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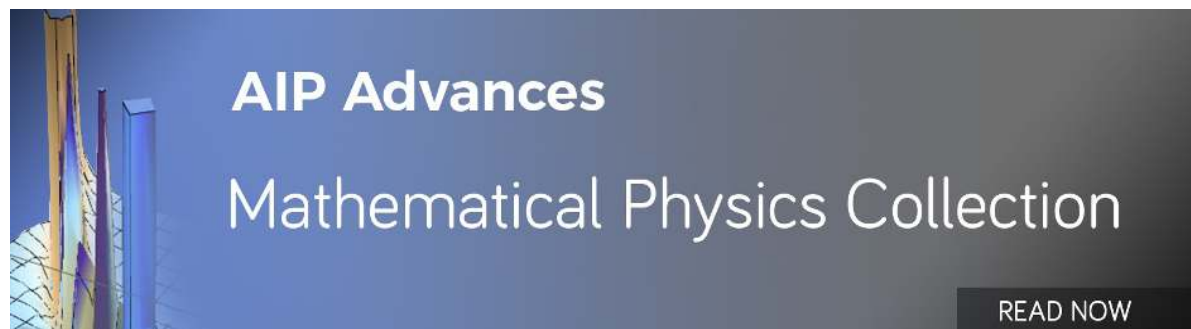
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## Influence of electrolyte proportion on the performance of dye-sensitized solar cells

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The performance of dye-sensitized solar cells (DSSC) depends strongly on the electrolyte. In this paper, the electrolytes with various solvents and different potassium iodide (KI) & iodine (I<sub>2</sub>) concentration were prepared and their influence on the DSSC performance were investigated. The results revealed DSSC with electrolyte prepared by organic solvent show better performance than that of inorganic solvent. Meanwhile, the increasing of KI concentration from 0.1 to 0.6 mol/L can effectively improve the short-circuit current density ( $J_{sc}$ ) from 0.2, correspondingly, the DSSC conversion efficiency ( $\eta$ ) increased from 0.04% to 0.11%. On the other hand, the  $J_{sc}$  and  $\eta$  of DSSC increased firstly then decreased with the I<sub>2</sub> concentration raised from 0.025 mol/L to 0.125 mol/L. It is worth to notice that the open-circuit voltage ( $V_{oc}$ ) of DSSC is almost unchanged with the concentration of KI and I<sub>2</sub>. In this paper, the best performance DSSC ( $J_{sc}=0.87$  mA/cm<sup>2</sup>,  $V_{oc}=0.57$  mV,  $\eta=0.23\%$ ) can be obtained with electrolyte at a KI and I<sub>2</sub> concentration of 0.6 mol/L and 0.075 mol/L respectively. Finally, the electrolytes used water and mixture of acetonitrile & ethylene glycol as solvents showed the latter presented better stability after setting at room temperature for 72 hours. © 2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). <https://doi.org/10.1063/1.5000564>

### I. INTRODUCTION

Among the new emerging photovoltaic technologies suitable for Building Integrated Photovoltaics (BIPV), dye-sensitized solar cells (DSSC), developed by O'Regan and Gratzel in 1991, have attracted great attention because of their various types, low cost, ease of manufacturing, unique applications, numerous options of efficiency enhancement, clean and affordable source of renewable energy.<sup>1-5</sup> A DSSC consists of an electrolyte that serves to restore the dye after oxidation of its molecules. The electrolyte basically contains a redox mediator in an organic matrix.<sup>6</sup> Different types of mediators are used in the electrolytes, such as I<sup>-</sup>/I<sub>3</sub><sup>-</sup>, SCN<sup>-</sup>/(SCN)<sub>2</sub>, S/S<sup>2-</sup> and Co(II/III).<sup>7</sup> The most mediator is the I<sup>-</sup>/I<sub>3</sub><sup>-</sup> couple because of slow recombination rate with injected electrons.<sup>8,9</sup> Traditionally, the liquid ionic electrolytes are commonly used in DSSC, but the quasi-solid or solid electrolytes gradually appeared in recent years, because of good stability and high efficiency of DSSC, but they also exhibited some defects such as high cost of preparation, low charge transfer, and the preparation process is complex, these factors limited their extensive application in reality. For now, the liquid ionic electrolyte based on redox couple of I<sup>-</sup>/I<sub>3</sub><sup>-</sup> still has potential application within a certain period of time due to many advantages in high diffusion rate, low cost, easy to prepare, and is beneficial to practical application. As is known to all, the function of electrolyte is a charge-transfer carrier, and its performance is closely relevant to the viscosity and conductivity of electrolyte which determine the rate and quantity of charge transfer respectively.<sup>10-12</sup> In fact, the viscosity and conductivity of the

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electrolyte can be regulated by its proportion. In this paper, several solvents with different concentration of KI and I<sub>2</sub> were prepared, and the influence of electrolytes proportions on the performance of DSSC was presented.

