

Improvement of Dye Sensitized Solar Cells Efficiency Utilizing Diethyl Carbonate in PVA Based Gel Polymer Electrolytes

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Abstract— Low conductivity of gel polymer electrolytes (GPEs) containing double iodide salts is critical for efficiency in dye-sensitized solar cells (DSSCs). The presence of diethyl carbonate (DEC) plasticizer affects the amorphousness and ionic conductivity of polyvinyl alcohol (PVA)-based GPEs and DSSCs performance. In this work, PVA-based GPEs containing a variation of DEC have been produced, characterized, and applied in the DSSCs fabrication. The structural properties of GPEs were analyzed using X-ray diffraction (XRD). The ionic conductivity was determined from electrical impedance spectroscopy (EIS). Based on XRD, GPEs for all prepared compositions have been identified as an amorphous phase. From the EIS measurement, it was found that GPE with the composition of 5.46 PVA - 8.19 EC - 10.92 PC - 60.06 DMSO - 5.73 TPAI - 5.73 KI - 1.34 I₂ - 2.57 DEC (in wt. %) having highest conductivity of 11.19 ± 0.20 mS cm⁻¹ with activation energy, E_a of 0.09 eV. The graph of conductivity versus temperature following the Arrhenius rule has been plotted. The GPEs dominate the highest conductive with 2.57% of DEC and showed the DSSCs efficiency of 6.42%. Common DSSCs parameters resulted in short-circuit current density (J_{sc}) of 17.58 mA cm⁻², fill factor (ff) of 0.63, and open-circuit voltage (V_{oc}) of 0.58 V. In conclusion, DEC improves the ionic conductivity as well as amorphous properties of the GPE, and therefore enhance the DSSCs' efficiency.

Keywords— Diethyl carbonate; electrical impedance spectroscopy; tetrapropyl ammonium iodide; potassium iodide; dye-sensitized solar cells.

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I. INTRODUCTION

For a century, energy harvesting or solar cells have already proposed three types, the primary, secondary, and third solar cells. The solar cells applying silicon were regarded as the primary stage of solar cells, generating photo-current with more than 26% efficiency [1]. The secondary stage of thin-layer solar cells has shown that the current performance achieves an efficiency of 22% [2]. In the current stage of solar cells, such as quantum dots [3], perovskite [4], and dye-sensitized [5], among others, compete to concentrate high-efficiency conversion. Dye-sensitized solar cells (DSSCs), acknowledged as Grätzel cells, implement a high conversion of sunlight to electrical efficiency [5]. When improved efficiency occurs, this alternate stage has accomplished by integrating the primary with the secondary stage of solar cells. This third stage manifests that solar cells have excellent

quality, such as low production cost, simple assembly, and excellent efficiency [6]. The fabrication of DSSCs is very customizable, flexible, and efficient under natural or artificial light [7]. The assembly of standard DSSCs configures an electrolyte (liquid or gel type) located between a working electrode and a counter electrode [8].

Last decade, some researchers in DSSCs typically employed liquid electrolytes served as ion mediums for the assembly of DSSCs [9]. However, due to liquid electrolytes' critical challenge, the gel polymer electrolytes (GPEs) have been discovered and implemented, which promoted advantages and high DSSCs performance [10]. DSSCs assembly with GPE displayed better performance than DSSCs with liquid electrolytes, such as easy fabrication handling, reliable performance, and eliminating leakage problems [10].

In past DSSCs research work, single iodide salts consist of a small cation such as potassium iodide (K⁺I⁻), lithium iodide

(Li⁺I⁻), and sodium iodide (Na⁺I⁻) has been used [11]. Besides, iodide salt carrying a large cation has been introduced in electrolytes to enhance the performance of DSSCs. There was also research on the utilization of double iodide salts such as tetrapropyl ammonium iodide (Pr₄⁺I⁻) – K⁺I⁻ [12], tetrabutyl ammonium iodide (Bu₄⁺I⁻) – Li⁺I⁻ [13], and tetrahexyl ammonium iodide (He₄⁺I⁻) - rubidium iodide (Rb⁺I⁻) [14]. The combination of iodide salts has been confirmed to promote a high efficiency of DSSCs. Even though the double iodide salts system shows the high efficiency of DSSCs, this system shows a low conductivity value. Thus, a plasticizer has been introduced to enhance the conductivity and improve the DSSCs efficiency [15].

