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Optimization of Silver *Doping* on Natural Dye Mixed Chlorophyll-Anthocyanin on The Efficiency of *Dye Sensitized Solar Cells* (DSSC)

Desita Suci Pangesti, Fahru Nurosyid*, Risa Suryana, Yofentina Iriani

Physics Department, Faculty of Mathematics and Natural Sciences, Universitas Sebelas Maret, Surakarta 57126, Indonesia

*Corresponding author email: fahrunurosyid@staff.uns.ac.id

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ABSTRACT. This study aimed to incorporate silver (Ag) in the mixture of chlorophyll-anthocyanin dyes to improve DSSC efficiency. This research evaluated the impacts of various natural dye modifications with different concentrations of Ag incorporation on the DSSC performance. Here, anthocyanin (A) was extracted from butterfly pea flowers, while chlorophyll (C) was extracted from spinach leaves. The two dyes were mixed with a volume ratio of (anthocyanin: chlorophyll) 1:3. From the ratio of anthocyanin: chlorophyll, the extraction of anthocyanin solution is 25% and chlorophyll is 75%. Ag was incorporated into the dye mixture (AC) with distinct concentrations of 0%, 3%, and 5%. The dyes were characterized using a UV-Vis spectrophotometer and Keithley I-V meter to investigate their optical and conductivity properties. The DSSC performance was assessed with a Keithley I-V meter. The yields exhibited that the Ag incorporation improved the absorbances and widened the absorption area of the AC dyes as compared to the single and mixed dyes without Ag addition. Further, the results demonstrated that Ag incorporation enhanced the conductivity of the AC dyes, showing that the values were 4.43×10^{-2} $(\Omega m)^{-1}$, 6.46×10^{-2} $(\Omega m)^{-1}$, and $8{,}21\times10^{-2}$ $(\Omega m)^{-1}$ for 0%, 3%, and 5% Ag incorporation. Moreover, the conversion efficiency values obtained by DSSCs prepared with AC dyes, Ag-incorporated AC 3%, and Ag-incorporated AC 5% were 0.073%, 0.209%, and 0.111%, respectively that Ag incorporation could significantly advance the DSSC performance. This study found that the AC dyes incorporated with 3% Ag were he optimal composition to produce the optimal DSSC performance.

Keywords: anthocyanin, chlorophyll, DSSC, dye, silver

INTRODUCTION

The need for energy continues to escalate along with the increasing population, causing a crisis in energy sources. The search for economical and environmentally friendly energy is growing towards renewable sources. One alternative energy source currently being developed is solar energy, which can be converted into electricity using solar cells. The development of solar cells is divided into three generations, where dye-based solar cells or Dye-Sensitized Solar Cells (DSSC) are the third one (Jariah et al., 2023).

DSSC is a photovoltaic device comprised of semiconductor working electrodes, counter electrodes, electrolytes, and dyes. One of the factors that affects the efficiency of DSSC is the absorption ability of the dye and the band gap of the semiconductor. The dye contained in the DSSC serves to provide a wider range of photon absorption from sunlight (Hariningtias & Setiarso., 2021).

There are two types of dyes commonly used in the manufacture of DSSC, namely synthetic dyes and natural dyes. Synthetic dyes can produce high efficiency and have good photochemical stability. However, they can pollute the environment, and they have expensive production costs, so natural dyes can be an alternative to overcome these problems. Natural dyes have abundant availability, affordable production, simple fabrication, and are environmentally friendly (Syafinar et al., 2015).

The ability of the dye to absorb light at certain wavelengths is a crucial factor affecting the efficiency of DSSCs. However, one of the main challenges in using natural dyes is the low efficiency of DSSCs due to their less effective absorption. Thus, the characteristics of the natural dyes need to be improved to enhance the efficiency of DSSCs. Several types of natural dyes can be mixed to expand the range of visible light absorption so as to increase the excitation of electrons transmitted to TiO_2 and increase the current in DSSC. Besides, incorporating metals into the dyes can be done to widen the range of absorption regions (Pramananda et al., 2021).