## II. EXPERIMENTAL

### A. Preparation of the TiO<sub>2</sub> photoanode

The commercial TiO<sub>2</sub> (P25, Degussa, Germany, a mixture of about 80% anatase and 20% rutile) powder 2 g, dilute acetic acid solution (1 ml), Triton X-100 (1 ml) and deionized water (5 ml) were mixed together and continuously grinded for 2 hours, TiO<sub>2</sub> slurry was obtained. Then the TiO<sub>2</sub> slurry was uniformly coated on the cleaned FTO glass through rolling of glass rods, and sintered at 450 °C for 0.5 hours after drying. TiO<sub>2</sub> photoelectrode was obtained and immersed in natural dyes with purple cabbage for 24 hours under darkness. Meanwhile, the low-cost graphite was used as counter electrode.

### B. Preparation of the electrolytes

#### 1. Solvent types

Three types of solvents were used to prepared electrolyte, include deionized water (25 ml), dimethylacetamide (25 ml), and mixture of acetonitrile (20 ml) and ethylene glycol (5 ml), and then put KI (0.9 g, powder) and I<sub>2</sub> (0.13 g, crystal) into these solvents respectively, allowing them stirring for 30 minutes and obtained samples Ai (i=1, 2, 3), then setting for 24 hours. The detailed proportions shown in Table I.

#### 2. Electrolytes with different KI concentration

Weighed 0.13 g I<sub>2</sub> crystal and dissolved in mixture solvent of acetonitrile (20 ml) and ethylene glycol (5 ml), and obtained the solution, then put KI powder (0.18 g, 0.36 g, 0.54 g, 0.72 g, 0.9 g, 1.08 g) into this solution individually, allowing them stirring for 30 minutes, and obtained sample Bi (i=1, 2, 3, 4, 5, 6), then setting for 24 hours. The detailed proportions shown in Table II.

#### 3. Electrolytes with different I<sub>2</sub> concentration

Weighed 1.08 g KI powder and dissolved in mixture solvent of acetonitrile (20 ml) and ethylene glycol (5 ml), and obtained the solution, then Put I<sub>2</sub> crystal (0.07 g, 0.14 g, 0.21 g, 0.28 g, 0.35 g) into this solution individually, allowing them stirring for 30 minutes, and obtained sample Ci (i=1, 2, 3, 4, 5), and then setting for 24 hours. The detailed proportions shown in Table III.

TABLE I. Electrolytes with different solvents.

Samples	Solvents	Electrolyte
A1	Water	0.5mol/L KI + 0.05mol/L I <sub>2</sub>
A2	Dimethylacetamide	0.5mol/L KI + 0.05mol/L I <sub>2</sub>
A3	Acetonitrile (20 ml) and Ethylene glycol (5 ml)	0.5mol/L KI + 0.05mol/L I <sub>2</sub>

TABLE II. Proportion of electrolytes with different KI concentration.

Samples	Solvents	Electrolyte
B1	Acetonitrile (20 ml) + Ethylene glycol (5 ml)	0.05mol/L I <sub>2</sub> + 0.1mol/L KI
B2	Acetonitrile (20 ml) + Ethylene glycol (5 ml)	0.05mol/L I <sub>2</sub> + 0.2mol/L KI
B3	Acetonitrile (20 ml) + Ethylene glycol (5 ml)	0.05mol/L I <sub>2</sub> + 0.3mol/L KI
B4	Acetonitrile (20 ml) + Ethylene glycol (5 ml)	0.05mol/L I <sub>2</sub> + 0.4mol/L KI
B5	Acetonitrile (20 ml) + Ethylene glycol (5 ml)	0.05mol/L I <sub>2</sub> + 0.5mol/L KI
B6	Acetonitrile (20 ml) + Ethylene glycol (5 ml)	0.05mol/L I <sub>2</sub> + 0.6mol/L KI

TABLE III. Proportion of electrolytes with different I<sub>2</sub> concentration.

