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Development of Environmentally Friendly Nano Photosensitizers from Zn-*Catharanthus roseus* and Cu-*Catharanthus roseus* Complexes for Dye-Sensitized Solar Cells

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Abstract: Dye-sensitized solar cells are a promising renewable energy technology that offers an environmentally friendly alternative source of electricity. One of main components of DSSCs is photosensitizer, which functions to absorb light and convert it into electrical energy. Use of natural-based photosensitizers, such as plant extracts from *Catharanthus roseus*, offers advantages in terms of sustainability and environmental friendliness. Nano sized photosensitizers based on transition metals such as zinc (Zn) and copper (Cu), combined with active compounds from *Catharanthus roseus* extract, can enhance properties of photosensitizer, particularly in light absorption and electron transfer efficiency. However, type of metal used in the nano photosensitizer Zn or Cu can have a significant impact on performance of solar cell, especially in terms of energy conversion efficiency and stability. This study aims to examine effect of different nano-photosensitizers, Zn-*Catharanthus roseus* and Cu-*Catharanthus roseus*, on performance of dye-sensitized solar cells. Method used includes ultrasonic-assisted extraction of *Catharanthus roseus* leaves using ethanol as the solvent. Nano-photosensitizers of Zn and Cu were then synthesized in equal ratios. Synthesized nano-photosensitizers were applied to TiO_2 electrodes as working electrode, with Pt used as the counter electrode in DSSC assembly. Efficiency testing was carried out using a halogen lamp as light source. The results showed that difference between the Zn-*Catharanthus roseus* and Cu-*Catharanthus roseus* nano-photosensitizers affected efficiency of the dye-sensitized solar cells. Highest efficiency was obtained using the Zn-*Catharanthus roseus* photosensitizer, with a value of 4.97×10^{-3} %, indicating that Zn-*Catharanthus roseus* has strong potential as a photosensitizer for dye-sensitized solar cells.

Keywords: Nano Photosensitizer, Zn-*Catharanthus roseus*, Cu-*Catharanthus roseus*, Dye-sensitized Solar Cells.

Abbreviations: Dye-Sensitized Solar Cell (DSSC), Fluorine-Doped Tin Oxide (FTO), Fill Factor (FF), Efficiency (η), Joint Committee on Powder Diffraction Standards (JCPDS)

Introduction

Availability of energy and the environmental impact of fossil fuel use have become global issues. Limited and non-renewable nature of fossil energy sources has led to rising energy costs, while the resulting carbon emissions negatively affect climate change and air pollution (Sukmawati and Hariyani 2025). Therefore, development of clean, efficient, and sustainable alternative energy sources is urgently needed to address these challenges. Solar energy is one of the most promising renewable energy sources due to its abundance and zero carbon emissions (Gunawan et al. 2021). In this context, Dye-Sensitized Solar Cell (DSSC) technology has emerged as an important innovation, offering high energy efficiency at a lower production cost compared to conventional silicon-based solar cells.

DSSC operate on principle of photoelectrochemistry, where photosensitizer plays a key role in capturing light energy and transferring electrons to semiconductor, typically

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titanium dioxide (TiO_2) (Prajapat et al. 2025). Performance of DSSC greatly depends on photosensitizer's ability to absorb visible light and efficiently transfer electrons (Umayya 2023). Ruthenium-based complex dyes, such as N719, have demonstrated efficiencies as high as 11.2%, but their use is limited due to high cost of ruthenium, complex synthesis processes, and environmental toxicity (Hardani, Cari, and Supriyanto 2020). These limitations have encouraged efforts to find more eco-friendly, cost-effective, and naturally derived photosensitizers.

Natural pigments from plants are promising candidates as photosensitizers in DSSC. One plant of interest is *Catharanthus roseus*, known to contain bioactive compounds such as anthocyanins, flavonoids, and alkaloids (Muna et al. 2025). These compounds can absorb visible light and act as natural electron donors. However, the main drawbacks of natural dye-based photosensitizers are their low photochemical stability and limited light absorption range (Ferdianto and Rohmawati 2024). To overcome these issues, chemical structure modification through formation of metal-ligand complexes has proven to be an effective approach.

Metals such as zinc (Zn) and copper (Cu) have strong coordination abilities with functional groups found in organic molecules such as carbonyl, hydroxyl, and amine groups present in natural pigments. Formation of these metal-ligand complexes enhances molecular stability, broadens light absorption range, and accelerates electron transfer to semiconductor (Govindan et al. 2020). Moreover, Zn and Cu are abundant, non-toxic, and environmentally friendly metals, aligning well with the principles of green chemistry. In the study by Mohamad et al. (2022), the use of black grape peel containing anthocyanin pigment as a sensitizer with ZnO as a photoanode achieved an efficiency of up to 3.63%. Meanwhile Puspitasari et al. (2023), reported that Cu ion doping in natural anthocyanin dye for DSSC, with a methanol:acid ratio of 3:0, resulted in an efficiency of 0.016% in the Cu-doped natural dye system.