Efforts to increase light absorption can be made by sensitizing semiconductors with transition metal complexes. Transition metal complexes that can act as photosensitizers should have the features of low oxidation numbers, low electronegativity, and excess electron density (Jariah et al., 2024). Incorporating silver (Ag) in the dyes can increase the number of free charge carriers, which directly improves the electrical conductivity. Silver has high electrical conductivity, so when added to the dyes, it can enhance the electron conduction path and accelerate the transfer of electrons in the solar cell (Reynolds et al., 2017). Based on previous research, the use of spinach leaf extract as a dye in DSSC has been found to have a high pigment content, which enhances its efficiency. Anthocyanin in DSSC can absorb sunlight across various wavelengths, particularly in the blue and green spectrum, allowing DSSC to capture a significant portion of available sunlight. Additionally, using a mixed dye system is more efficient than a single dye, and the addition of silver (Ag) doping can improve the conductivity of DSSC. Therefore, this study combines two natural dyes, including anthocyanin from butterfly pea flower extract and chlorophyll from spinach leaf extract, along with silver (Ag) doping to enhance DSSC performance (Mahajan et al., 2024).

In the *I-V* characteristics of solar modules, short circuit print current (I_{sc}) occurs at zero volts, while zero current occurs at open circuit voltage (V_{oc}). Solar modules achieve optimal performance when the *I-V* characteristics are at the maximum power point (I_{mp}, V_{mp}). Under normal conditions, solar modules are capable of producing maximum power when the *I-V* characteristics reach that maximum power point (Amna dkk., 2021). Performance evaluation based on current density and voltage measurements. I_{sc} indicates the ability of the dye to absorb sunlight energy and convert it into electrical energy. In the visible region of the solar spectrum, the amount of accumulated sunlight is represented by I_{sc}. The higher the absorption of sunlight by the dye molecules, the higher the Isc, so that the energy conversion efficiency of the device is high (Ezike et al., 2021).

This study aimed to incorporate silver (Ag) in the mixture of chlorophyll-anthocyanin dyes to improve DSSC efficiency. This research also evaluated the impacts of the dye modifications with different concentrations of Ag incorporation on the DSSC performance. Further, this study sought to find the optimal compositions of silver and dye mixtures that revealed the optimum DSSC efficiency. The main focus of the research is to obtain the absorbance value, electrical conductivity, and the effect of silver doping on DSSC performance efficiency.

EXPERIMENTAL SECTION

Preparation of Natural Dyes

One of the solute separation techniques often used is extraction. In this study, chlorophyll (C) was extracted from spinach leaf extraction, while anthocyanin (A) was obtained from butterfly pea leaf extraction. Five grams of spinach leaves were dissolved in 100 mL of ethanol and stirred using a magnetic stirrer at 300 rpm for 2 hours. Separately, two grams of butterfly pea leaf flowers were dissolved in 25 mL of methanol, 21 mL of distilled water, and 4 mL of acetic acid and stirred on a hotplate at 300 rpm at 30°C for 4 hours. The two solutions were then left for 24 hours, called the maceration process. After the process, the solutions were filtered using filter paper to obtain dyes and covered with aluminum foil to prevent evaporation.

$AgNO_3$ Incorporation in Natural Dyes

Anthocyanin and chlorophyll dyes obtained were mixed with a volume ratio of 1:3 mL (called AC dyes). The incorporation of Ag was done by mixing the powder with the dye solution. The weights of $AgNO_3$ were varied at 0%, 3%, and 5% of the total volume of the AC dyes. The powders were then diluted in the AC dyes and stirred with a magnetic stirrer at 300 rpm for 2 hours.

Preparation of Working Electrodes

The working electrodes of the prepared DSSCs in this study were made of FTO glass substrates coated with TiO_2. First, 2 grams of TiO_2 gel were mixed with 4 mL of ethanol and stirred using a magnetic stirrer for 4 hours, producing TiO_2 paste. Simultaneously, the FTO substrates were sterilized with an ultrasonic cleaner for 30 minutes, followed by heating at 100°C for 10 minutes on a hotplate. The TiO_2 paste was then deposited on the substrates using a spin coater. After that, the TiO_2-coated FTO glasses were immersed in the prepared dyes, including chlorophyll, anthocyanin, anthocyanin-chlorophyll mixture, Ag-incorporated AC 3%, and Ag-incorporated AC 5% dyes, for 24 hours.

Preparation of Counter Electrodes

The counter electrodes were made of FTO glass substrates coated with platinum (Pt). Before this, FTO glass substrates were cleaned using the procedures mentioned above. The platinum paste was then coated on the FTO glasses using a spin coater. The samples were then annealed at 450 °C for 30 minutes.