This research, GPEs containing polyvinyl alcohol (PVA), ethylene carbonate (EC), propylene carbonate (PC), and tetrapropyl ammonium iodide (TPAI) - potassium iodide (KI) was prepared. DEC plasticizer has been applied to improve the conductivity and efficiency of DSSCs. GPEs have been investigated using x-ray diffraction (XRD) and electrical impedance spectroscopy (EIS). In the assembly section of DSSCs, photoanode production consisted of double layers; first thin layer and second thin layer. Double-layer TiO₂ with different particle sizes influence the efficiency of DSSCs for better penetration by improving light-harvesting in this working electrode [16].

II. MATERIALS AND METHOD

A. Materials and gel polymer electrolyte preparation

Chemicals such as ethylene carbonate (EC), tetrapropyl ammonium iodide (TPAI), polyvinyl alcohol (PVA), propylene carbonate (PC), diethyl carbonate (DEC), and potassium iodide (KI) were obtained from Sigma Aldrich. TiO₂ powder was purchased from Aeroxide brand. Platinum (Pt) liquid and Ruthenium N3 dyes were received from Solaronix. Dimethyl sulfoxide (DMSO) has obtained from Friendemann Schmidt Chemicals.

The preparation GPEs have fixed amounts of 0.2 g of PVA, 2.2 g of DMSO, 0.3 g of EC, 0.4 g of PC, and 0.42 g of TPAI-KI. The amount of DEC has been varying and precisely weighted. In Table 1, the weight percentage for each material used was listed. All prepared GPEs have been labeled as GP. Firstly, the preparation of GPEs was done by dissolving the polyvinyl alcohol in a solvent (DMSO) using a glass container. The double iodide salt (TPAI-KI) was then well stirred in a EC and PC solvents mixture using a different beaker.

Lastly, DEC has been added to the mixture. This mixture was stirred at a temperature of 60 °C on the heater plate. The heater was turned off once the mixture was turned into gel. Before the gel was slowly cooled down to 25°C, iodine (I₂) crystal has been added in GPEs. In the end, all GPEs were stored in a closed desiccator containing silica gels in a dark place. These GPEs were ready for use in the characterization and assembly of DSSCs.

TABLE I
WEIGHT PERCENTAGE FOR ALL GPEs PREPARED

	(wt. %)							
GPE	PVA	EC	PC	DMSO	KI	TPAI	I ₂	DEC
GP1	5.61	8.41	11.21	61.64	5.88	5.88	1.37	0.00
GP2	5.52	8.28	11.04	60.70	5.79	5.79	1.35	1.53
GP3	5.46	8.19	10.92	60.06	5.73	5.73	1.34	2.57
GP4	5.40	8.10	10.80	59.40	5.67	5.67	1.32	3.64
GP5	5.31	7.96	10.62	58.39	5.57	5.57	1.30	5.28

B. X-ray diffraction (XRD)

XRD is a specific measurement to identify the GPEs in an amorphous region or crystalline region. In the setting of XRD (BTX-II Olympus Benchtop), the x-ray beam (wavelength, $\lambda = 1.540598 \text{ \AA}$) was set at different angles of the samples. XRD measurement was done under the radiation of Cu-K α . For XRD measurement, 2θ range has been set between 5° and 55°. In order to obtain more reliable results, the XRD measurement was carried out with a high precaution measure by preventing any device from receiving frequency response close to the XRD machine. Thus, XRD measurement was operated in a closed room without any noise or disturbance.

C. Impedance Spectroscopy and Ionic Conductivity

HIOKI LCR Hi-Tester *ac* (frequency of 1 Hz to 100 kHz) with computer connection was utilized to measure electrical impedance spectroscopy (EIS). The GPEs have been placed in a sample holder (Teflon based). The Teflon holder (two stainless-steel electrodes) was then located in a closed oven with variation temperatures. The measurements began at an interval of 5°C from 25°C to 100°C. The Nyquist plot was obtained when the measurement was done finished. Then the conductivity was obtained from the formula of:

$$\sigma = \frac{t}{R_b A} \quad (1)$$

where t is the GPEs thickness; bulk resistance, R_b was observed from Nyquist plot which is the intersection between plotted graph and x -axis; A is contact area between electrode couples.

