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The Effect of Copper-Silver (Cu-Ag) Double Doping on ZnO Photoanode on Dye Sensitized Solar Cell (DSSC) Efficiency

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Abstract

A DSSC has a good performance seen from the ability of the photoanode to absorb sunlight. The use of photoanodes uses a semiconductor material, namely ZnO. However, in use, ZnO has the disadvantage of low optical and electrical properties, which results in low DSSC efficiency. So in this research, efforts are made to add Cu-Ag double doping to improve optical and electrical properties that will increase the efficiency of DSSC. ZnO/Cu-Ag photoanodes were prepared using the sol-gel spin coating method with doping variations of 0%, 1%, 2%, and 3%. Characterization was carried out using a UV-Vis spectrophotometer to see the absorption area and determine the band gap energy of the ZnO photoanode. I-V Keithley was used for current and voltage characterization to determine the conductivity of the ZnO photoanode and the efficiency of the DSSC. The results of the research show the effect of increasing the optical properties of ZnO photoanodes by decreasing the band gap energy along with the increase in doping concentration, namely 3.29 eV, 3.24 eV, 2.44 eV, and 2.40 eV. In addition, there is an increase in conductivity on the ZnO photoanode with the highest conductivity at doping concentration of 3% which is 0.1006 $\Omega^{-1}m^{-1}$ with an increase of 6 times compared to the pure ZnO photoanode of 0.0163 $\Omega^{-1}m^{-1}$. The improved optical and electrical properties of ZnO/Cu-Ag photoanodes have an impact on increasing the efficiency value of DSSC with the optimum efficiency achieved at 1% doping concentration of 0.0291% with an increase of 16 times compared to the use of pure ZnO photoanode of 0.00018%. The addition of Cu-Ag doping to ZnO improves the optical and electrical properties of the photoanode, enhancing DSSC efficiency with an optimum at 1% doping concentration.

Keywords: *Copper; Silver; Double Doping; ZnO Photoanode; DSSC Efficiency*

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INTRODUCTION

A dye Sensitized Solar Cell (DSSC) is the third generation of solar cells as a renewable energy device that can convert solar energy into electrical energy [1,2]. DSSC is attractive for development due to its low cost, easy fabrication, environmental friendliness, and can be an alternative to

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conventional silicon based solar cells [3–5]. A DSSC has a good performance seen from its efficiency level determined by the ability of the photoanode to absorb sunlight so that the dye and electron transfer can achieve the best performance [6].

Generally, DSSC photoanode uses semiconductor materials, one of which is ZnO. ZnO attracts the attention of researchers because it has large electron mobility, low resistance, can reduce the recombination rate, and has a suitable light-capturing character in DSSC application [7–9]. However, ZnO also has a weakness of low optical and electrical properties that results in low efficiency of DSSC [10]. This can be resolved by doping efforts to adjust the optical and electrical properties of ZnO, also expanding the absorption of solar energy to increase the electron transfer rate to increase the efficiency of DSSC [11]. Mehmood et al. [6] showed an increased efficiency of DSSC from the combination of two types of transition metals, titanium (Ti) and copper (Cu) doped on ZnO, with the highest efficiency reaching 2.38%. These results underlie this research to modify the combination of two types of dopants, namely copper (Cu) and silver (Ag) in ZnO.

The combination of two doping copper (Cu) and silver (Ag) on ZnO is done to complement each other's deficiencies in carrying electric charges to optimize the performance of the photoanode, which will increase the efficiency of the DSSC. It is because the transition metal types copper (Cu) and silver (Ag) have high conductivity, can suppress the ZnO recombination rate, increase the rate of electron injection into the conduction band, and can stimulate the interface electron transfer process [12–16]. Research by Alqadi et al. [17] and Kant et al [18] were able to show the effect of a combination of copper (Cu) and silver (Ag) dopants on improving the electrical properties and optical absorption of ZnO so that ZnO can be applied as a photovoltaic device, while Windayani et al. [19] found that adding Ag to ZnO:Cu enhanced DSSC efficiency, though not surpassing the efficiency without Ag doping. This highlights the need for further exploration of Cu-Ag doping combinations for DSSC optimization by combining both doping together.

