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Applications of Factorial Design in Dye-Sensitized Solar Cells with Titanium-Silicon Dioxide as Photoanode

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Abstract**: *Titanium-silicon dioxide (TiO_2-SiO_2) nanocomposite thin films have been synthesized by sol-gel process as a photoanode in dye-sensitized solar cells (DSSC). The photovoltaic performances, i.e. J_{SC}, V_{OC}, FF, and η were explored using I–V measurement. The effects of electrolyte type (iodolyte, PAN-based gel polymer: E1 and E2) and the preparation of photoanode (annealed temperature) on the performance of DSSC was significantly studied using factorial design methodology. It reveals that in factorial design, both main factors (electrolyte type and annealed temperature) and the interaction between these two factors were found statistically significant. It means that the effects of electrolyte type and annealed temperature, are the significant variables influencing the energy efficiency, as well as the two-factor interactions of electrolyte type and annealed temperature. The determination coefficient (R^2) also in good alliance, which confirms that there exist a high association between these factors with the energy efficiency of DSSC. The optimum conditions of the energy efficiency occurs for a PAN-based gel polymer E2 when the photoanode was annealed at 350 ℃, exhibits a highest energy efficiency of 1.5%.***

Keywords : analysis of variance, determination coefficient, dye-sensitized solar cells, electrolytes, energy efficiency factorial design.

I. INTRODUCTION

Recently, dye-sensitized solar cells (DSSC) obtained global focus as a resort to solar silicon photovoltaics due to their intelligibility of construction, low-cost materials, and higher photoelectric conversion efficiency [1]–[3]. Since power conversion efficiency of titanium dioxide (TiO_2) have been recorded up to 10%, so this metal oxide type has been fully utilized in DSSC as semiconductor. Furthermore, this popular semiconductor grants high chemical and stability, nontoxicity, and economy-size [4].

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However, the performance of DSSC is constrained due to its high electron recombination with the electrolyte, and low electron mobility and transport properties. Therefore, many alternatives have been explored such as substituting TiO_2 photoanode with other metal oxide [5]–[7], modifying the morphology of TiO_2 by increasing dense layers (known as hybrid) [8]–[9], or added a doping [7], [11] as replacements of nanoparticles to accelerate electron transfer. To improve the performance of DSSC, zinc oxide (ZnO), magnesium oxide (MgO), aluminum oxide (Al_2O_3), and silicon dioxide (SiO_2) are the possible oxide materials which able to reduce the electron recombination [5], [10].

The performance improvement in DSSC is investigated on TiO_2-SiO_2-based DSSC as potential semiconductor [8], [12]–[13]. Ennaoui et al. claimed that the factor which leads to poor efficiencies of solar cell is contributed by low open-circuit voltage (V_{OC}). Furthermore, Nguyen et al. presented that the energy efficiency has been boosted by 0.15% by adding SiO_2 into $TiO_2/(SiO_2)_{0.53}$ electrodeposition nanocomposite films [13].

The most practical statistical tool for the study of the effects of two factors is factorial design. Using factorial design, each replication of DSSC are examined and this method is identified more reliable. Furthermore, it's beneficial when the interactions present, since it can avoid misleading conclusions. Another advantage of factorial design is to allow the effects of a factor to be projected at several levels of the other factors [14].

In this study, the energy efficiency improvement is investigated. The effects of electrolyte type and annealed temperature were studied using factorial design and was significantly determined by analysis of variance (ANOVA).the final paper but after the final submission to the journal, rectification is not possible.

II. EXPERIMENTAL DETAILS

A. Preparation of TiO_2-SiO_2

In this research, the synthetization of silicon dioxide nanocomposite powder by a sol-gel technique. Firstly, tetraethyl-orthosilicate (TEOS) was mixed with ethanol (C_2H_5OH) solution under magnetic stirring for 15 minutes. This partial hydrolysis of TEOS was followed by adding the deionized water and dropping 3.3 mL acid ammonia. Then, the reaction is continued stirring and warmed up at 60˚C for an hour, until a stable, and translucent solution obtained.

To remove any extra chemical, the resulting mixture was then filtered and cleaned with ethanol. Then, the solution was withered for eight hours at 80°C in an oven to produce a homogeneous powder. The dried SiO_2 were mortar grounded and pestle into a fine powder before mixed with TiO_2. Hence, the photoanode film was annealed at 300°C, 350°C and 400°C for 30 minutes.

The photoanodes were then prepared by dipping the hardened TiO_2-SiO_2 in 0.5 mM ethanolic solution of N719 dye solutions at room temperature for 24 hours. After 24 hours-absorption period, the photoanodes were rinsed in ethanol. Before fabricating the DSSCs, a slim layer of platinum (Pt) was deposited on the cleaned FTO substrate. Then, the electrodes were warmed up at 100°C, succeeded by annealing procedure at 450°C for an hour. Hence, the DSSC was fabricated by clamping the two electrodes together like a sandwich. Finally, iodolyte was inserted into space between the two electrodes.