DSSCs Assembly

Figure 1. presents the DSSC structures assembled in this study. The working and counter electrodes were stacked like a sandwich structure and glued using a clip. The electrolyte solution was then injected between the two electrodes.

Characterizations of Optical and Conductivity Properties

A UV-Vis spectrophotometer was used to observe the optical properties of the dyes prepared in this study. The form of characterization is the result of extraction from spinach leaves and telang flowers, which are then doped with the addition of Ag metal in the form of a solution. The results of the sample test using a *UV-Visible* Spectrophotometer will be presented in the form of a graph containing information on the relationship between wavelength and absorbance value. A Keithley I-V Meter tool was used to assess the conductivity of the prepared dyes.

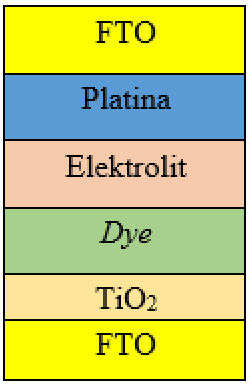

Figure 1. DSSC sandwich structure

Evaluation of the DSSC Performance

The prepared DSSCs were tested using a Keithley I-V meter to determine the efficiencies under a Halogen lamp with an intensity of 1000 W/m². The results were current-voltage (I-V) curves containing V_{oc}, I_{sc}, and power data. In the *I-V* characteristics, short circuit print current (I_{sc}) occurs at zero volts, while zero current occurs at open circuit voltage (V_{oc}). These parameters were then exploited to determine the fill factor (FF) values and further calculate the DSSC efficiencies, as Equations 3 - 6 (Ezike et al., 2021).

$$FF = \frac{I_{max} V_{\max}}{I_{SC} V_{oc}} \quad (3)$$

$$P_{max} = I_{sc} V_{oc} FF \quad (4)$$

$$\eta = \frac{P_{out}}{P_{input}} \; x\, 100\% \quad (5)$$

$$\eta = \frac{I_{sc} x V_{oc} x FF}{P_{input}} \; x\, 100\% \quad (6)$$

Where FF is fill factor, I_{max} is maximum current density (mA.Cm^{-2}), V_{max} is maximum voltage (mV), I_{sc} is Short circuit current density (mA.Cm^{-2}), V_{oc} is open circuit voltage (mV), P_{out} is output power (mW.cm^{-2}),P_{input} is input power (mW.cm^{-2}), and η is DSSC efficiency (%).

RESULTS AND DISCUSSION

Optical Properties of the Prepared Dyes

The volume of the extracted solution containing chlorophyll was 100 mL, as all the dissolved chlorophyll was present in the 100 mL ethanol solvent. The absorbance spectra of chlorophyll (C), anthocyanin (A), AC, Ag-doped AC 3%, and Ag-doped AC 5% were depicted in **Figure 2.** Based on the figure, chlorophyll dye prepared from spinach leaves could absorb light in the wavelength range of 400-450 nm (blue band) and 640-680 nm (red band). Chlorophyll pigment can be used as a sensitizer dye in DSSC because of its ability to absorb sunlight photons and had an invisible light spectrum that can be harvested and proven by testing the observation rate using a UV-Vis spectrophotometer. The peaks identified at 413 and 668 nm are considered characteristic peaks of chlorophyll-a, while the other two peaks observed are associated with characteristic peaks of carotenoids (Kaliramna et al., 2022). Further, the figure reveals that anthocyanin obtained from butterfly pea flower extract could absorb light in the wavelength range of 450-550 nm (blue band) and 500-620 nm (green to red band).

Further, the dye mixture (AC) exhibited a similar absorbance curve to the chlorophyll dye with an increased absorbance intensity. **Figure 2** showed the impact of silver incorporation on the optical characteristics of the AC mixed dyes. It revealed that Ag incorporation caused the absorbance intensities of the AC dyes to improve and the absorbance area to widen. Ag-incorporated AC 3% had a maximum absorbance value of 3.279 a.u at 660 nm. The absorbance value increased compared to the pure AC because the interaction between Ag^+ ions and dye molecules enhanced the molar absorptivity coefficient (ε) or causes more dye molecules to contribute to the absorbance.

