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Dye-Sensitized Solar Cell Utilizing NiPd-rGO Counter Electrode: Comparative Study of Deposition Technique and Effect of $NiCl_2$ Concentration on its Performance

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ABSTRACT

This paper reports the performance of dye-sensitized solar cell (DSSC) utilizing NiPd-rGO composite as counter electrode of the device. Various method of preparation of the counter electrode have been employed. It is found that the device utilizing the counter electrode prepared via liquid phase deposition technique (LPD) performs the highest power conversion efficiency that is 1.18 %. Next, the effect of the concentration of nickel chloride ($NiCl_2$) on the performance of the device utilizing the composite prepared via LPD technique has been investigated. The device with the counter electrode prepared using of 50 mM $NiCl_2$ demonstrates the highest efficiency of 1.07 %.

Keywords: Alloy, Counter Electrode, Deposition Technique, DSSC, rGO.

1. INTRODUCTION

Dye-sensitized solar cell (DSSC) was first developed back in 1991 by O'Regan and Grätzel [1]. The device consists of three major components, photoanode, redox couple electrolyte and counter electrode [2]. Photoanode usually made of titania (TiO_2) coated with dye on a transparent conductive oxide (TCO). As the electrolyte, triiodide/iodide (I_3^-/I^-) redox couple is widely used as an electrolyte in a DSSC. Meanwhile, platinum (Pt) coated onto TCO substrate is commonly used as a counter electrode of the device [3]. This is due to Pt has high catalytic activity and high durability [4]. However, due to its scarcity, Pt is not suitable for large scale production of the device and is also high in cost [5].

Attempts in utilizing carbon film as counter electrode in DSSC is an alternatives to the costly platinum due to its electrochemical reactivity, resistance towards corrosion and high conductivity of electronic [6]–[8]. Reduced graphene oxide (rGO) can also substitute platinum as counter electrode because its price is much lower than platinum and possesses good conductivity. Many researchers had employed rGO film as counter electrode in DSSC [5], [9], [10]. They found that the efficiency of the DSSC employing graphene film counter electrode is at the same level with the device utilizing platinum films counter electrode [7].

The DSSC employed gold doped reduced graphene oxide (Au-rGO) exhibited a low power conversion efficiency of 0.175 % [11]. The efficiency was improved to 0.75 % by replacing gold with silver to form silver doped rGO (Ag-rGO) [12]. A new composite of NiPd alloy doped with reduced graphene oxide (NiPd-rGO) film is believed to improve the performance of DSSC. This composite is used as counter electrode of the device in this work.

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Therefore, in this work, we report the preparation of NiPd counter electrode via various deposition techniques of liquid phase deposition (LPD), spin coating and dip coating. The first objective of this work is concerned with the comparative study of the performance of DSSC utilizing NiPd counter electrode prepared via those techniques. The second objective is to investigate the effect of $NiCl_2$ concentration on the performance of DSSC employing NiPd-rGO counter electrode prepared via LPD technique.

2. METHODOLOGY

Indium tin oxide (ITO) coated glass substrate was used as the TCO for both counter electrode and photoanode. The substrate glass was cleaned with deionized water, acetone and ethanol for 15 minutes respectively in ultrasonic water bath.

2.1 Counter Electrode

2.1.1 Preparation of NiPd Films on ITO Substrate

The NiPd precursor consists of 50 mM $NiCl_2$ and 10 mM $PdCl_2$ in 50 % ethanol. It was prepared by vigorous stirring for 4 hours. The precursor was deposited on the cleaned ITO substrate using dip coating, spin coating and liquid phase deposition (LPD) techniques, respectively.

In dip coating technique, the substrate was held and dipped into the NiPd precursor 10 times by a dipping machine with 30 seconds interval for each dipping. Then, the formed film was dried in air.

For the second technique, spin coater was used to spread NiPd precursor onto the substrate. Firstly, the substrate was heated on hot plate at 100 ^{0}C before spreading 200 μL of the precursor onto it by using a spin coater at 1500 rpm for 30 seconds. The process of heating and coating were repeated for 5 times.

