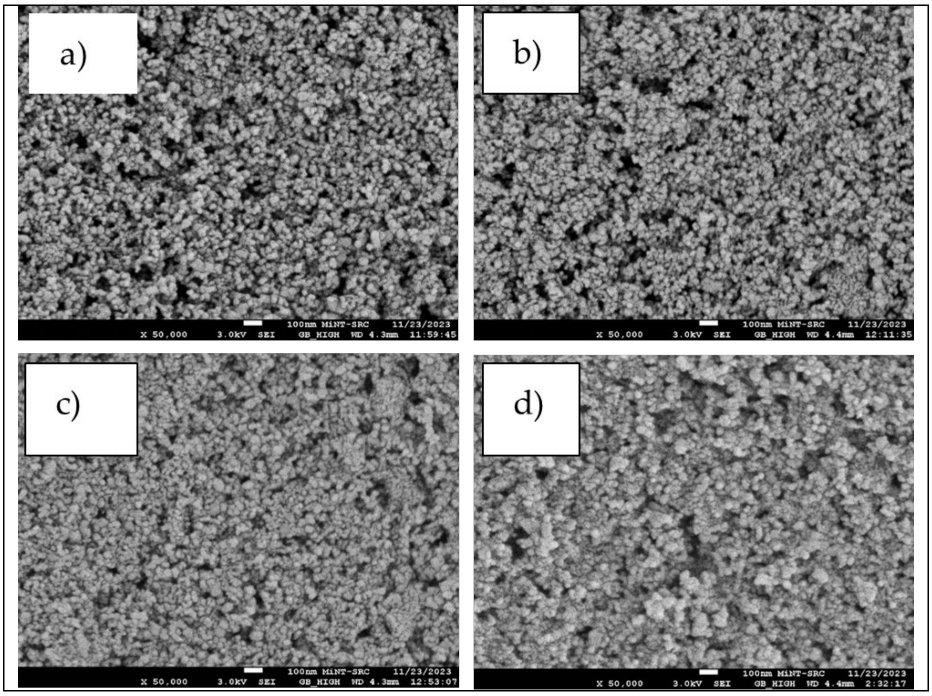

Fig. 5 *FESEM images of (a) TiO_2 film; (b) TiO_2 film-N719; (c) TiO_2 film-gambier; (d) TiO_2 film-hybrid (N719+gambier)*

This study effectively demonstrates the optimization of semiconductor surfaces for DSSC through comprehensive investigation that includes I-V measurements, XRD, and FESEM characterization. By adjusting the mixing ratios of TiO_2 between P25 and TiO_2 powder (TTIP), which comprises both rutile and anatase phases. The research emphasizes the importance of achieving an ideal balance between small and large particles from both phases. Small particles will provide a high surface area for dye absorption, while larger particles enhance light scattering. These findings highlight the potential of tailoring semiconductor surface properties to achieve higher photovoltaic efficiency and advance sustainable energy solutions.

3.4 Optical Properties

To evaluate the light absorption behavior of the prepared sensitizers, UV-Visible spectroscopy was conducted, and the corresponding optical output is presented in Figure 6. This figure shows the UV-Vis absorption spectra of TiO_2 films sensitized with gambier dye (red curve), N719 dye (blue curve), and a hybrid combination of both dyes (black curve). These spectra provide crucial insight into the optical properties of each dye and their interaction with the TiO_2 photoanode.

As shown, the gambier dye exhibits a relatively broad absorption profile in the visible region, indicating its potential for solar light harvesting. However, the absorption intensity is lower compared to the commercial N719 dye, which displays a distinct peak around 550–600 nm, characteristic of its strong molar extinction coefficient. The hybrid dye spectrum, meanwhile, demonstrates overlapping features from both components, suggesting a complementary absorption effect that can enhance light harvesting efficiency [27], [28]. This optical output graph serves to confirm the capability of each dye system in absorbing light within the visible range, which directly correlates to the potential photocurrent generation in the DSSCs. Understanding these differences is essential in explaining the photovoltaic behavior discussed in Section 3.1 and supports the rationale behind hybrid dye utilization to improve performance.