At Ag-incorporated AC 5%, the maximum absorbance value increased to 3.117 at 664 nm. It demonstrates that the absorbance slightly declined compared to 3% Ag incorporation. Basically, almost all types of metals cannot absorb light properly; thus, the addition of Ag metal causes a decreased dye absorption in the light given. Another thing that explains the phenomenon of reducing absorbance value is due to the hypochromic effect (Suhartati., 2017). The hypochromic impact not only caused the decreased absorbance value of a solution but also shift the wavelength absorption, as occurs in the 3% and 5% Ag addition. The declined absorbance value can be due to the aggregation of dye molecules or quenching by Ag^+ ions. In this case, although the dye concentration is fixed, the effectiveness of c that contributes to absorbance is reduced because not all dye molecules are in an effective state of absorbing light. Aggregation reduces the number of individual dye molecules that can absorb light, so the effective absorptivity coefficient (ε) is reduced, resulting in a declined absorbance. Dye aggregation is influenced by the structure of the dye, dye solution concentration, temperature, solvent, and so on. This occurs in the sensitization process, where the dye aggregates in solution (caused by interactions between dye molecules and/or other molecules) and then adsorbs onto the metal oxide semiconductor. The dye aggregates after being adsorbed onto the metal oxide semiconductor surface to form a dye/metal oxide semiconductor interface. However, dye aggregation usually causes quenching of the molecular excited state and unfavorable back electron transfer, which affects the electron injection efficiency and subsequently reduces the efficiency. Additionally, H-aggregation causes a blue-shift in the absorption spectrum, which is not conducive to improving light absorption capacity (Fang Xu et al., 2020).

Besides, the cause of this occurrence was that metals do not match for light absorption. Still, the addition of metals can attach their properties to dyes, thereby affecting the conductivity properties of the prepared samples. This statement is also supported by Supriyanto et al. (2021), reporting that the high conductivity value of the metal caused the decreased Ag metal uptake.

Electrical Conductivity of Ag-Incorporated AC Dyes

The current-voltage curves of the AC dyes with and without Ag incorporation are presented in **Figure 3.** It reveals that the more Ag added the higher the currents the voltages are given. The gradient or resistivity (R) values obtained from the graphs could be used to calculate the conductivity values, as listed in Table 1 (Faqih et al., 2020).

Figure 3 showed that the graph produced by the Ag doping solutions of 0%, 3%, and 5% produces an ideal linear graph and indicate that the resistivity value is small and the conductivity value is high**. Table 1**. shows that the single chlorophyll and anthocyanin had conductivity values of 0.64×10^{-2} $(\Omega m)^{-1}$ and 2.23×10^{-2} $(\Omega m)^{-1}$, respectively. Meanwhile, the conductivity value of the mixed AC dye was enhanced to 4.43×10^{-2} $(\Omega m)^{-1}$. The Ag incorporation at 3% and 5% levels further could significantly improve the conductivity values to 6.46×10^{-2} $(\Omega m)^{-1}$ and 8.21×10^{-2} $(\Omega m)^{-1}$.