LPD was carried out by hanging the substrate with tape and submerged it into the precursor. The NiPd film was grown on the substrate at 60 ^{0}C for 15 hours. Then, the substrate was rinsed by dipping it in deionized water and dried with nitrogen gas to remove residue on the film. The concentration of $NiCl_2$ was varied with 50, 100 and 150 mM.

The samples prepared via all techniques underwent annealing treatment in the argon atmosphere at 350 ^{0}C for 30 minutes to strengthen the adhesion of the alloy on the substrate.

2.1.2 Preparation of rGO and NiPd-rGO Films on ITO Substrate

rGO film preparation begins by sonicating10 wt% GO solution containing GO flakes and deionized water for 1 hour and stirring for 3 hours. The GO flakes were synthesized earlier using modified Hummers method. GO solution was deposited on preheated clean ITO substrate before spreading 200 μL of the precursor onto it by using spin coater. The spinning speed was 500 rpm for 6 seconds, then followed at 3000 rpm for 30 seconds. The coating process were repeated for 3 times in order to obtain GO film with sufficient thickness. The GO films were finally reduced to rGO through annealing treatment in the argon atmosphere at 350 ^{0}C for 30 minutes. The GO solution also deposited on NiPd films to obtain NiPd-rGO film as the final product. The NiPd films were prepared with LPD technique as mentioned in the previous section.

2.2 Fabrication of DSSC

The DSSCs were fabricated by assembling rGO, NiPd and NiPd-rGO films as counter electrodes and titania film as photoanodes. The porous photoanodes consisting 4 layers of TiO_2, annealed for 30 minutes at 500 ^{0}C for each layer were immersed in 0.5 mM N719 dye solution for 24 hours. Teflon was used as a spacer between the electrodes while parafilms were used as adhesive of the teflon for both electrodes. Adequate amount of EL-HPE electrolyte containing redox iodide/triiodide couple was injected into the cavity between the electrodes. The cavity was sealed with silicone paste sealant. The assembled devices were put in air for 20 minutes to cure the sealant to avoid leakage during testing. A standard DSSC utilizing Pt film as counter electrode was also fabricated in this work as a reference device.

2.3 Performance Study

The photovoltaic performance of the DSSCs was studied with Keithley 2401 source meter in dark and under 1000 W/m^2 sun simulator. The photovoltaic parameters such as short-circuit current density (J_{SC}), open circuit voltage (V_{OC}), fill factor (FF) and efficiency were determined from the J-V curves under illumination. The illuminated area of the devices was 0.23 cm^2. All measurements were repeated three times to ensure reliability and repeatability of the photovoltaic parameters.

3. RESULTS AND DISCUSSION

3.1 Deposition Method of NiPd as Counter Electrode on Performance of DSSC

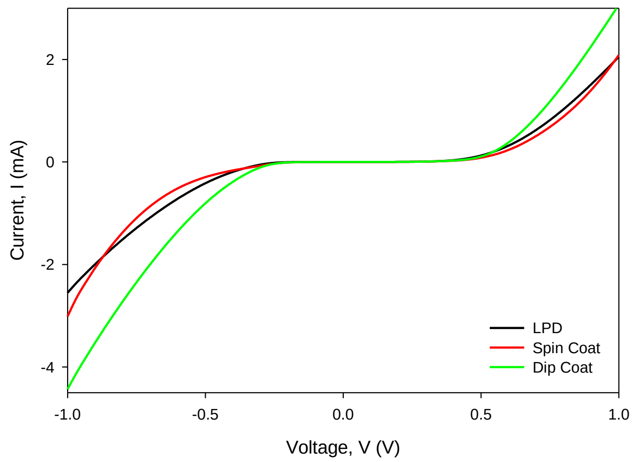

Figure 1. *I*-*V* curves of the DSSC in dark with multiple film deposition methods.