Nanotechnology-based approaches in photosensitizer development play an important role in improving DSSC efficiency. Nanometer-scale particles increase the active surface area, enhancing the interaction between photosensitizer and the semiconductor (Sova and Setiarso 2021). Complexes of Zn-*Catharanthus roseus* and Cu-*Catharanthus roseus* have potential to produce nano-photosensitizers with improved optoelectronic performance and sustainability. Development of natural nano-photosensitizers based on metal-pigment complexes not only aims to improve DSSC energy conversion efficiency but also supports the advancement of eco-friendly energy materials (Sukmawati et al. 2024). The use of renewable bio-based natural materials and non-toxic metals represents a concrete contribution to sustainable energy technology innovation, in line with the Sustainable Development Goals (SDGs), particularly in the field of clean and environmentally friendly energy.

Therefore, this study aims to develop environmentally friendly nano-photosensitizers from Zn-*Catharanthus roseus* and Cu-*Catharanthus roseus* complexes for application in Dye-Sensitized Solar Cells. Through optical, structural, and photovoltaic characterizations, this research seeks to provide scientific insights into the potential of metal–natural pigment complexes in enhancing solar cell efficiency. The results are expected to serve as an initial step toward integrating natural materials into renewable energy technologies that are efficient, affordable, and environmentally sustainable.

Materials And Methods

Study area

Research was conducted at Pharmacy Laboratory of Nahdlatul Ulama Sunan Giri University, Bojonegoro. Equipment used included a rotary evaporator, beaker glass, dropper pipette, stirrer, ultrasonic water bath, hotplate, DC power supply, digital multimeter, lux meter, halogen lamp, and XRD instrument. The materials used were *Catharanthus roseus* leaves, distilled water, ethanol, $ZnSO_4$, $CuSO_4$, $H_2PtCl_6{\cdot}6H_2O$, platinum metal, TiO_2, HCl, NaOH, and conductive FTO glass.

Procedures

Sample Preparation and Extraction of Catharanthus roseus

Fresh Catharanthus roseus leaves weighing 2 kg were used as the raw material in this study. Leaves were washed thoroughly to remove any impurities and then dried in an oven at 50°C until moisture content was reduced. Once dried, simplicia were ground using a blender into a fine powder to facilitate the extraction process. Extraction was carried out using ultrasonic method, where 200 g of powdered simplicia were placed in a beaker and mixed with 500 mL of ethanol. Mixture was sonicated for 1 hour. Resulting extract was then concentrated using a rotary evaporator, and any remaining solvent was removed with a water bath, yielding a thick extract of Catharanthus roseus leaves.

Synthesis of Zn and Cu Nanoparticles from Catharanthus roseus

Stock solutions of 0.02 mol/L $ZnSO_4$ and 0.02 mol/L $CuSO_4$ were prepared by dissolving 0.48 g of each salt in 150 mL of distilled water. Then, 50 mL of each stock solution was transferred into separate beakers. Subsequently, 20 mL of Catharanthus roseus leaf extract with a 10% concentration was added to each beaker. Solutions were stirred continuously with a magnetic stirrer at a constant speed while being slowly mixed. To prepare the extract solution, 4 g of Catharanthus roseus leaf powder was dissolved in 20 mL of distilled water. Mixture was stirred for 1.5 hours at room temperature using a magnetic stirrer. Then, 0.2 mol/L sodium hydroxide (NaOH) solution was slowly added until pH reached 8. Stirring was continued for another 3.5 hours to ensure complete reaction. Resulting mixture was centrifuged at 4000 rpm for 5 minutes to separate precipitate from the supernatant. Obtained precipitate was dried at 70°C until completely dry.

Coating of TiO_2 Working Electrode

TiO_2 coating on the working electrode was prepared on a cleaned conductive FTO glass, which had been washed with ethanol and treated in an ultrasonic cleaner. TiO_2 paste was made by mixing TiO_2 powder with a solvent and a binder, then applied onto the FTO substrate surface within a 0.5 × 0.5 cm tape-defined area. Coating was performed using the doctor blade technique with a glass rod. Coated substrate was then dried at 80°C to remove the solvent, followed by sintering at 400–500°C to form a strong and homogeneous TiO_2 layer.