Samples	Solvents	Electrolyte
C1	Acetonitrile (20 ml) + Ethylene glycol (5 ml)	0.6mol/L KI + 0.025mol/L I <sub>2</sub>
C2	Acetonitrile (20 ml) + Ethylene glycol (5 ml)	0.6mol/L KI + 0.050mol/L I <sub>2</sub>
C3	Acetonitrile (20 ml) + Ethylene glycol (5 ml)	0.6mol/L KI + 0.075mol/L I <sub>2</sub>
C4	Acetonitrile (20 ml) + Ethylene glycol (5 ml)	0.6mol/L KI + 0.100mol/L I <sub>2</sub>
C5	Acetonitrile (20 ml) + Ethylene glycol (5 ml)	0.6mol/L KI + 0.125mol/L I <sub>2</sub>

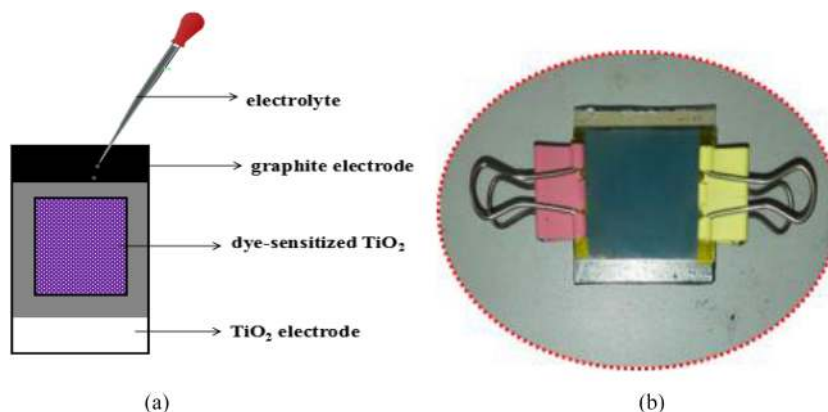


FIG. 1. Fabrication of DSSC: (a) schematic diagram and (b) actual picture.

### C. Fabrication and characterization of the DSSC

Dye-sensitized TiO<sub>2</sub> electrode contact with graphite electrode closely through clamp, and in order to achieve detection, both of electrodes should be staggered around 0.5 cm. Finally, different types of electrolytes are immersed in gap between the two electrodes through dropper, then the dye-sensitized solar cells were fabricated,<sup>13</sup> and this process shown in figure 1(a), and corresponding to actual picture shown in figure 1(b).

The DSSC performance was measured by solar cell tester (XJCM-8) under simulated sunlight (AM 1.5, 100 mW/cm<sup>2</sup>), and the open-circuit voltage (*V*<sub>oc</sub>), short-circuit current density (*J*<sub>sc</sub>), fill factor (*FF*), conversion efficiency (*η*) can be obtained.

## III. RESULT AND DISCUSSION

### A. Solvents on the DSSC performance

Figure 2 shows the current density-voltage (*J*-*V*) characteristics of DSSC with electrolyte prepared by different solvents. Table IV shows the performance parameters of the DSSC. It can be found sample A1 exhibited small short-circuit current and low open-circuit voltage using the water as solvent, and the conversion efficiency is only 0.01 % which can be attributed to most organic dyes are unstable in water. Meanwhile, compared sample A2 with A3, the latter shows larger short-circuit current density and conversion efficiency, which may be caused by different viscosity of organic solvents. Generally, low viscosity contributes to improve the charge transfer rate, which is beneficial to accelerate the dye redox process and increase the DSSC performance.<sup>14,15</sup>

### B. KI concentration on the DSSC performance

Figure 3 shows the current density-voltage (*J*-*V*) characteristics of DSSC with electrolyte prepared by different KI concentration. In this series, the I<sub>2</sub> concentration was fixed at 0.05 mol/L and acetonitrile (20 ml) and ethylene glycol (5 ml) was used as solvent. Table V shows the performance parameters of the DSSC.