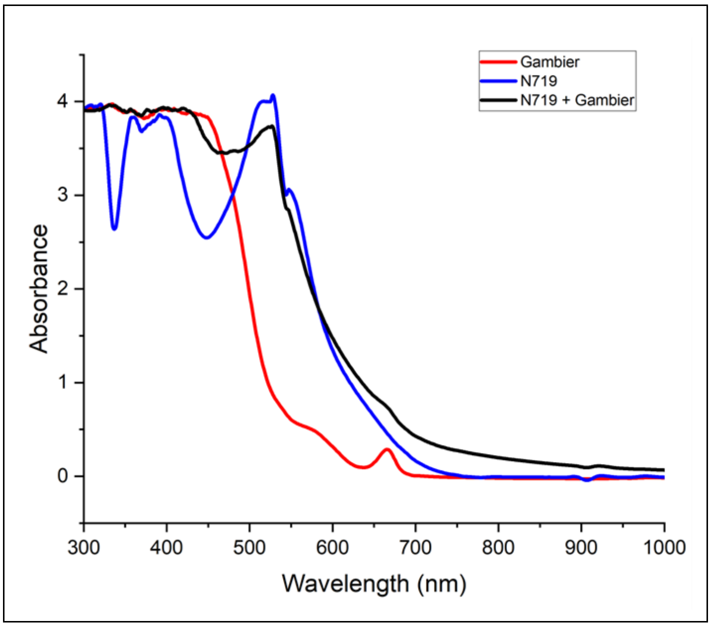

Fig. 6 *UV absorption of (a) TiO_2 film-gambier; (b) TiO_2 film-N719; (c) TiO_2 film-hybrid (N719+gambier)*

3.5 Electrical Properties

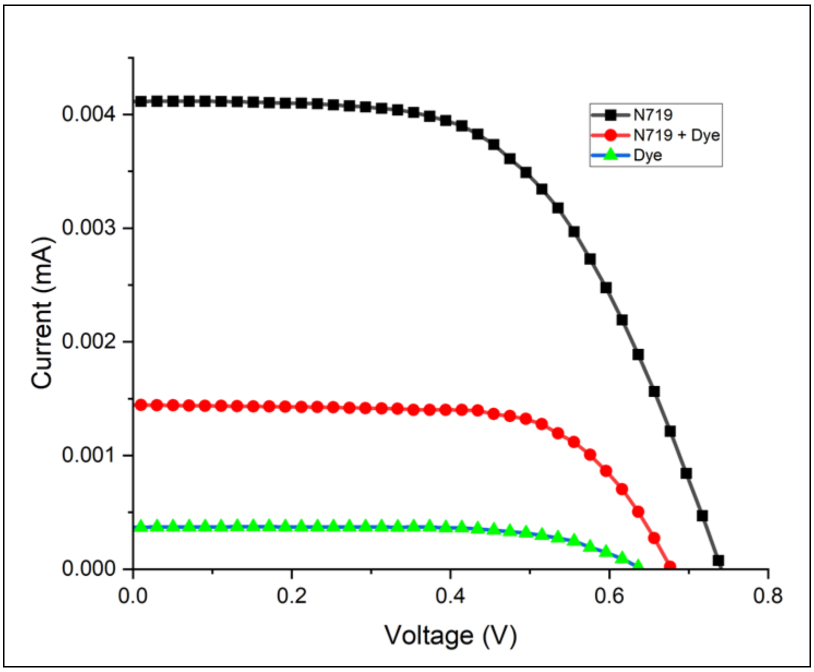

Fig. 7 *Current voltage (Jsc-V) characteristics of film with dye N719, dye gambier, and hybrid*