Preparation of Pt Counter Electrode

Electrolyte solution was prepared by dissolving 0.065 g of $H_2PtCl_6 \cdot 6H_2O$ powder in a small amount of distilled water. Then, 10 mL of 1 M HCl was added, and the total volume was adjusted to 100 mL with deionized water while stirring until homogeneous. Platinum coating process was carried out on conductive FTO glass (Fluorine-doped Tin Oxide), as shown in Figure 1. In this setup, FTO substrate served as the working electrode connected to negative terminal of power supply, while platinum metal was connected to the positive terminal. Distance between two electrodes was maintained at 2 cm. Platinum deposition was carried out for 30 minutes at a current of 15 mA, solution temperature of 40°C, and a maximum voltage of 0.755 V.

Figure 1. Schematic of FTO Glass Coating with Pt

Testing of DSSC Solar Cells

Each Zn and Cu nanoparticle sample from Catharanthus roseus extract was weighed at 0.0015 g. Working electrode was paired with the counter electrode and clamped together using a binder clip. TiO_2 semiconductor layer on the working electrode was dropped with an electrolyte solution in gap between the electrodes. Efficiency of the DSSC solar cell was measured using a static resistor to obtain current–voltage data at various resistance values. Resistance values used were 0, 2.2, 3.3, 4.7, 10, 22, 47, 100, 220, 470, 800, and ∞ ohm. Test was conducted under halogen lamp illumination with an intensity of 180,000 lux, measured at exact position of the solar cell using a lux meter.

Data analysis

Measurement of DSSC Solar Cell Efficiency

Maximum current and voltage data obtained were used to calculate the fill factor (FF) value according to Equation 1.

$$FF = \frac{Vmpp}{Voc} \times \frac{Impp}{Isc} \quad (1)$$

Meanwhile, to measure efficiency, use equation 2:

$$\eta = \frac{Voc \times Isc \times FF}{PPchy} \quad (2)$$

Description

Vmpp : maximum cell voltage
Impp : maximum cell current
Voc : open-circuit cell potential
Isc : closed-circuit cell current
Pchy : light power delivered to the cell

XRD Characterization

Crystal structure of Zn and Cu nanoparticles from *Catharanthus roseus* was analyzed using X-Ray Diffraction (XRD) method. XRD characterization was performed to determine the crystal structure, crystallite size, and phase purity of the Zn and Cu nanoparticles. By analyzing obtained diffraction patterns, the type and arrangement of crystal lattice formed in nanoparticles could be identified. In addition, the XRD results also helped determine whether synthesized nanoparticles were amorphous or crystalline and ensured that no impurity phases or other compounds were formed during the synthesis process.

Results And Discussion

Result of TiO_2 Working Electrode

Coating of the TiO_2 working electrode produced results as shown in Figure 2, which illustrates that TiO_2 layer appeared pure white and adhered well to active surface of conductive FTO glass.

Figure 2. Result of TiO_2 Working Electrode Coating

Result of Pt Counter Electrode

Coating of the Pt counter electrode produced results as shown in Figure 3, which displays the deposition of Pt on FTO glass characterized by typical metallic silver-gray color of Pt, adhering well to the active surface of conductive FTO glass.

Figure 3. Result of Pt Counter Electrode Coating

Result of DSSC Solar Cell Efficiency Measurement

Efficiency measurement of DSSC solar cells using Zn and Cu nanoparticles from *Catharanthus roseus* was carried out by calculating the fill factor (FF) value. Measurement results for the DSSC with nanoparticles of Zn-*Catharanthus roseus* are shown in Table 1, with an FF value of 0.2031 and an efficiency of 4.97×10^{-3}. Meanwhile, DSSC with nanoparticles of Cu- *Catharanthus roseus* showed an FF value of 0.045 and an efficiency of 2.6×10^{-4}, as presented in Table 2.

Table 1. Measurement Results of DSSC Solar Cell Efficiency Using Zn Nanoparticles from *Catharanthus roseus.*

Potential (mV)	Current (μA)	Power (nW)	FF	η (%)
0	41,4	0		
1,4	42,0	58,8		
2,3	32,8	75,44		
3,2	36,1	115,52		
2,0	43,1	86,2		
3,2	35,8	114,56	0,2031	$4{,}97 \times 10^{-3}$
4,9	29,6	145,04		
6,2	45,6	282,72		
9,1	32,8	298,48		
23,8	14,3	340,34		
46,6	9,6	447,36		
53,2	0	0		

Table 2. Measurement Results of DSSC Solar Cell Efficiency Using Cu Nanoparticles from *Catharanthus roseus.*

Potential (mV)	Current (μA)	Power (nW)	FF	η (%)
0,0	21,3	0,00		
0,4	11,2	4,48		
1,6	5,4	8,64		
2,2	2,9	6,38		
1,4	2,1	2,94		
1,9	1,8	3,42	0,045	$2{,}6 \times 10^{-4}$
1,6	1,9	3,04		
1,3	1,9	2,47		
1,6	1,5	2,40		
23,7	1,0	23,70		
24,3	0,3	7,29		
24,6	0,0	0,00		

Result of XRD Characterization

Crystal structure of Zn and Cu nanoparticles synthesized using *Catharanthus roseus* leaf extract was analyzed using XRD technique, and resulting diffractogram is shown in Figure 4.