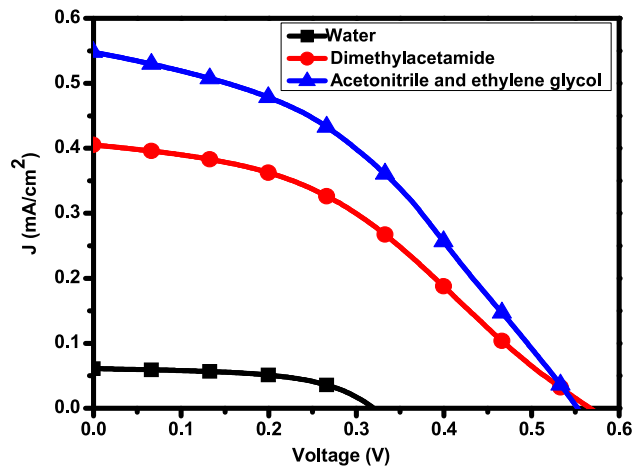


FIG. 2. J-V curves of DSSC with different solvents.

TABLE IV. Photovoltaic performance of DSSC with different solvents.

Samples	$J_{sc}$ (mA/cm <sup>2</sup> )	$V_{oc}$ (v)	FF (%)	$\eta$ (%)
A1	0.06	0.33	62.5	0.01
A2	0.40	0.56	45.6	0.10
A3	0.55	0.55	46.4	0.14

We can find the  $J_{SC}$  and  $\eta$  of DSSC increased monotonically with the KI concentration, but the  $V_{OC}$  almost unchanged. The increasing of KI concentration from 0.1 to 0.3 mol/L leads to an obviously increase of  $J_{sc}$  from 0.16 to 0.30 mA/cm<sup>2</sup>, and correspondingly the  $\eta$  improved from 0.04% to 0.09%. It can be explained by the equation (1)–(3).



Where  $D^+$  is the oxidative dye molecules. So, the increasing in KI concentration (supply  $K^+$  and  $I^-$  ionics) would increase  $I_3^-$  concentration because high concentration of  $I^-$  would promote to occur

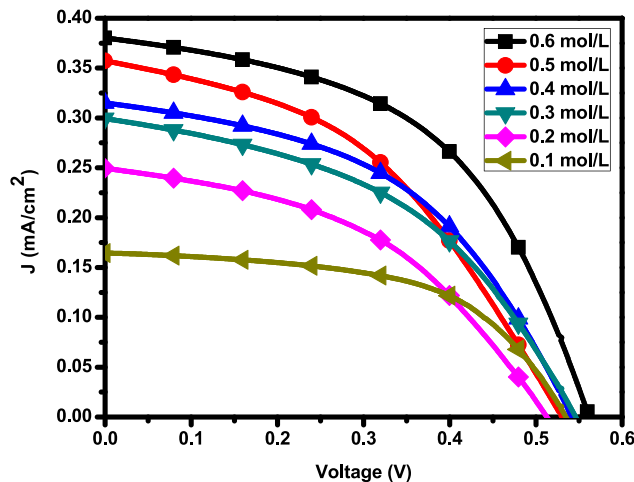


FIG. 3. J-V curves of DSSC with different KI concentrations.

TABLE V. Photovoltaic performance of DSSC with different KI concentrations.

Samples	$J_{sc}$ (mA/cm <sup>2</sup> )	$V_{oc}$ (V)	FF (%)	$\eta$ (%)
B1	0.16	0.55	47.7	0.04
B2	0.25	0.54	45.8	0.06
B3	0.30	0.55	52.0	0.09
B4	0.32	0.55	52.2	0.09
B5	0.36	0.52	54.3	0.10
B6	0.38	0.56	54.5	0.11

