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Response surface modeling of photogenerated charge collection of silver-based plasmonic dye-sensitized solar cell using central composite design experiments

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In this study, silver nanoparticles (AgNP) have been prepared and successfully incorporated in TiO_2 nanopowder and used in dye-sensitized solar cell as photoanode. The effect of the AgNP concentration and photoanode film thickness on the charge collection efficiency of a photogenerated electron at the external circuit was investigated using response surface methodology. A multiple regression analysis of second order polynomial was employed to fit the experimental data and an empirical model was subsequently developed using analysis of variance (ANOVA). The results show that two independent variables (AgNP concentration and photoanode film thickness) have significantly influenced the charge collection efficiency of the silver-based plasmonic DSSC. An optimum charge collection of 64.3% was obtained at AgNP concentration and film thickness of 5%wt and 10 μm, respectively.

Introduction

The Economic and social wellbeing of any nation or community are largely dependent on the extent of its energy utilization, thus, energy, fresh water, and air are considered as the most important commodities for human existence. Fossil fuels such as petroleum, natural gas, and coal are the most widely used sources of energy for industrial and domestic purposes. The depletion of the earth, fossil fuel reserves and environmental concern such as greenhouse gas emission are some of the drawbacks of these highly efficient carbon-based fuels [1]. Solar energy is a renewable source of energy that has attracted global attention as a substitute for the fossil fuels owing to its numerous advantages of being a naturally infinite commodity that is free from environmental pollution. Hence solar energy is indeed one of the most viable alternatives to consider in overcoming world's energy crisis.

Among the various solar photovoltaic cells, dye-sensitized solar cell (DSSC) has been considered as a promising solar cell device for solar electricity generation owing to its low cost, flexibility, ease of production, and low toxicity [2].Titanium dioxide (TiO_2) has been widely used in DSSC photoanode due to its large surface area which is beneficial in maximizing dye loading onto the semiconductor surface [3]. The modification of TiO_2 with a metal nanoparticles such as gold (Au), silver (Ag) [4] and platinum (Pt) has been widely reported to prevent the recombination of the photogenerated electron–hole pairs and improve the charge transfer efficiency in DSSC [5]. Silver nanoparticles have been utilized to improve photon absorption in the TiO_2-based DSSC photoanode because of its surface plasmon resonance effect which concentrates and then scatters the incoming solar radiation [6] which enhances light absorption by the sensitizing molecules.

In addition to their surface plasmon resonance effect, a noble metal to TiO_2 can also enhance the photovoltaic performances by changing the surface properties of the semiconductor, since the work function of the metal is higher than that of TiO_2, such that electrons are displaced from the TiO_2 in the vicinity of each metal particle which then create a Schottky barrier at each metal–semiconductor region, thereby decreases the charge recombination [7].

For high conversion efficiency to be achieved, there must be an efficient collection of nearly all the photogenerated electrons which means that the incident photon to- Current-efficiency should be close to unity under visible light region. This can be realized if the carrier diffusion length is greater than the film thickness [8].

Response surface methodology (RSM) is a resourceful statistical modeling tool that has been developed and used in testing process

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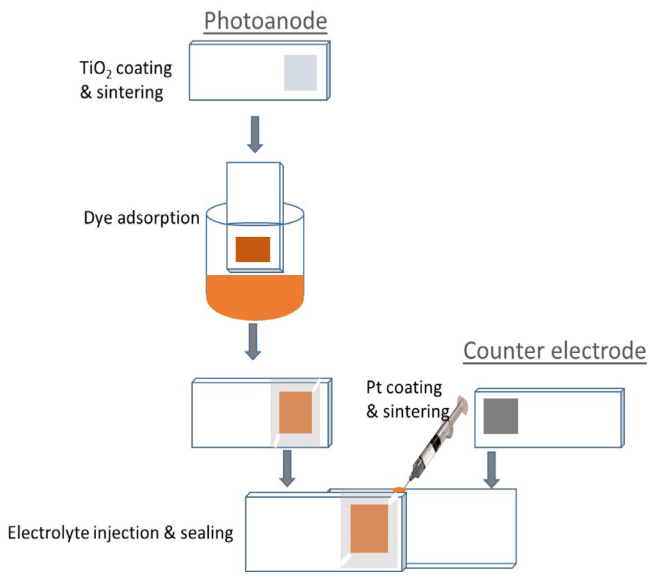

Fig. 1. DSSC fabrication process.

parameters and their effects on the resulting output in a given experiment [9], RSM is used in design optimization in order to reduce the cost of expensive analysis methods such as finite element method and computational fluid dynamic analysis and their attendant numerical noise. Additionally the problem can be estimated with smooth functions that increase the convergence of the optimization process because they reduce the effects of noise which allow for the use of derivative-based algorithms. This mathematical technique can also be used in developing an approximately accurate prediction of system throughput and subsequent development of a mathematical model that can exactly describe the overall process [10], the developed model identifies the links between various operational factors and their individual responses with multiple ideal criteria by determining the influence of these effective parameters on the coupled output variables, thereby minimizing the experimental costs and more importantly, reducing the inconsistency around the expected result [11].