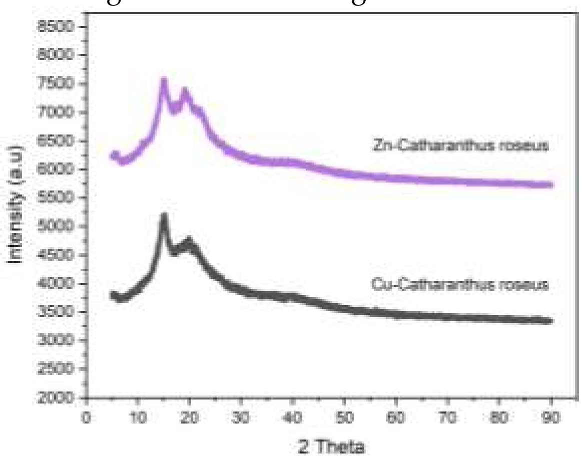

Figure 4. XRD Diffractogram Pattern of Zn and Cu Nanoparticles from *Catharanthus roseus*

Discussion

Coating result of TiO_2 working electrode, as shown in Figure 2, demonstrates that deposition process was successful. TiO_2 layer functions as a semiconductor in the DSSC, acting as an electron acceptor injected by the photosensitizer. In addition, TiO_2 serves as an anchoring medium for dye molecules that absorb light, generate electrons, and transfer them to the conductive FTO glass, thus producing electrical energy (Aprillia and Haris 2016).

Result of the Pt counter electrode coating, as shown in Figure 3, also indicates a proper and uniform deposition. Coating process occurs through a redox reaction, in which Pt ions from the H_2PtCl_6 electrolyte solution undergo reduction and subsequently deposit on conductive FTO surface via electroplating method. During this process, electrolyte solution acts as both an electric current conductor and a source of Pt metal ions that are deposited onto electrode surface (cathode) (Winata, Fikri, and Mujirudin 2022).

DSSC solar cell performance test produced efficiency values as presented in Table 1 for Zn-*Catharanthus roseus* nanoparticles and Table 2 for Cu-*Catharanthus roseus* nanoparticles. Highest efficiency was obtained for Zn-*Catharanthus roseus* nanoparticles, which reached 4.97×10^{-3}, while Cu–*Catharanthus roseus* nanoparticles showed a lower efficiency of 2.6×10^{-4}. These results indicate that both Zn and Cu nanoparticles derived from *Catharanthus roseus* can generate electric current in DSSC systems due to continuous electron transfer. This process occurs when light irradiates semiconductor surface bound to Zn and Cu nanoparticles, causing molecules to become excited from their ground state to a higher energy level. The excited electrons then possess sufficient energy to be injected into conduction band of the semiconductor. This electron injection process occurs due to the interaction between Zn and Cu nanoparticles and TiO_2 through bonding formed between them (Hatib, Anwar, and Soso 2024).

Characterization of Zn and Cu nanoparticles synthesized from *Catharanthus roseus* was performed using XRD, as shown in Figure 4. Phase identification was carried out by matching diffraction peak positions with reference data from Joint Committee on Powder Diffraction Standards (JCPDS), specifically JCPDS No. 01-070-1255 for Zn and JCPDS No. 01-077-1900 for Cu. Zn–*Catharanthus roseus* nanoparticles exhibited a diffraction peak at 2θ around 21.2° with low intensity, indicating a cubic crystal structure. Meanwhile, Cu–*Catharanthus roseus* nanoparticles displayed main peaks at 2θ around 15.2° and 19.1° with moderate intensity, suggesting an anorthic crystal structure. Observed peak shifts and broadening in diffraction pattern indicate that resulting particles are nanosized (Ali, Chiang, and Santos 2022). Furthermore, all samples showed high-intensity peaks around 15°, corresponding to crystalline structure of *Catharanthus roseus* bonded with Zn and Cu atoms. Sharp and intense peaks suggest that the synthesized materials possess good crystallinity.

Conclusions

Conclusion of this study is that Zn and Cu nanoparticles derived from *Catharanthus roseus* are capable of producing natural pigments with potential as environmentally friendly photosensitizers. The highest DSSC solar cell efficiency was obtained from Zn-*Catharanthus roseus* nanoparticles, with a value of 4.97×10^{-3}, while the Cu-*Catharanthus roseus* nanoparticles showed a lower efficiency of 2.6×10^{-4}.

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Conflict of Interest: Authors declare that there is no conflict of interest related to the research or publication of this article.

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