D. DSSCs parameters

Double layers of titanium dioxide (TiO₂) have been layered on the active surface layer fluorine-doped tin oxide (FTO) glass for photoanode preparation. The bottom layer has been prepared by grounding 0.5 g of TiO₂ (P90 in powder form) with 2 ml of nitric acid, HNO₃ (pH = 1) for 30 minutes using agate mortar. Once homogenous slurry was produced, it was deposited on the FTO glass. FTO glass with a drop the slurry was spun at 1000 rpm for the first five seconds and 2300 rpm for the next 60 seconds using a spin coater. The coated FTO glass was then sintered at 450°C for 30 minutes and labelled as TiO₂ first layer. The second layer of TiO₂ was prepared using a doctor blade technique which is coated on top TiO₂ first layer. 0.5 g of P25 TiO₂, 100 mg of carbowax, 2 ml of HNO₃ (pH=1) and ~1 or 2 drops of Triton X-100 was grounded and another slurry was produced. The slurry was ground in a span of 30 minutes or more. Slurry should appear in a homogeneous form without any clumps before it was spread to FTO glass. Then the FTO glass was sintered in a closed furnace at 450°C for 30 minutes. The TiO₂-FTO glass-coated solution was soaked in 0.3 M Ruthenium-based dye solution N3 with low humidity and dark environment place at the final preparation step. After the soaking process, the TiO₂-dye electrode was thoroughly cleaned using ethanol liquid to eliminate unbound or loosely bound TiO₂ particles. This TiO₂-dye electrode was ready to be used for DSSCs assembly.

DSSCs were carried out by constructing the GPEs, squeezed in between the counter electrode and photoanode. The counter electrode was prepared by coating the platinum metal solution on the active side of FTO glass. The working

electrode is a double-layer of TiO₂ (P90 and P25) with dye-soaked Ruthenium N3. The assembly of DSSCs was tested under visible white light (1000 W m⁻² intensity power: Xenon brand). This operating test was performed in a dark place. A source meter (Keithley 2400 brand) has been used to collect the photovoltaic parameter of DSSCs. Fill factor (*ff*) was calculated using the following equation:

$$ff = \frac{V_{max} \times J_{max}}{V_{oc} \times J_{sc}} \quad (2)$$

Here, fill factor, *ff* has obtained by current density at maximum power output, J_{max} (mA cm⁻²), and voltage at the maximum power output, V_{max} (V) were divided with the multiplication of the open-circuit voltage, V_{oc} (V) and short-circuit current density, J_{sc} (mA cm⁻²). DSSCs efficiency has been obtained from this equation:

$$\eta(\%) = \frac{V_{oc} J_{sc} ff}{P_{in}} \times 100\% \quad (3)$$

P_{in} represents the incident light power.

III. RESULTS AND DISCUSSION

A. XRD curves and FWHM

The plot of XRD curves displayed by the GPEs comprising PVA-PC-EC-DMSO-TPAI-KI and varying DEC amounts is illustrated in Fig. 1. The XRD curves for all GPEs, revealed a wide peak located at $2\theta = 20^\circ$ [17]. This wide peak was attributed to the addition of DEC plasticizer in the GPEs. DEC plasticizer has improved the amorphous region of the GPEs. In other words, this phenomenon happened when the plasticizer changed the flexibility and eased the segmental motion of the polymer chain for ion mobility [18].

Full-width half maximum (FWHM) was obtained by deconvoluting all the XRD patterns using a nonlinear least square analysis. 2θ (°) and FWHM values for each GPE were listed in Table 2. As seen in Table 2, the FWHM value for GP1 and GP2 samples was 0.20 and slightly increased to 0.21 for GP3, GP4 and GP5 samples. The results of FWHM and the wider peak of XRD curves confirmed that the GPEs were an amorphous type [19]. GPEs with an amorphous identity have been the propensity for ion mobility, thus, enhancing ionic conductivity [20]. The relationship between GPE amorphousness and ionic conductivity has been further discussed in the section on electrical impedance spectroscopy.