B. Preparation of PAN-based gel polymer electrolyte

In this research, the gel polymer electrolyte have been prepared using polyacrylonitrile (PAN), ethylene carbonate (EC), propylene carbonate (PC), potassium iodide (KI) and iodine (I_2). Table 1 was shown the ratio of PAN:EC:PC:KI:I_2 used for preparing PAN-based gel polymer electrolyte. EC, PC and KI are stirred until KI is totally dissolved. Then, PAN was added and continuously stirred and heated at 110°C for 5 minutes. Finally, I_2 is added to the solution to get the desired gel polymer electrolyte.

III. METHODOLOGY

A. DSSC photovoltaic properties

The photovoltaic parameters inclusive of short-circuit current density (J_{sc}), open-circuit voltage (V_{oc}), fill factor (*FF*), and energy efficiency (η) were calculated. Table 2 shows the corresponding photovoltaic properties. J_{sc} is defined as the maximum current attained when the cells are short-circuited. While, V_{oc} is described as the maximum voltage that the cells can transported. The fill factor (*FF*) is defined as the rate between the maximum power and the product of V_{oc} with J_{sc}.

The energy efficiency was computed with the (1):

$$\eta = (FF*J_{sc}*V_{oc}) / P_{in} \quad (1)$$

where P_{in} was identified as the incident power, and the fill factor (*FF*) can be expressed as:

$$FF = (V_{max}*J_{max}) / (V_{oc}*J_{sc}) \quad (2)$$

where V_{max} and J_{max} were the maximum voltage and photocurrent measured, respectively [16].

B. Factorial Design

This study involved the effects of two factors. Hence, factorial designs are most efficient for this study. In the two-factor factorial, the main effect is defined as an outcome that can show consistent difference between levels of factor i.e. electrolyte type and temperature. Furthermore, factorial ANOVA also can examine the occurrence of interaction effect between these factors.

- *Electrolyte Effect:* The null hypothesis states that the means efficiency is statistically equal across the three types of electrolytes i.e. iodolyte and different ratio of gel polymer PAN-based gel polymer electrolyte (E1 and E2). Because the null hypothesis assumes all the means are equal, so the alternative hypothesis is:
  Alternative hypothesis: At least one type of electrolyte is statistically significant.

- *Annealed Temperature Effect:* Another main factor that contribute to energy efficiency of DSSC is heat treatment i.e. annealed temperature. Hence, the hypotheses about the equality temperature treatment effects are:
  Null Hypothesis: The means efficiency between each temperature (300°C, 350°C and 400°C) is statistically equal.
  Alternative Hypothesis: At least one temperature is statistically significant.

- *Interaction Effect:* The statistical test for possible interaction effects is also tested. Thus, the hypotheses for interaction effects are:
  Null Hypothesis: The two main effects (types of electrolyte and annealed temperature) are independent.
  Alternative Hypothesis: At least one interaction is significant.

For the present work, the parameters have been estimated by using a statistical software package (SPSS v16). This software was used to assign two independent variables, namely, electrolyte and annealed temperature as shown in Table 3, which resulted in 36 experimental sets.

Table- I: Percentage Weight (wt.%) of PAN-based gel polymer electrolyte

Gel Polymer	PAN : EC : PC : KI : I2
E1	10 : 43 : 38 : 4 : 4
E2	10 : 38 : 43 : 4 : 4

Table- II: Independent Variable Coded Level

Name	Coded Level			
	Factor	*1*	*2*	*3*
Electrolyte	*A*	Iodolyte	E1	E2
Temperature	*B*	300°C	350°C	400°C

IV. RESULT AND DISCUSSION

A. Photovoltaic Properties

The DSSC efficiency (%) versus annealed temperature (°C) is shown in Figure 2. From the plot, the highest efficiency (about 1.5%) values were obtained with gel polymer E2 when the annealed temperature was 350°C. However, the lowest efficiency values were obtained with iodolyte when the annealed temperature was 400°C. As we can see from the Figure 1, changing from low to intermediate annealed temperature, DSSC efficiency with gel polymer E1 and E2 has increased, whereas it decreased for iodolyte. From intermediate to high temperature, the DSSC efficiency decreased for all electrolyte type. Gel polymer E2 seems to give the results as the temperature changed.

Table 4 presents the photovoltaic performance of TiO_2- and TiO_2-SiO_2-based DSSC annealed at different temperature (300°C, 350°C and 400°C). From Table 4, gel polymer E2 showed the highest J_{SC} (annealed temperature 350°C), 23.85 mA cm^{-2} with V_{OC} of 0.2 V and FF of 0.314. However, iodolyte generated the lowest efficiency at annealed temperature 400°C with 13.132 mA cm^{-2} of J_{SC}, 0.08 V of V_{OC} and FF of 0.304.

It is reported that greater surface area and high absorption coefficient of the sensitizer are correlated with the increment of J_{SC} [15-16].