This research focuses on improving the optical and electrical properties with a combination of copper (Cu) and silver (Ag) doping on ZnO photoanode in DSSC applications. The improvement of optical and electrical properties can have an impact on the high efficiency of DSSC. In this research, a comparison was also made between the results of absorption, conductivity, and efficiency of DSSC with double-doped ZnO photoanode of copper (Cu) and silver (Ag) so that the optimum doping concentration can be obtained for the photovoltaic properties of DSSC.

METHOD

The ZnO/Cu-Ag photoanodes were synthesized by sol-gel spin coating method. The materials used in this research include CuSO4.5H2O (Merck, 99%), AgNO3 (Merck, 99,8%), Zn(CH3COOH)2.2H2O (Merck, 99%). Cu doping was obtained from CuSO4.5H2O dissolved in ethanol, acetic acid, and distilled water. Meanwhile, Ag doping was obtained from AgNO3 dissolved in ammonia and ethanol. ZnO/Cu-Ag photoanode solution was prepared by dissolving Zn(CH3COOH)2.2H2O into 2-propanol at 70 ^{0}C – 85 ^{0}C for 15 minutes. After 15 minutes, diethanolamine (DEA) and Cu-Ag doping was added with concentration of each 0%, 1%, 2%, and 3% until a homogenous sol solution formed. Dye was obtained from a mixed anthocyanin-chlorophyll with a ratio 2:1. Electrolyte solution was prepared from a mixture of KI and I2.

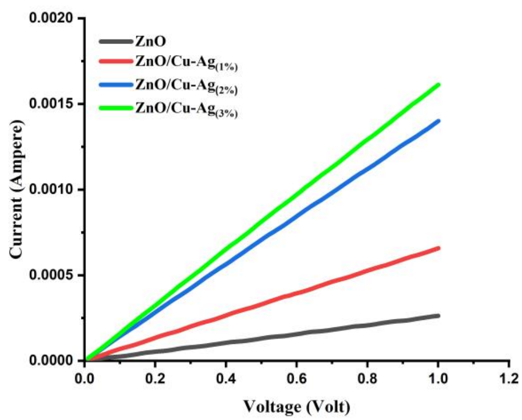
The working electrode was deposited from the ZnO/Cu-Ag photoanode solution on FTO glass with an active area 1 x 1 cm 3 times using spin coating method. Then, an annealing process was carried out using a furnace at 250 ^{0}C for 60 minutes. The working electrode is then immersed in the dye mixture solution for 24 hours until dye is absorbed into the ZnO. The counter electrode was prepared by depositing a platinum paste using the brush painting method on the FTO glass, placed on the hotplate at 200 ^{0}C for 15 minutes, then continued annealing process at 300 ^{0}C fot 15 minutes.

Characterization of the optical properties of ZnO/Cu-Ag photoanode using UV-Vis spectrophotometer at a wavelength of 300 – 800 nm to determine the absorption and band gap energy of ZnO/Cu-Ag photoanodes. Conductivity characterization was retrieved on the ZnO/Cu-Ag photoanodes solution using an I-V Keithley meter 2620A to determine the current and voltage variations. Characterization J-V DSSC was carried out on the DSSC device using an I-V Keithley meter 2620A to determine current density and voltage produced in determine the efficiency value of the DSSC.

RESULTS AND DISCUSSION

Conductivity Characterization of ZnO/Cu-Ag Photoanode Solution

Conductivity characterization of the ZnO/Cu-Ag photoanode solution displayed a photoconductivity response indicated by an increase in electric current due to a semiconductor material capturing electromagnetic waves [20]. The photoconductivity response is displayed in the current and voltage relationship curves as shown in Figure 1a. Figure 1a displays an increasing current value with increasing doping concentration and an increasingly linear curve.