Figure 7 shows the current-voltage characteristics performance of dye concentration for dye N719, dye gambier and combination of dye gambier and N719. Dye N719 is commonly used in DSSC because of its stability and good performance [28], [29]. Table 3 summarizes the open circuit voltage, short circuit current, fill factor and the efficiency of three different kinds of dye. It can be seen that dye N719 $\boldsymbol{V_{oc}}$ value of 0.7V. Previously research stated that this value is a standard value [30], [31]. While dye gambier and combination of dye gambier and N719 have almost the same $\boldsymbol{V_{oc}}$ value of 0.6V. It may occur due to their same thickness but a little bit thin than the dye concentration of dye N719. The highest efficiency value is produced by dye N719 where $\boldsymbol{J_{sc}}$ = 16.474 mA/$\boldsymbol{cm^2}$, FF = 58.523 and ŋ = 7.301% and the lowest efficiency value is ŋ = 0.799% (with $\boldsymbol{J_{sc}}$ = 1.845 mA/$\boldsymbol{cm^2}$ and FF = 67.764) from dye gambier.

Although the Voc value of gambier dye (0.6 V) is lower than that of N719 (0.7 V), this range is still within the typical values reported for natural dye-sensitized solar cells [30], [31]. The reduced Voc may be attributed to limited electron injection efficiency and higher charge recombination rates, which are common limitations in organic dye systems. Despite these drawbacks, gambier dye demonstrates the ability to generate stable photovoltage, indicating its potential viability as a green sensitizer. While its current efficiency (0.799%) is not yet sufficient for large-scale solar panel applications, its eco-friendly nature and low-cost position as a strong candidate for future development [31], especially when combined with performance-enhancing strategies such as hybridization or structural refinement.

Table 3 *I-V performance of variation of dye*

Dye concentration	V_{oc}	J_{sc} (mA/cm^2)	FF	η%
N719	0.757	16.474	58.523	7.301
Gambier	0.639	1.845	67.764	0.799
Gambier + N719	0.674	5.785	70.914	2.764

4. Conclusion

This study investigated the potential of gambier dye as a natural sensitizer for dye-sensitized solar cells (DSSCs), both in standalone application and in combination with the synthetic dye N719. Through optimization of the TiO_2 photoanode specifically using a 1:1 mixing ratio of commercial P25 and synthesized TiO_2 enhanced morphological and structural properties were achieved, contributing to improved light scattering and electron transport. The UV-Visible spectra confirmed that the hybrid dye system extended the absorption range, leading to stronger light harvesting. Correspondingly, the hybrid DSSC achieved the highest power conversion efficiency of 2.764%. These findings suggest that hybrid sensitization using gambier and N719 dyes is a promising strategy to improve DSSC performance, and further validate the role of natural dyes in advancing eco-friendly photovoltaic technologies

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Conflict of Interest

The manuscript has not been published elsewhere and is not under consideration by other journals. All authors have approved the review, agree with its submission and declare no conflict of interest on the manuscript

Author Contribution

The authors confirm contribution to the paper as follows: ***Conceptualization:*** *D.G. Saputri, M.K. Ahmad, A. Supriyanto, A.B. Faridah, A.M.S. Nurhaziqah;* ***Writing-Original Draft Preparation:*** *D.G. Saputri, N.H. Rasman;* ***Methodology:*** *D.G. Saputri, A.B. Faridah, A.M.S. Nurhaziqah, Ahmad Ramli Shazleen;* ***Software:*** *N.H. Rasman, Ahmad Ramli Shazleen, Noor Kamalia Abd Hamed, Abdinasir Hirsi, M.Y. Ahmad;* ***Validation:*** *M.K. Ahmad, A. Supriyanto, A.B. Faridah, Noor Kamalia Abd Hamed, M.Y. Ahmad;* ***Writing-Reviewing and Editing:*** *M. N. Ribuan. All authors reviewed the results and approved the final version of the manuscript.*

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