Figure 1 illustrates the behavior of dark current in the devices with the increase in voltage. According to the figure, it shows that the reverse current is higher than the forward current. The reverse current is also called leak current [11]. The highest leak current is observed in the device utilizing NiPd counter electrode prepared by dip coating, followed by that with the counter electrode prepared via spin coat and LPD.

The photovoltaic parameters of the DSSC utilizing NiPd alloy film as counter electrode deposited by dip coater, spin coater and LPD technique are illustrated in Table 1. The DSSC utilizing the sample prepared by dip coating technique demonstrates the highest efficiency of 1.22 %. Meanwhile, the device using the cathode prepared by spin coating technique exhibits the highest J_{SC} of 5.21 mA/cm^2 but the lowest FF of 0.23. Low FF indicates high power loss in the device and leads to low power conversion efficiency as shown in Table 1. This result indicates this method is not suitable for preparing NiPd counter electrode.

The LPD technique, in the other hand, has reliable performance in term of J_{SC}, *FF* and efficiency as shown in Table 1 even though they are not in the highest value. This is due to its *J-V* curve as shown in Figure 2 has more stable current density compared to the spin coat technique and higher J_{SC} than the dip coating technique. Hence, LPD is suitable to be used for preparing NiPd counter electrode in this work.

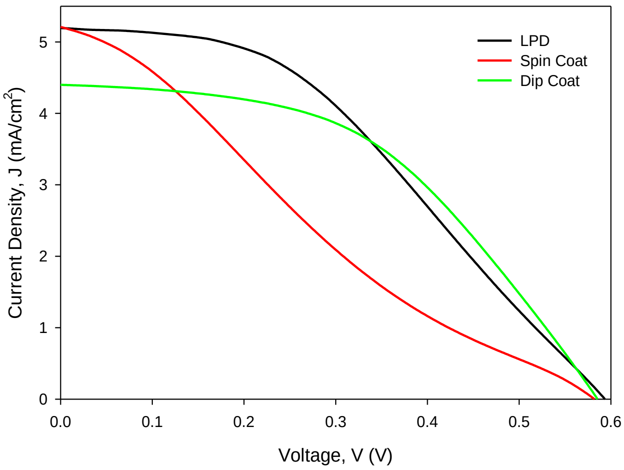

Figure 2. *J-V* curves of the DSSC under illumination with multiple film deposition method.

Table 1 Photovoltaic parameters of DSSC utilizing NiPd films of different deposition method

Deposition method	J_{SC} (mA/cm²)	V_{OC} (V)	*FF*	Efficiency (%)
LPD	5.01 ± 0.19	0.59 ± 0.02	0.41	1.18 ± 0.05
Spin coat	5.21 ± 0.04	0.58 ± 0.00	0.23	0.70 ± 0.02
Dip coat	4.40 ± 0.02	0.58 ± 0.00	0.48	1.22 ± 0.00

3.2 $NiCl_2$ Concentration in NiPd-rGO as Counter Electrode on Performance of DSSC

Figure 3 demonstrates the *I-V* curves in dark of the DSSC with different $NiCl_2$ concentrations. From the figure, it is observed that the leak current trend increases with increasing $NiCl_2$ concentration as highest $NiCl_2$ concentration of 150 mM produced highest leak current followed by 100 mM and 50 Mm.

In Figure 4, the *J-V* curves of the DSSC with NiPd-rGO and rGO counter electrodes under illumination were illustrated. The curves have almost same shape for which the current starts to decrease drastically as the voltage increases from about 0.2 V to 0.5 V. However, the shape of the DSSC utilize Pt and NiPd shows slightly different pattern as they are not containing rGO.

Table 2 shows the photovoltaic parameters of the DSSC utilizing Pt, rGO, NiPd and NiPd-rGO composite film as counter electrode. DSSC with Pt has highest value for all parameters analysed. Meanwhile, the device employing the NiPd-rGO has highest efficiency of 1.07 % belongs to device prepared with 50 mM $NiCl_2$ followed by the device with 150 and 100 mM $NiCl_2$. However, the highest J_{SC} is obtained at the $NiCl_2$ concentration of 150 mM and its lowest at 100 mM. The FF values have insignificant difference since the *FF* is in the range between 0.30 to 0.33.