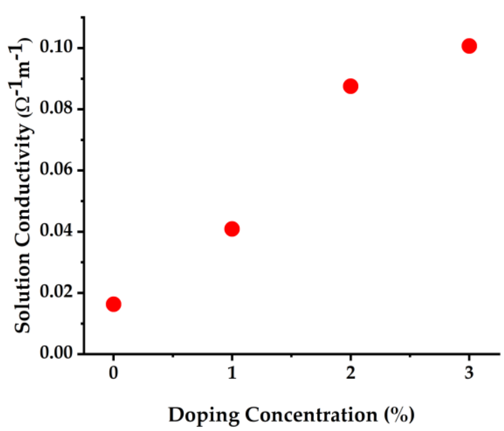


Figure 1. a) Conductivity graph of Zno/Cu-Ag Photoanode solution and b) Solution Conductivity as a Function of Doping Concentration

Based on Ohm's law, an increasingly linear curve shows that the value of constant electrical resistance does not depend on voltage and current. The resistance value of this sample can be determined using a linear graphic method where the slope of the curve determines the size of this value. The smaller resistance value indicates that most charge carriers in the semiconductor can move freely without much resistance. It shows that the material has good quality and can be applied as an active layer for photoanodes. The electrical values that have been obtained will determine the conductivity value of the sample.

The presence of photoconductivity response is due to the phenomenon of electron transport from the valence band to the conduction band, which is the effect of the addition of Cu-Ag double doping on ZnO photoanode. Figure 1b clearly shows the relationship obtained from the effect of the addition of Cu-Ag double doping on ZnO photoanode on the conductivity value is directly proportional where the conductivity increases with the addition of doping. This happens because the addition of Cu-Ag doping affects increasing the majority carrier charge of ZnO semiconductors, namely holes, which affect the electrical conductivity of ZnO [17,18]. According to theory, the conductivity value of a semiconductor is affected by the concentration of most charge carriers. The greater the concentration of the majority of charge carriers due to the addition of doping, the greater the electrical conductivity value obtained from this research. The electrical conductivity values obtained from the overall ZnO/Cu-Ag solution are shown in Table 1.

Table 1. The Values of the Conductivity of Pure Zno and Zno/Cu-Ag Photoanode Solution

Solution Sample	Conductivity (Ω^{-1} m^{-1})
ZnO (0%)	0.0163
ZnO/Cu-Ag (1%)	0.0409
ZnO/Cu-Ag (2%)	0.0875
ZnO/Cu-Ag (3%)	0.1006

According on Table 1, the conductivity value in the sample of pure ZnO photoanode solution is equal to 0.0163 ⊚-1 m-1. The addition of Cu-Ag double doping with concentrations of 1%, 2%, and 3% resulted in the conductivity value of ZnO/Cu-Ag photoanode conductivity values that increased as the doping concentration increased. These results prove that the addition of Cu-Ag double doping can increase the conductivity of ZnO photoanode with an increase of about 2.5 times, 5 times, and, 6 times compared to the conductivity value of pure ZnO photoanode solution.

