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Study the effect of nickel and aluminium doped ZnO photoanode in DSSC

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ABSTRACT

Dye sensitized solar cells (DSSC) is one of the promising candidates which are efficient, low-cost, and clean hybrid molecular solar cell devices. Zinc oxide (ZnO) has been widely used as the phoanode in DSSC due to its excellent charge conduction mechanism, yet still suffers from poor cell efficiency. In this study, aluminium doped ZnO (ZnO:Al) and Ni doped ZnO (ZnO:Ni) were studied as photoanode material in DSSC using solar cell capacitance simulator (SCAPS) simulation, and the electrolyte liquid considered a single solid p-type layer as hole transporting materials. Both studied photoanodes have demonstrated better cell performance than pure ZnO photoanode due to the small amount of aluminium (Al) and nikel (Ni) impurities added have enhanced the physiochemical properties of ZnO films. A power conversion efficiency (PCE) of 3.96% was obtained at 3 mol% ZnO:Al photoanode with optimized key parameters. These simulation results proved an opportunity to improve the performance of the DSSCs via doping engineering into the ZnO photonaode.

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1. INTRODUCTION

In recent years, dye-sensitized solar cells (DSSC) have gained attention with good photovoltaic performance, specifically under low-light conditions along with flexibility in terms of color and structure. In addition, DSSC is also known for its simple fabrication procedures and low production costs. The first DSSC was reported by Regan and Grätzel in 1990s, using mesoporous titanium oxide (TiO_2) prepared from colloidal TiO_2 nanoparticles for light absorption [1]. DSSC is photo-electrochemical cell-based, in which energy generation for charge carrier transport is driven from the absorption of photon energy and produces a chemical reaction [2]. However, there are many challenges faced by DSSC to improve power conversion efficiency (PCE) without reducing the stability characteristics. Theoretically, the external circuit through the semiconductor photo anode needs to function properly so that the absorption of light by the organic dye can generate and produce a flow of current which in turn produces energy, and this is known as the electrochemical effect. The semiconductor photo-electrode layer plays an important role when current reduction occurs during the recombination of photo-generated carriers in the semiconductor which in turn reduces the efficiency of the solar cell. On the other hand, the photo-anode must be highly transparent to allow sunlight to reach the dye molecules. The concept working mechanism of DSSC was different compared to other types of the solar cell, where DSSC is a photoelectrochemical solar cell that contains a dye-sensitized mesoporous TiO_2 work electrode (WE), a redox mediator (electrolyte) and a counter electrode (CE) [1], [3]-[5]. However, WE and CE can be (semi) transparent that allowing for illumination of the solar cell. Since the beginning of TiO_2-based DSSC research, zinc oxide (ZnO) has been the best TiO_2 alternative for DSSC. Both TiO_2 and ZnO consists same electron affinities and almost same band gap energies approximately 3.2 eV and 3.3 eV, respectively, and ZnO has much higher electron diffusivity than TiO_2 [6], [7] and higher electron mobility of 115 $cm^2\ V^{-1}\ s^{-1}$ -155 $cm^2\ V^{-1}\ s^{-1}$ [8], which is for efficient electron transport in the semiconductor and reduction of recombination rate. The electrical conductivity of ZnO can be enhanced further with a small amount of aluminium (Al) and nikel (Ni) impurities. The ion radius of Zn^{2+} (0.74 Å) are larger than Al^{3+} (0.54 Å) and Ni^{2+} (0.69 Å), therefore Zn^{2+} able to replace at the lattice site when a small quantity of Al^{3+} and Ni^{2+} enters. Subsequently, an additional dopant can be used to improve the conductivity of the ZnO photoanode while maintaining excellent transparency in the visible light region [9], [10].

In this works, the performance of DSSC with Al and Ni doped ZnO as photoanode were investigated using solar cell capacitance simulator (SCAPS) simulation software. Several key parameters including layer thickness, doping concentration, operating temperature as well as defect density were studied in order to obtain a optimum value of PCE in DSSC structureIt is observed that the electrical conductivity in ZnO photoanode is able to increase hgiher with dopant a small amount of material Al and Ni. The best cell performance was achieved with 3 mol% and 4 mol% Al doped ZnO photoanode, which produced PCE of 3.96%. Similarly, Ni doped ZnO photoanode of concentration 4 mol% and 6 mol% provided PCE as high as 3.9% compared to PCE without doped ZnO photoanode which is low. The PCE in DSSC is able to increase and provide best cell performance as the concentration of material increase, however at certain point, Al^{3+} and Ni^{2+} consist of a solubility limit to substitute in ZnO material and these findings were found in previous reports [9]-[12]. The obtained results can be used as a guide and direction for future device fabrication.