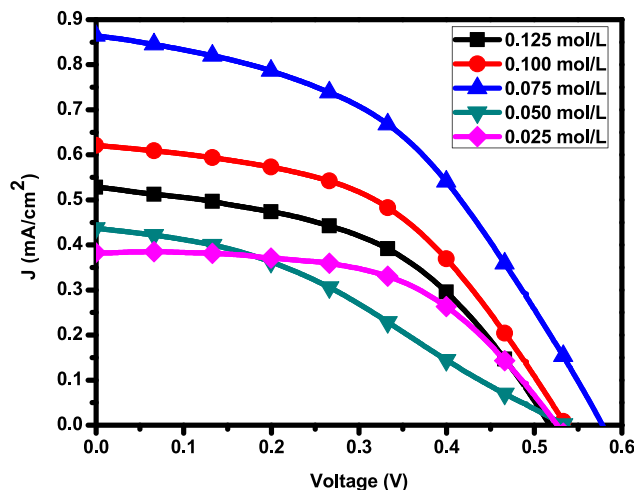
for equation (1).<sup>16</sup> This process could improve the charge transfer quantity from counter electrode (equation (2)) which is a key process to the reduction of excited dye (equation (3)). The increased quantity of reborn dyes can improve the absorptive capacity to light, and also beneficial to form the loop current, finally enhance the value of  $J_{SC}$ . Also, the increasing of  $J_{SC}$  and  $\eta$  get slowly because the dye reduction gradually saturated. As for the  $V_{OC}$ , it is well known that the open-circuit voltage is just determined by the potential difference between the Fermi level of the photoelectrode ( $E_{Fn}$ ) under illumination and the Nernst potential of the redox couple ( $E_{red}$ ) in the electrolyte. In this paper, the concentration of  $I^-/I_3^-$  has no influence on its  $E_{red}$ , and the  $E_{Fn}$  can be considered as constant.<sup>17</sup> Therefore, the  $V_{OC}$  is usually unchanged when the photoelectrode materials and redox couple fixed, just shown in Tables V and VI.

### C. $I_2$ concentration on the DSSC performance

Figure 4 shows the current density-voltage (J-V) characteristics of DSSC with electrolyte prepared by different  $I_2$  concentration. In this series, the KI concentration was fixed at 0.6 mol/L and acetonitrile (20 ml) and ethylene glycol (5 ml) was used as solvent. Table VI shows the performance parameters of the DSSC.

TABLE VI. Photovoltaic performance of DSSC with different  $I_2$  concentrations.

Samples	$J_{sc}$ (mA/cm <sup>2</sup> )	$V_{oc}$ (V)	FF (%)	$\eta$ (%)
C1	0.37	0.52	56.5	0.11
C2	0.44	0.52	52.6	0.12
C3	0.87	0.57	45.5	0.23
C4	0.62	0.53	53.2	0.17
C5	0.53	0.52	45.8	0.13

FIG. 4. J-V curves of DSSC with different  $I_2$  concentrations.

We can find the  $J_{SC}$  and  $\eta$  of DSSC were increased at first then decreased as the  $I_2$  concentration increased from 0.025 to 0.125 mol/L. Similar to the KI, the increasing of  $I_2$  concentration in the beginning leads to an increase of reduction for oxidative dye molecules (see equation (1)–(3)) then improve the cell performance. The maximum  $J_{SC}$  (0.87 mA/cm<sup>2</sup>) and  $\eta$  (0.23%) were obtained with 0.075 mol/L  $I_2$ . However, as the  $I_2$  concentration was over 0.075 mol/L, the  $J_{SC}$  and  $\eta$  of DSSC decreased to 0.53 mA/cm<sup>2</sup> and 0.13% respectively, G. Paruthimal Kalaigan with his coworker revealed that  $I_2$  can be converted to  $I_3^-$  fully and Iodine concentration is proportional to  $I_3^-$ .<sup>16</sup> According to equation (4).<sup>18</sup>

$$A = A_0 e^{k'l} \quad (4)$$

Where  $A_0$  is the viscosity of water at room temperature,  $k'$  is the instrument parameters and keep a constant,  $l$  is ion strength which depend on ion concentration.<sup>19</sup> From the equation (4), we could find that the increasing concentration of  $I_3^-$  would lead to increase the viscosity of the electrolyte solution.<sup>18,20</sup> So the electrolyte viscosity is proportional to  $I_2$  concentration. Increased viscosity of electrolyte would lead to strengthen the interaction of  $I_3^-$  in the electrolyte, caused the migration of charge transfer carrier ( $I_3^-$ ) blocked and declined reduction performance for the oxidative dyes, then reducing the absorption capacity of the nature dyes to light and lower  $J_{SC}$  was obtained.<sup>21–23</sup> Moreover, higher  $I_2$  concentration would increase the turbidity of electrolyte and then affect the dye absorption of sunlight.

#### D. Stability of dye - sensitized solar cell

The stability of DSSC is important to practical application, and electrolyte stability play a critical role in that. Figure 5(a) shows electrolyte with 0.6mol/L KI and 0.075mol/L  $I_2$  while the water as solvents. However, Figure 5(b) showed identical solutes except use the mixture solvent with acetonitrile and ethylene glycol. From figure 5(a), after setting 48 h, we find the color of electrolyte becomes darker, which attributed to the water evaporation.