Optical Characterization of ZnO/Cu-Ag Photoanodes

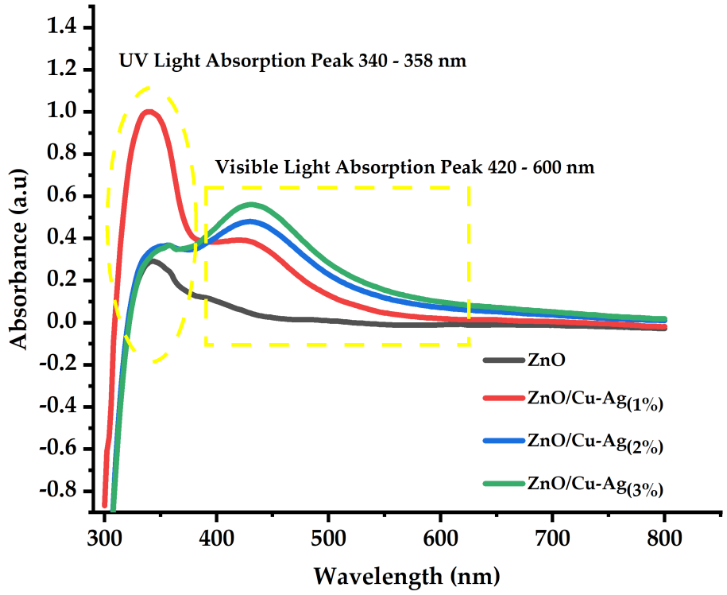


Figure 2. Absorption Spectra of Pure Zno and Zno/Cu-Ag Photoanode

The optical characterization aims to determine the absorption area and the band gap energy of the ZnO/Cu-Ag photoanode sample. The absorption spectrum of each sample shown in Figure 2 shows strong absorption in the UV and visible light region [18,21]. The absorption in the UV light region (<400 nm) occurs due to the effect of using pure ZnO as a semiconductor material [22,23], and absorption in the visible light region (420 – 600 nm) occurs due to the strong interaction between Zn ions with Cu ions and Ag ions also the scattering of light [24,25]. In addition, in the visible light region there is also a wavelength shift toward the redshift. The redshift indicates a decrease in the band gap energy value [17], whose value can be determined using the Tauc Plot method, as shown in Figure 3.

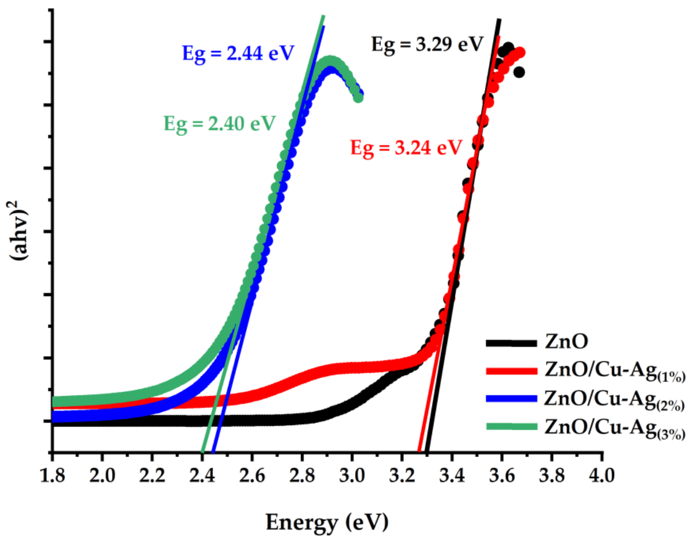


Figure 3. Band Gap Energy of a Zno and Zno/Cu-Ag Photoande

Table 2. The Value of the Eg of a Pure Zno and Zno/Cu-Ag Photoanode

Sample	Band Gap Energy (eV)
ZnO	3.29
ZnO/Cu-Ag (1%)	3.24
ZnO/Cu-Ag (2%)	2.44
ZnO/Cu-Ag (3%)	2.40