2. DSSC SIMULATION

SCAPS, a one-dimensional computer simulation software was used in this work where it was able to simulate the characteristics of solar cells in terms of AC and DC electricity. This simulation software is based on three coupled differential equations that is Poisson's (1) and continuity equations for holes (2) and electrons (3) where D is diffusion coefficient, ψ is electrostatic potential, q is electron charge, G is generation rate, ξ is permittivity, and n, p, n_t and p_t are free holes, electrons, trapped holes, and trapped electrons, repectively. Meanwhile, ionized acceptor-like doping concentration is denoted by N_a^-, and ionized donor-like doping concentration is denoted by N_b^- [13].

$$\frac{d}{dx}\left(-\varepsilon\,(x)\frac{d\psi}{dx}\right) = q\,[p(x) - n(x) + N_d^+(x) - N_a^-(x) + p_t(x) - n_t(x)] \quad (1)$$

$$\frac{dp_n}{dt} = G_p - \frac{p_n - p_{n0}}{\tau_p} - p_n\mu_p\frac{d\xi}{dx} - \mu_p\xi\frac{dp_n}{dx} - D_p\frac{d^2p_n}{dx^2} \quad (2)$$

$$\frac{dn_p}{dt} = G_n - \frac{n_p - n_{p0}}{\tau_n} - n_p\mu_n\frac{d\xi}{dx} - \mu_n\xi\frac{dn_p}{dx} - D_n\frac{d^2n_p}{dx^2} \quad (3)$$

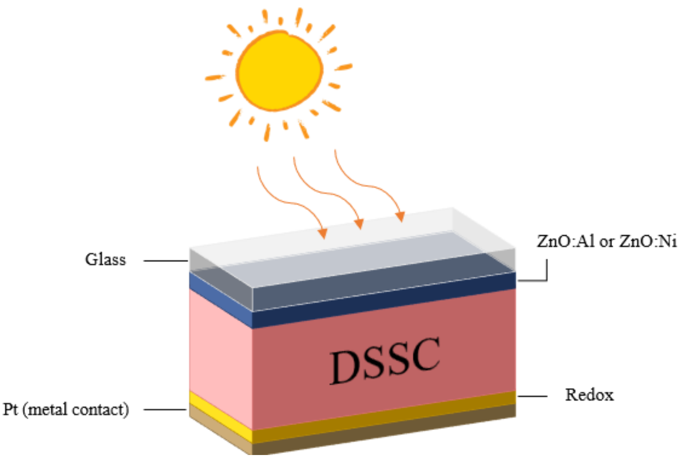


Figure 1. The structure of DSSC with aluminium doped ZnO (ZnO:Al) or nickel doped ZnO (ZnO:Ni) photoanodes

The studied device consists of five main layers which are indium tin oxide (ITO), ZnO:Al or ZnO:Ni as photoanode, N719 dye as dye sensitizer, redox as electrolyte and platinum (Pt) as back contact as shown in Figure 1. The energy level of each layer is illustrated in Figure 2. Herein, the redox acting as an electrolyte is considered to be the solid state *p*-layer [13]. All parameters for each simulated layer are shown in Table 1. The simulation was performed under an illumination of 1000 W/m^2 and an air mass of 1.5 G. The main objective of this work is to investigate the effect of Al and Ni dopant on the ZnO photoanode and to optimize the key parameters of the layer, thus increase the cell efficiency value. The optimization of the main parameters of ZnO: Al and ZnO: Ni photoanodes in terms of thickness, doping concentration, operating temperature and interface defect density has been extensively studied. This work thereby provides a clear insight that with the small addition of Al and Ni dopant on the ZnO photoanode with the optimization of several important parameters has improved the efficiency of DSSC [9], [10].