And after 72 h, there is nothing left except KI crystal owing to the sublimation of  $I_2$  crystal. From figure 5(b), the electrolyte color gets lighter slowly with the time going which is

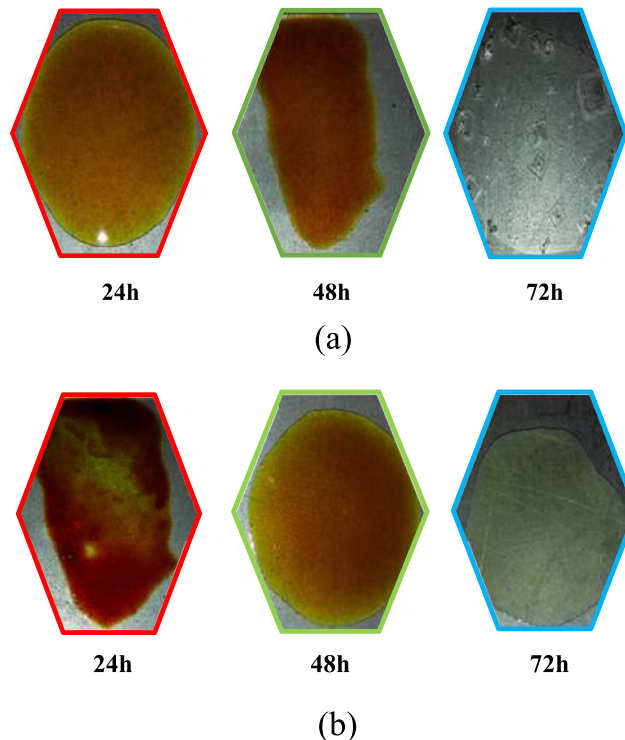


FIG. 5. Different setting time of electrolyte: (a) water as solvent (b) acetonitrile and ethylene glycol as solvent.

TABLE VII. Stability of photovoltaic performance of DSSC with different solvent.

Setting time	water		acetonitrile and ethylene glycol	
	$J_{sc}$ (mA/cm <sup>2</sup> )	$V_{oc}$ (V)	$J_{sc}$ (mA/cm <sup>2</sup> )	$V_{oc}$ (V)
24 h	0.130	0.390	0.870	0.570
48 h	0.100	0.100	0.470	0.335
72 h	/	/	0.110	0.254

attributed to the sublimation of I<sub>2</sub> crystal and the organic solvent show better stability than water.<sup>24</sup>

Table VII shows the photovoltaic parameters of DSSC used water and organic reagent as solvent after different setting time. It can be seen that the performance of DSSC obviously declined due to volatile or sublimation of electrolyte. However, compared to water solvent, the mixture solvent of acetonitrile and ethylene glycol exhibits better stability which is accordance with the results of figure 5.

#### IV. CONCLUSION

The influence of electrolytes proportion on the DSSC performance has been investigated. The results show DSSC with electrolyte prepared by organic solvent show better performance than that of inorganic solvent due to the lower viscosity. The increasing of KI concentration can reaction generated more I<sub>3</sub><sup>-</sup> ionic which effectively accelerates the reduction process of oxidative dye molecules and then improves the sunlight absorption. Therefore, the  $J_{SC}$  and  $\eta$  of DSSC increased with the increasing of KI concentration but get slowly improvement as the concentration further increased owing to the dye reduction saturated. Similar to KI, the  $J_{SC}$  and  $\eta$  of DSSC increased firstly as the I<sub>2</sub> concentration raised, but decreased as the concentration was over 0.075 mol/L. This phenomena can be explained by the fact that the excessive concentration would increase the viscosity of electrolyte and improve the ionic strength which blocked the migration of ionic (I<sup>-</sup>/I<sub>3</sub><sup>-</sup>). Moreover, the increased I<sub>2</sub> concentration make the transmittance of electrolyte declined. In addition, the  $V_{OC}$  of DSSC was found almost unchanged with the KI and I<sub>2</sub> concentration, because it is determined mainly by the potential difference between Fermi level of the photo electrode and Nernst potential of the redox couple. Finally, we found the electrolyte used organic solvent is more stable than that of water, this conclusion is helpful for the design and fabrication of DSSC with high stability.

#### ACKNOWLEDGMENTS

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