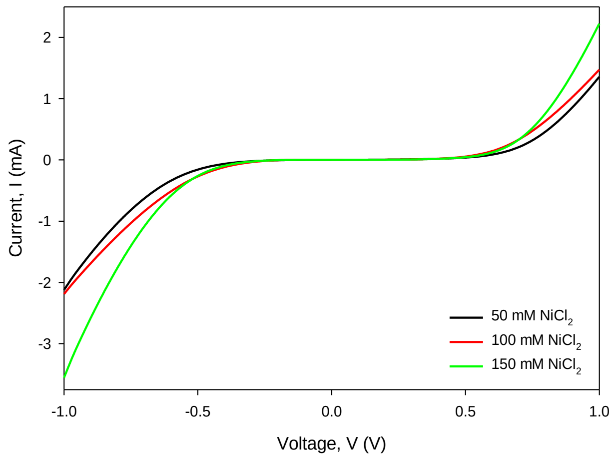

Figure 3. *I-V* curves of the DSSC in dark with different nickel concentration.

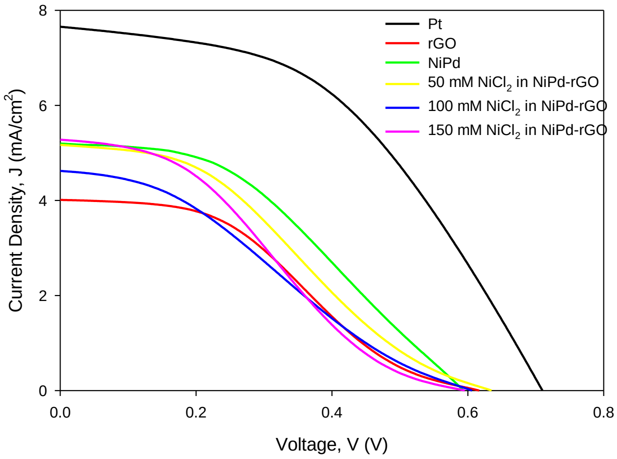

Figure 4. *J-V* curves for the DSSC under illumination with Pt, rGO, NiPd and NiPd-rGO counter electrodes of different $NiCl_2$ concentration.

Table 2 Photovoltaic parameters of DSSC utilizing Pt, rGO, NiPd and NiPd-rGO counter electrodes of different $NiCl_2$ concentration

Film	$NiCl_2$ Concentration (mM)	J_{SC} (mA/cm^2)	V_{OC} (V)	FF	Efficiency (%)
Pt	-	7.66 ± 0.05	0.72	0.46	2.54 ± 0.05
rGO	-	4.01 ± 0.02	0.62	0.36	0.90 ± 0.01
NiPd	50	5.01 ± 0.19	0.59	0.41	1.18 ± 0.05
NiPd-rGO	50	5.17 ± 0.00	0.63	0.33	1.07 ± 0.00
	100	4.62 ± 0.01	0.59	0.30	0.82 ± 0.00
	150	5.28 ± 0.01	0.59	0.31	0.97± 0.01

4. CONCLUSION

This paper highlights the photovoltaic performance of DSSC utilizing NiPd counter electrode prepared via deposition method. DSSC with the counter electrode deposited with LPD technique in terms of photovoltaic parameter is found suitable for this work. The effect of $NiCl_2$ concentration on the performance of the DSSC employing NiPd-rGO counter electrode prepared via LPD has been investigated. The device with the counter electrode prepared using 50 mM NiCl2 has the highest power conversion efficiency of 1.07 %. This work shall continue to find optimum $NiCl_2$ concentration for the highest efficiency of the DSSC employing NiPd-rGO counter electrode.

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