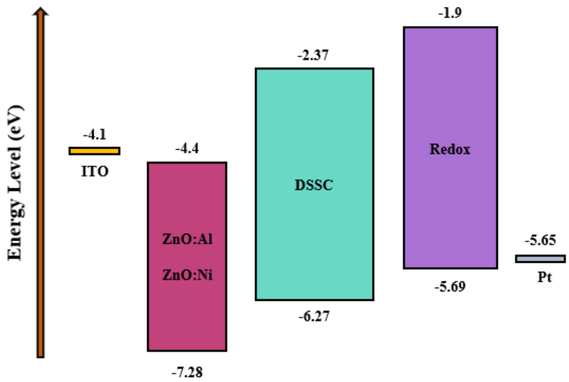


Figure 2. Energy level diagram of simulated DSSC

Table 1. Parameters for DSSC layer

	ITO	N719 Dye	ZnO	Concentration of Al (mol%) [9]				Concentration of Ni (mol%) [10]				Redox
	[14]	[15]	[9]	1	2	3	4	1	2	4	6	[16]
Thickness (nm)	60	500	220	220	220	220	220	180	180	180	180	100
Eg (eV)	3.6	2.37	3.28	3.25	3.1	3.08	3.05	3.18	3.16	3.14	3.13	1.9
X (eV)	4.1	3.9	4.4	4.4	4.4	4.4	4.4	4.4	4.4	4.4	4.4	3.79
$\varepsilon/\varepsilon_o$	10	30	9	9	9	9	9	9	9	9	9	10
N_c (1/cm^3)	2.2×10^{18}	2.4×10^{20}	3×10^{18}	3×10^{18}	3×10^{18}	3×10^{18}	3×10^{18}	3×10^{18}	3×10^{18}	3×10^{18}	3×10^{18}	6.0×10^{21}
N_v (1/cm^3)	1.8×10^{19}	2.5×10^{20}	1.7×10^{19}	1.7×10^{19}	1.7×10^{19}	1.7×10^{19}	1.7×10^{19}	1.7×10^{19}	1.7×10^{19}	1.7×10^{19}	1.7×10^{19}	6.0×10^{21}
μ_e (cm^2/V_s)	50	5	43.00	14.03	13.84	13.35	15.02	36.73	37.01	35.49	55.39	0.1
μ_h (cm^2/V_s)	75	5	31	31	31	31	31	31	31	31	31	0.3
N_D (1/cm^3)	1×10^{19}	0	2.9×10^{15}	7.2×10^{18}	1.02×10^{19}	1.46×10^{19}	1.06×10^{19}	6.09×10^{18}	7.53×10^{18}	8.14×10^{18}	8.80×10^{18}	1×10^{15}
N_A (1/cm^3)	0	1×10^{17}	0	0	0	0	0	0	0	0	0	1×10^{15}
V_e (cm/s)	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}
V_h (cm/s)	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}

3. RESULTS AND DISCUSSION

3.1. Effects of Al and Ni doped ZnO photoanode in thickness

In the structure of a solar cell, the thickness of the layer plays a very important role in improving cell performance. Here, the layer thicknesses for Al doped ZnO (ZnO:Al) and Ni doped ZnO (ZnO:Ni) photoanodes were varied with different dopant concentrations in the range of 5 nm to 1000 nm for SCAPS simulations. PCE for both ZnO:Al and ZnO:Ni photoanodes showed a gradual increase with layer thickness as shown in Figure 3. However, this trend was different for ZnO photoanodes with no dopant addition, where PCE had declined sharply by 20% at 100 nm thickness compared to 5 nm thickness. This trend persists with no significant change with further increase in layer thickness. It was also found that, concentrations of 3 mol% – 6 mol% Al and Ni doped ZnO photoanodes have shown higher PCE values for all layer thicknesses compared to others by achieving PCE values as high as 3.9%. The increase in cell efficiency at higher Al and Ni doped ZnO photoanode layer thicknesses can be attributed to enhanced photon absorption, producing more electron hole pairs and thus increasing charge carrier conduction. However, the trend is the differ for pure ZnO without doping, where there is a decrease in PCE with increasing layer thickness. This is probably due to the development of surface containing valleys and peaks at higher thickness [17], [18]. In addition, the chance for recombination to occur is high which results in a decrease in PCE and this is due to the charge requiring a longer journey to conduct current and thus generate energy [19].