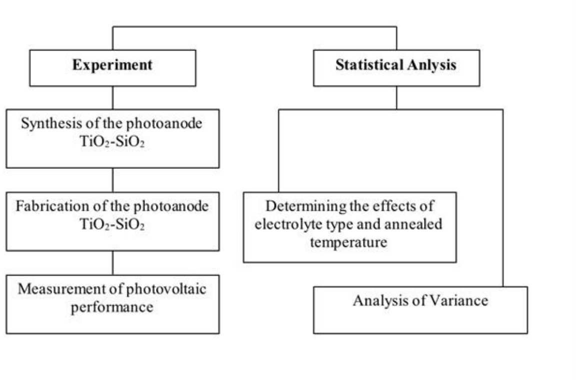

Fig. 1. Flowchart of the Methodology

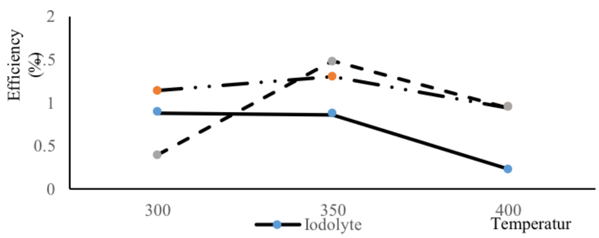

Fig. 2. Electrolyte Type vs Annealed Temperature

Table- III: Photovoltaic performance of iodolyte, gel polymer E1 and E2

Electrolyte	Annealed Temperature 300°C			
	J_{sc} [mA cm^{-2}]	V_{oc} [V]	*FF*	η [%]
Iodolyte	16.6600	0.1499	0.3688	0.9243
Gel Polymer E1	19.2840	0.1699	0.3508	1.1516
Gel Polymer E2	18.2400	0.1598	0.2755	0.8008
	Annealed Temperature 350°C			
Iodolyte	21.6440	0.1599	0.3681	1.2742
Gel Polymer E1	20.1080	0.1998	0.3614	1.4512
Gel Polymer E2	23.8500	0.1998	0.3145	1.5009
	Annealed Temperature 300°C			
Iodolyte	13.1320	0.0799	0.3039	0.3186
Gel Polymer E1	16.7000	0.1599	0.3652	0.9753
Gel Polymer E2	16.2960	0.1598	0.3548	0.9258

Table-IV: Efficiency (%) of TiO_2-SiO_2-based DSSC Data

Electrolyte	Temperature (°C)					
	300		*350*		*400*	
Iodolyte	0.8796	0.8764	0.2087	1.0747	0.2635	0.2266
	0.9045	0.9210	1.1465	1.0495	0.2352	0.2293
Gel Polymer E1	1.1422	1.1492	1.3588	1.3022	0.9664	0.9535
	1.1371	1.1568	1.3416	1.2272	0.9752	0.9240
Gel Polymer E2	0.4512	0.3770	1.4988	1.5116	0.9664	0.9534
	0.3291	0.4512	1.4820	1.4752	0.9752	0.9240

Table-V: Analysis of variance of energy efficiency

Source of Variation	Sum of Squares	Degree of Freedom	Mean Squares	*F* values	Probability value
A	1.333	2	0.667	29.263	0.000
B	1.734	2	0.867	38.060	0.000
*A*B*	1.998	4	0.499	21.917	0.000
Error	0.615	27	0.23		
Total	36.013	35			

B. Analysis of Variance

Table 5 shows the results of analysis of variance (ANOVA). As stated in Table 5, the probability value equal to 0.000, means that there is a statistically significant interaction between types of electrolyte and annealed temperature. Furthermore, the main effects of electrolyte and temperature are also significant. The ANOVA also demonstrated that the determination coefficients (R^2) was 0.892, indicating a high association between electrolyte and temperature with the energy efficiency of DSSC. It interprets that the factors A and B (types of electrolyte and annealed temperature) contributed with 89.2% to the DSSC efficiency.

V. CONCLUSION

Performances of DSSC were studied as a factor of electrolyte type and annealed temperature during preparation of nanocomposite TiO2-SiO2-based DSSC. By employing the synthetizations process parameters, the energy efficiency of DSSC through modifying the particle characteristics can be altered.

To analyze the main factors and interaction among the main factors, a factorial design was employed in this research. The energy efficiency of DSSC was influenced by the single factor effect of electrolyte type and the annealed temperature. Also, the two-factor interactions of electrolyte type and annealed temperature was also another key variable influencing the energy efficiency of DSSC. In addition to that, the determination coefficients (R2) was in good alliance, indicating a high association of the electrolyte and temperature with the DSSC efficiency. The optimum conditions of the energy efficiency occurred for a PAN-based gel polymer E2 when the photoanode was annealead at 350 · C, generating a highest energy efficiency of 1.5%..

APPENDIX

It is optional. Appendixes, if needed, appear before the acknowledgment.

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