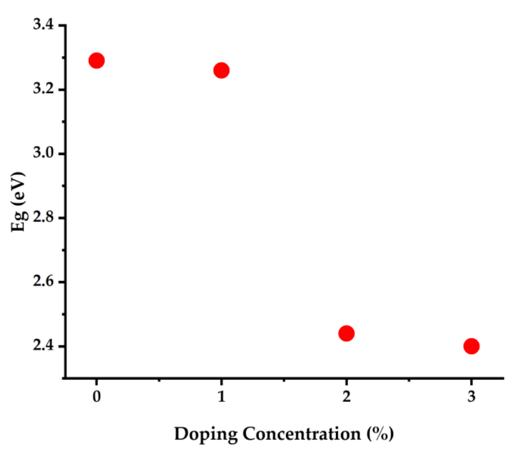


Figure 4. Band Gap Energy as a Function of Doping Concentration

Table 2 displays the band gap energy value of pure ZnO and ZnO/Cu-Ag photoanode. These results based on Figure 4 show a decrease in the band gap energy value as the concentration of Cu-Ag double doping applied to ZnO photoanodes increases. The decrease in the band gap energy value occurs due to the replacement of Zn ions by Cu ions and Ag ions, resulting in the emergence of attractive forces that makes the distance between atoms tighter [12,26]. In addition, Cu-Ag double-doped semiconductors make semiconductors have excess holes so that the acceptor energy appears, which makes it easier for electrons to be excited from the valence band to the conduction band due to the decrease in the required energy value [18]. This ease helps the performance of ZnO photoanodes due to the high number of electrons that can move around [27].

J-V Characterization

A DSSC device shows good performance results in terms of its efficiency in converting solar energy into electrical energy. J-V characterization of DSSC displays the relationship curve between current density and voltage. Figure 5 shows the J-V test results on each ZnO/Cu-Ag photoanode sample.

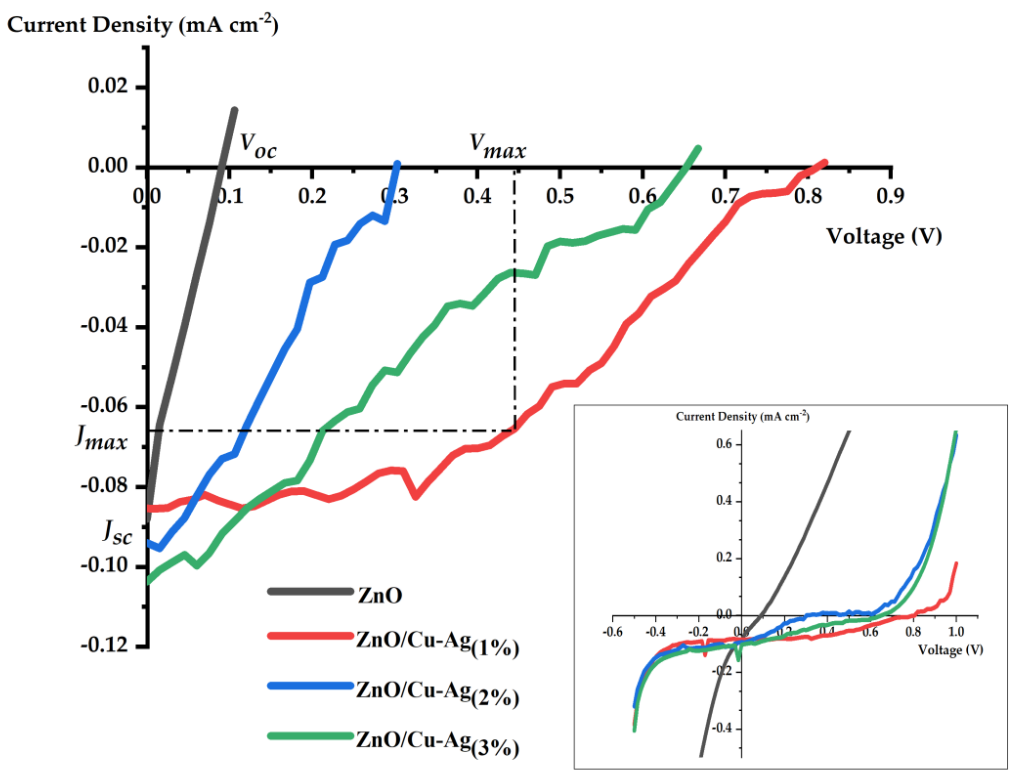


Figure 5. J-V Curve of DSSC

Figure 5 shows the acquisition of photovoltaic parameters of the DSSC, including short circuit current density (J_{sc}), open circuit voltage (V_{oc}), maximum current density ($Jmax$), and maximum voltage (V_{max}), then calculated to obtain the fill factor value and efficiency of the DSSC as shown in Table 3.