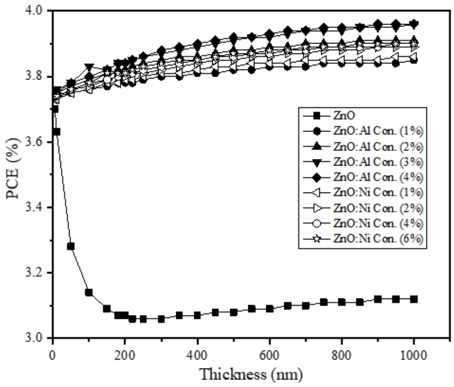


Figure 3. PCE for Al and Ni doped ZnO photoanodes with different dopant concentrations at different layer thicknesses

3.2. Effects in doping density (N_D)

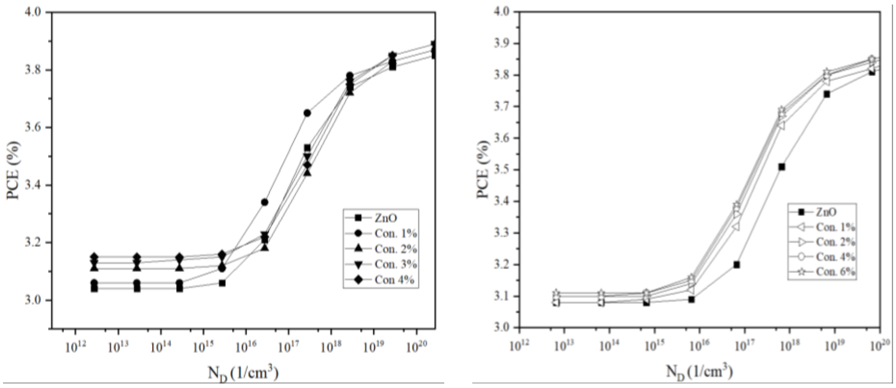
The doping density of ZnO:Al and ZnO:Ni photoanode were varied from 1×10^{12} cm^{-3} to 1×10^{20} cm^{-3}. Figure 4(a) and Figure 4(b) show similar trend curve where the PCE of ZnO:Al and ZnO:Ni photoanode rises gradually as the doping density increases. The increase in photoanode doping concentration will enhance the conductivity and reduce the reverse saturation current, thus leading to PCE growth [20]. Generally, the conductivity (σ) in N-type doping is establish as:

$$\sigma = nq\mu_n \quad (4)$$

Where n is the electron concentration, μ_n is the electron mobility and q is the electric charge. The equation in low doping were obtain as:

$$\sigma \approx N_d q\mu_n \quad (5)$$

Where $n \approx N_d$ and N_d is the ionized donor concetration. The positive correlation between σ and N_d occurs since the mobility does not change substantially with N_d in case of low doping. It can also be observed that PCE grows slowly as the doping density exceeds 10^{15} cm^{-3}. The Moss-Burstein effect will create substantial doping effects if doping density continues to rise to a certain point which will limit the DSSC performance [21].



(a) (b)

Figure 4. Effect of the doping concentrations on the PCE for (a) Al doped ZnO photoanodes DSSC and (b) Ni doped ZnO photoanode DSSC

3.3. Effects of Al doped ZnO and Ni doped ZnO at different working temperature

In thin film solar cell applications, the operating temperature has a considerable impact on device performance. The solar cell panels are expected to be functional at temperatures above 300 K. So, in this investigation, operating temperature for both Al and Ni doped ZnO are analyzed from ranged 300 K to 400 K to understand its effect on the cell performance. It can be observed that all PCE devices performance slightly decrease as the temperature rises, approximately by -0.02 ± 0.01 %/K hence proving that an increase in temperature causes an impact on all parameter as shown in Figure 5. As the temperature rises, all parameter includes the material carrier concentration, band gaps, electron and hole mobility values changed, resulting in a reduced cell efficiency [22]. In addition, the significant absorption of energy by electrons at high temperatures causes the efficiency of solar cells to decline with temperature. The electrons are stimulated to move to an unstable state and trigger recombination before entering the conduction band [19], [23].