In this research, a mathematical model has been developed based on several experimental trials in order to gain an insight on the influence of film thickness and concentration of silver nanoparticles on the collection efficiency of the photogenerated electron in DSSC. To realize this objective, a central composite design (CCD) based on the polynomial model of quadratic order was employed where each factor is varied over five levels for efficient design optimization. Subsequently, the model was fortified through regressive analysis and examination by analysis of variance (ANOVA) technique to minimize error thereby improving its resultant accuracy.

Materials and methods

Materials

Titanium dioxide (TiO_2), Flourine Tin oxide (FTO) coated glass ($7sq^{-1}$), Di-2 Cis-bis (isothiocyanato) bis-bipyridyl-4-4′-dicarboxy lato) ruthenium (II) (N719) dye were obtained from Sigma-Aldrich Co., (USA). Silver nitrate was purchased from Qrec chemicals, and liquid electrolyte was obtained from Kyutech Laboratory, Japan

Methods

Silver nanoparticles were synthesized according to the procedure reported by Silvert et al. [12] Firstly, 4 g of polyvinylpyrrolidone(PVP) was dissolved in 50 mL ethylene glycol at room temperature, also 100 mg $AgNO_3$ was added to this solution. The resultant suspension was stirred using magnetic stirrer for 30 min. thereafter, the solution was put in a Teflon autoclave and heated in a furnace at 120 °C for 6 h and allowed cooling to room temperature, the PVP, and the solvent were removed by centrifugation. Secondly, TiO_2/Ag paste was obtained by mixing a required amount of the AgNP with 2g TiO_2 powder in ethanolic solution of ethyl cellulose.

TiO_2/Ag layer was deposited on FTO-coated glass substrates by screen printing technique followed by drying at 100 °C in an oven. The desired film thickness (from 5 μm to 30 μm) was achieved by reperative coating, the obtained films were sintered in a furnace at 450 °C for 30 min. A monolayer dye coating of the electrodes was achieved by immersing the films in a 0.2 mM ethanolic solution of N719 for 12 h. The counter electrodes were fabricated by coating a platinum paste on a pre-drilled FTO glass substrates by screen printing followed by sintered 400° for 30 min. The dye-loaded photoanode and the Pt-counter electrodes were assembled together sandwiched by 60 μm surlyn polymer sheet. The assembled devices were heated on a hot plate until the two electrodes were firmly glued together. Finally, liquid electrolyte was administered into the device through the drilled holes at the counter electrode and then sealed with a polymer sheet (Fig. 1).

Determination of charge transfer process

Electrochemical impedance spectroscopy (EIS) was carried out on the DSSCs using Autolab PGSTAT204, the electrochemical impedance spectra were measured in the frequency range between 0.01 Hz and 100KHz The series resistance (R_s) of the cell was deduced from the Nyquist plot (Fig. 4.) as the point of intercept of the semicircle on the x-axis while the charge transfer resistance (R_{ct}) at the dye/electrolyte interface is the value of the arc length of the semicircle [13]. Electrochemical impedance data represented in a Bode phase plot was used for the determination of maximum frequency, here, the log of the frequency is plotted on the x-axis and the phase shift on the y-axis. The maximum frequency(ω_{max})

Table 1
Comparison of photovoltaic performance of some plasmonic $DSSC_s$.

Photoanode	NP synthesis method	J_{sc} (mA cm^{-2}	V_{oc} (V)	FF	?? (%)	Refs.
TiO_2-Ag	Chemical reduction	8.4	0.63	51	2.7	[15]
TiO_2-Ag	Chemical reduction	12.19	0.77	0.52	4.86	[16]
Au-Ag/TiO_2	Chemical reduction	23.5	0.76	0.41	7.33	[17]
Ag-TiO_2	Thermal evaporation	10.0	0.62	0.41	2.55	[18]
Al-TiO_2	Solution process	17.6	0.72	0.56	6.95	[19]
Ag/TiO_2	Photoreduction	16.2	0.76	–	8.9	[20]
TiO_2-Ag	Biosynthesis	11.80	0.79	0.55	5.12	[21]

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