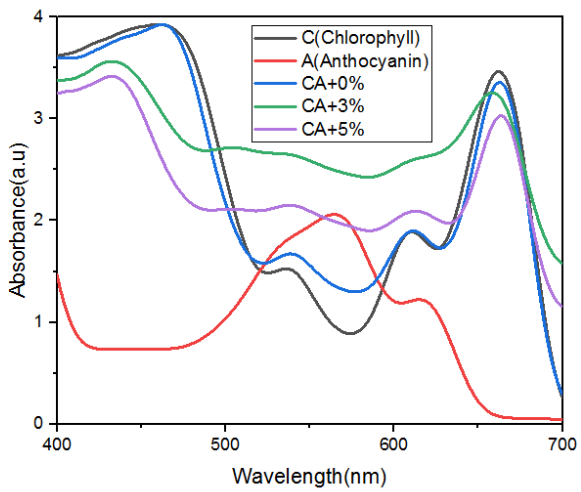

Figure 2. Absorbance spectra of dye, mixed, and Ag-incorporated dyes

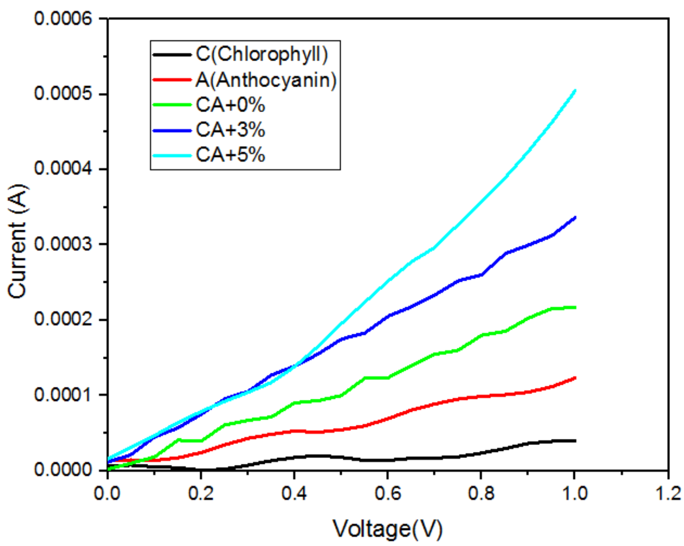

Figure 3. I-V curves of dye, mixed, and Ag-incorporated dyes

Table 1. Conductivity values of single, mixed, and Ag-incorporated dyes

Samples	Resistivity (Ωm)	Conductivity $(\Omega m)^{-1}$
C (Chlorophyll)	154.78	0.64×10^{-2}
A (Anthocyanin)	44.79	2.23×10^{-2}
AC	22.52	4.43×10^{-2}
Ag-incorporated AC 3%	15.46	6.46×10^{-2}
Ag-incorporated AC 5%	12.17	8.21×10^{-2}

Hence, the results prove that the addition of metal to the dyes can enhanced their conductivity value. The improved conductivity is due to the influence of the basic nature of the metal itself. Silver incorporation escalates the number of free charge carriers in the dyes, which directly advances the electrical conductivity. Silver has high electrical conductivity, so when added to the dye, it can improve the electron conduction path and accelerate electron transfer in the solar cell (Reynolds et al., 2017) (Yadav et al., 2022).

Efficiency of DSSCs

Current-voltage testing was carried out in bright conditions where the DSSC deviced was directly illuminated by a lamp with an intensity of 1000 W/m². The current and voltage data that have been obtained through the Keithley I-V Meter tool then sought for their efficiency value using equation 6.

DSSCs were characterized using the Keithley I-V Meter with an active area of 0.025 m². **Figure 4.** showed the I-V curves of DSSCs composed of single, mixed, and Ag-incorporated dyes, where the efficiency values were presented in **Table 2**. It revealed that the conversion efficiency values of DSSCs generated from the single chlorophyll and anthocyanin dyes were 0.031% and 0.045%, respectively. Meanwhile, the efficiencies of the mixed AC dyes improved to 0.073%. The incorporation of 3% Ag further advanced the efficiency up to 0.209%. Nevertheless, the efficiency slightly declined to 0.113% with the incorporation of 5% Ag. At 5% doping, the V_{oc} and I_{sc} values decrease compared to 3% doping, resulting in lower efficiency. The decrease in V_{oc} and I_{sc} values at 5% doping is due to the higher Ag concentration, which blocks the active surface area of the DSSC. Ag becomes a center for electron-hole recombination, leading to a decrease in V_{oc} and I_{sc}, which in turn lowers the DSSC efficiency (Kalimrana et al., 2022).

The efficiency value of DSSC doping variations of 0%, 3% and 5% increases because it is influenced by the value of V_{oc} and I_{sc}. V_{oc} values of 0%, 3% and 5% doping variations were 0.500 V; 0.45 V; 0.530 V, while I_{sc} values of 0%, 3% and 5% doping variations are 1.29×10^{-4} A; 1.56×10^{-4} A; 0.79×10^{-4} A, respectively. The dye mixture with 0%, 3% and 5% doping variations shows an increase in V_{oc} and I_{sc} values. This makes the curve area on the current-voltage graph wider, so that it will affect the magnitude of I_{max} and V_{max} values and result in increased DSSC efficiency values. The results demonstrate that the Ag incorporation could enhance the efficiency of the DSSCs as compared to single and mixed dyes. The Ag incorporation influenced the values of V_{oc} and I_{sc}, thereby I_{max} and V_{max} values, resulting in increased DSSC efficiency.