Table 3. DSSC Efficiency Based on Zno/Cu-Ag Photoanode

Cu-Ag doping concentration (%)	V_{oc} (V)	J_{sc} (mA/cm²)	V_{max} (V)	J_{max} (mA/cm²)	*FF*	η (%)
0	0.090	0.088	0.045	0.040	0.225	0.0018
1	0.820	0.085	0.045	0.065	0.416	0.0291
2	0.303	0.094	0.151	0.055	0.291	0.0083
3	0.667	0.101	0.303	0.051	0.232	0.0156

The addition of Cu-Ag double doping on ZnO photoanode increases the value of short circuit current density (J_{sc}) and open circuit voltage (V_{oc}) as shown in table 3 regarding the characteristic of DSSC samples for each variation in the use of ZnO/Cu-Ag. This increase in value of J_{sc} shown in table 3 indicates that electron mobility from ZnO/Cu-Ag photoanode occur well because the Cu-Ag doping has high conductivity so it is easy to conduct electrons [28–30]. Meanwhile, the increase in the V_{oc} value indicates that light scattering from ZnO/Cu-Ag photoanode occurs well and this is related to a decrease in the energy banda gap value [31]. The J_{sc} and V_{oc} values produced on each DSSC device with the use of ZnO/Cu-Ag photoanodes 0-3% determine the efficiency value of the DSSC. Each 0-3% Cu-Ag double doping concentration gives different efficiency results depending on the amount of doping concentration. The difference in the efficiency value of each DSSC device due to the addition of Cu-Ag double doping is inseparable from the influence of the absorption value, band gap energy, and conductivity on the ZnO photoanode. The increasingly widening absorption value towards higher wavelengths results in a decrease in the energy band gap value. This decrease makes it easier for electrons to move from the valence band to the conduction band. This easy excitation of electrons certainly impacts the increasing number of electrons acting as conduction along with the high conductivity test results obtained, which help in the productivity of the current produced in the DSSC.

On average, the addition of Cu-Ag double doping on ZnO photoanodes 1-3% in DSSC applications has increased compared to the use of pure ZnO photoanodes. The highest efficiency was achieved in DSSC with a 1% ZnO/Cu-Ag photoanode of 0.0291%, with an increase of about 16 times compared to DSSC with a pure ZnO photoanode. The high efficiency obtained by DSSC indicates the high performance of DSSC in converting solar energy into electrical energy. However, these results are compared with research by Windayani et al. (2019) with the efficiency obtained, namely 0.38%, which is indeed lower [19]. This is due to the use of synthetic dyes, which have a much longer dye life compared to natural dyes. Based on the research results, the efficiency obtained still tends to be low. Apart from that, this can also be caused by limitations in determining effective methods to increase efficiency. Although it cannot yet be used commercially due to low efficiency, this research has the potential to help in the development of DSSCs that are cheap, environmentally friendly, and have high efficiency through more effective experimental methods.

CONCLUSION

Adding Cu-Ag double doping on the ZnO photoanode successfully improved its optical and electrical properties. The improvement of optical properties is indicated by the expansion of the absorption region (redshift) and a decrease in the energy band gap value. Meanwhile, the improved electrical properties are shown by the increased conductivity value of ZnO/Cu-Ag photoanodes achieved at certain doping concentrations. The improvement of these properties impacts increasing the efficiency value of DSSC. The results obtained provide the potential for further development on doping concentration variation, synthesis method optimization, and DSSC stability testing to develop DSSCs with high efficiency and environmentally friendly commercially.

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AUTHOR CONTRIBUTIONS

Rhiska Aria Berliani: Conceptualization, Methodology, Visualization, Writing – Original Draft, Resources, Investigation, Formal Analysis; Fat Hana Rizqi Haq: Methodology, Investigation Resources, Eka Silvia Ningrum: Methodology, Investigation, Resources; Putri Lestari: Investigation; Yofentina Iriani: Supervision, Review & Editing and Fahru Nurosyid: Supervision, Review & Editing.

DECLARATION OF COMPETING INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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