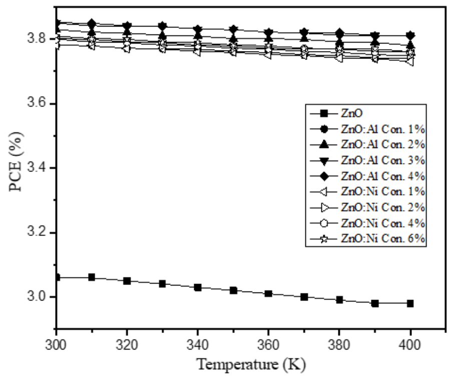


Figure 5. Effect of increasing temperature to the ZnO, ZnO:Al and ZnO:Ni photoanodes DSSC

3.4. Effects in defect density

The parameters of the interface layer have a great influence on the perfomance of solar cells. The interface layer quality depends on defect density, where higher defect density generating the recombination rate, thus decreasing the quality of the interface layer. To observe the effect in interface layer of N719 Dye/ZnO:Al and N719 Dye/ZnO:Ni, defect density was varied from range 1×10^{10} cm^{-2} to 1×10^{20} cm^{-2}. At lower value of defect density, ZnO photoanode without doped obtained the lowest PCE which is around 3.06% compared to photoanode with Al doped ZnO and Ni doped ZnO. Furthermore, PCE for N719 Dye/ZnO:Al and N719 Dye/ZnO:Ni interface layer began to show a downward trend at value of 1×10^{12} cm^{-2} which can be seen in Figure 6. PCE defect density decreased due to a lower charge recombination rate and carrier recombination at the N719 Dye/ZnO interface, which was achieved by a lower concentration of surface defects in ZnO material [24]. Furthermore, if the interface layer defect density continues to increase, the DSSC performance will degrade because high defect densities in N719 Dye/ZnO:Al and N719 Dye/ZnO: Ni lead to increase the traps and form the recombination centres [25].

The optimum PCE values for each Al-doped ZnO and Ni-doped ZnO photoanode in DSSC that obtained from layer thickness, doping density, operating temperature and defect density are shown in Table 2. It is observed that 3 mol% - 6 mol% concentration of Al and Ni yield highest PCE through out the studies. This demonstrate that by adding a small amount of Al and Ni material can increase the performance of ZnO in their electrical conductivity.

Table 2. Optimum PCE for Al and Ni dopant concetration in ZnO photoanode

Photoanode	Dopant concentration (mol%)	CE (%)
ZnO	ZnO	3.7
Al doped ZnO	1	3.85
Al doped ZnO	2	3.91
Al doped ZnO	3	3.96
Al doped ZnO	4	3.96
Ni doped ZnO	1	3.86
Ni doped ZnO	2	3.89
Ni doped ZnO	4	3.9
Ni doped ZnO	6	3.9

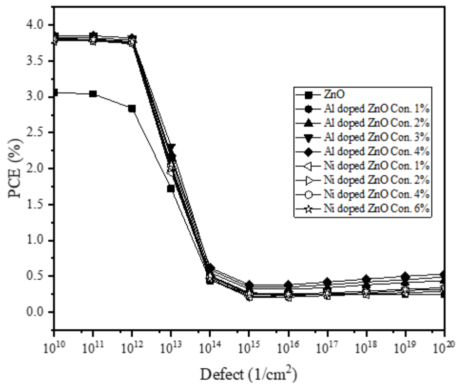


Figure 6. Cell performance with increasing interface defect density for different dopant of Al doped ZnO, Ni doped ZnO and ZnO photoanode DSSC

4. CONCLUSION

Herein, we report the effect of Al and Ni impurities in ZnO photoanode of DSSC on cell performance using SCAPS-1D. DSSC has been succesfully simulated with different doping concentrations along with optimization of several key layer parameters. Cell efficiencies as high as 3.96% and 3.9% were obtained using above 3 mol% ZnO:Al photoanode and 4 mol% ZnO:Ni photoanode, respectively. The cell performance obtained was enhanced than that found in pure ZnO photonoade DSSC, indicating that the additional dopant had improved the charge carrier conduction and thus increased the PCE. However, further increase in dopant concentration above 6 mol% will result in deterioration of cell performance due to larger particle size and Burstein-Moss effect. So, this work thereby provides a clear insight that the addition of dopants into ZnO photoanodes with optimized key parameters has successfully improved the performance of DSSC cells. The addition of Ni and Al dopants has contributed to an increase in cell efficiency and in turn further elevates PCE. This is most probably due to the radius of Al^{3+} and Ni^{2+} of 0.54 Å and 0.69 Å, respectively, are smaller than the Zn^{2+}, 0.74 Å. Therefore, the Al^{3+} and Ni^{2+} ions can sneak in and replace the Zn^{2+} ions at the lattice site as well as passivate any possible defect in the ZnO photoanode. The results obtained are useful as a guide and direction in the manufacture of future devices.

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