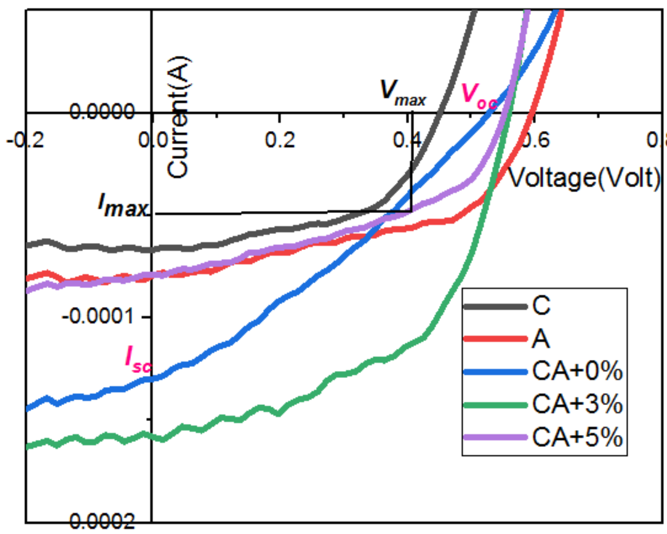

Figure 4. I-V curves of the prepared DSSCs with various dye modifications

Table 2. I-V characteristics of the prepared DSSCs

Sample	V_{oc} (V)	I_{sc} (A)	V_{max} (V)	I_{max} (A)	P_{max} (W)	FF	η (%)
C (Chlorophyll)	0.439	0.65×10^{-4}	0.166	0.47×10^{-4}	0.79×10^{-5}	0.274	0.031
A (Anthocyanin)	0.590	0.79×10^{-4}	0.469	0.24×10^{-4}	1.14×10^{-5}	0.244	0.045
AC	0.500	1.29×10^{-4}	0.167	1.09×10^{-4}	1.83×10^{-5}	0.284	0.073
Ag-incorporated AC 3%	0.545	1.56×10^{-4}	0.378	1.38×10^{-4}	5.24×10^{-5}	0.615	0.209
Ag-incorporated AC 5%	0.530	0.79×10^{-4}	0.242	1.17×10^{-4}	2.84×10^{-5}	0.677	0.113

The results of this study found that the incorporation of 3% Ag was the optimal number to obtain the optimal DSSC efficiency. The incorporation increases absorbance and conductivity. This allows efficient electron transfer from the dye to the electrode and reduces electron-hole recombination, resulting in the highest efficiency. Meanwhile, the incorporation of 5% Ag decreased the V_{oc} and I_{sc} values. This decrement are because the excessive Ag incorporation can block the surface of the active area of the DSSC so that Ag becomes the center of the recombination of electrons and holes, thereby reducing the DSSC efficiency (Kalimrana et al., 2022).

According to the results, the absorbance peaks of Ag-incorporated AC dyes were higher, and the absorption areas were wider than those of the single and mixed dyes without doping. However, the absorbance of the AC dyes with 5% Ag incorporation slightly decreased as compared to that of 3% Ag. This result was in contrast to the results of the conductivity values, where the conductivity of DSSC with 5% Ag was higher than that of 3% Ag. At 5% doping, although conductivity increases, the decrease in absorbance due to dye quenching and aggregation causes a decrease in overall efficiency. Electron transfer is suboptimal because most of the dye molecules are unavailable for electron excitation and transfer. This increase in conductivity makes it easier for the device to conduct electric current at an applied voltage.

Supriyanto et al. (2021) stated that the increasing and excessive Ag concentration will cause aggregation between the dye and Ag metal. The decreased DSSC efficiency value at 5% Ag is caused by increased electron-hole recombination and aggregation of silver nanoparticles. Besides, the excessive Ag incorporation can damage the dye structure, resulting in the disrupted interaction between dye and TiO_2 for electron transfer, non-optimal plasmonic effects, and increased internal resistance, thereby reducing the DSSC efficiency. However, overall, the Ag incorporation into the dyes causes an increase in DSSC efficiency (Gupta et al., 2016).

CONCLUSIONS

The incorporation of silver metal (Ag) into the mixture of chlorophyll and anthocyanin could improve the optical properties, conductivity, and efficiency of the prepared DSSCs. It revealed that the incorporation of Ag broadened the light absorption spectrum, optimized electron transfer, and enhanced the stability and overall efficiency of solar cells. The conductivity values of the AC dyes incorporated with 3% and 5% Ag increased as compared to the single and mixed dyes without Ag. This improvement is due to the natural conductive properties of Ag metal, which increases the number of free charge carriers and improves the electron conduction pathways in the dye, thereby increasing the efficiency of the DSSC. Further, the Ag incorporation could advance the energy conversion efficiency of the prepared DSSCs. The results revealed that the optimal efficiency of DSSC of 0.209 % was achieved by AC dyes incorporated with 3% Ag.

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