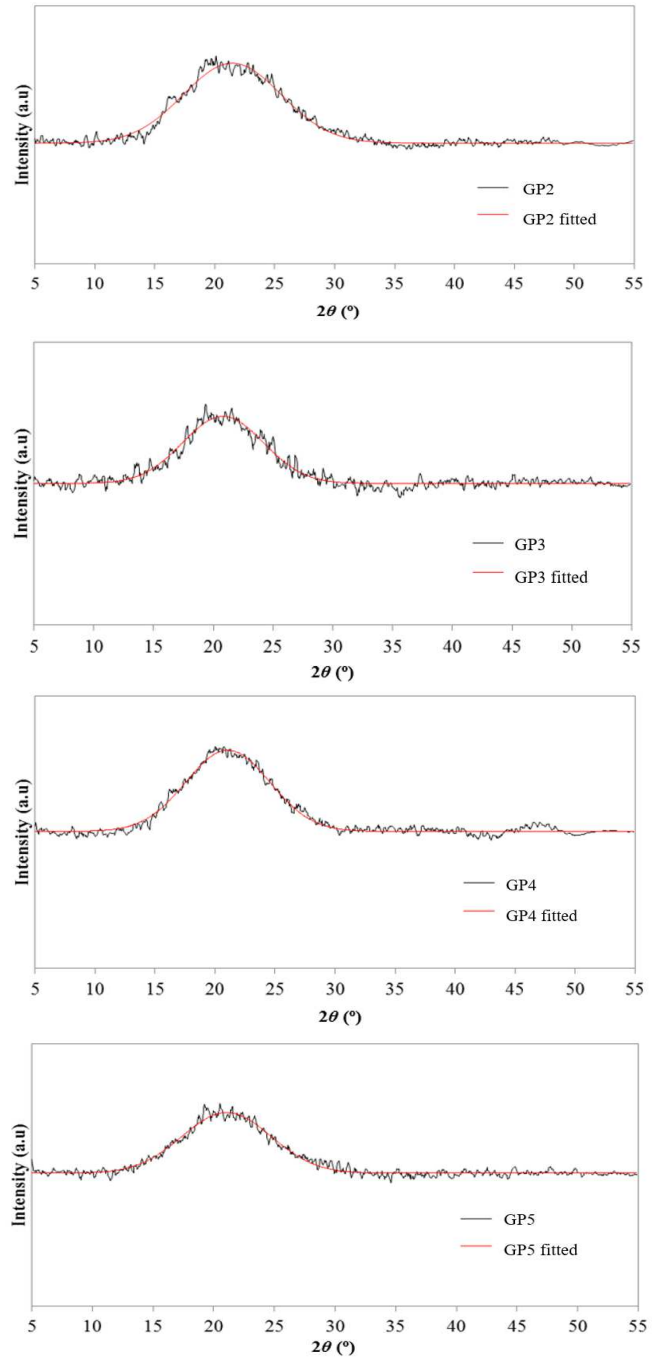
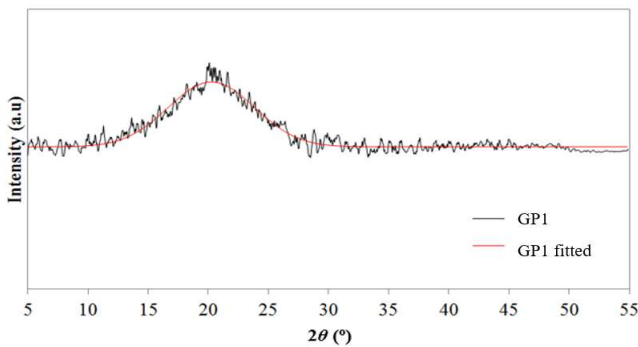


Fig. 1 XRD patterns for all GPEs

TABLE II
FWHM VALUES FOR ALL GEL POLYMER ELECTROLYTES

GPEs	2θ (°)	Full-width half maximum (rad)
GP1	20.4	0.20
GP2	22.0	0.20
GP3	21.0	0.21
GP4	21.2	0.21
GP5	21.2	0.21

B. Electrical impedance spectroscopy (EIS) analysis

Fig. 2 indicates that the conductivity values for different amounts of DEC in the GPEs. From the plotted graph, GP1 sample had a conductivity of 7.58 mS cm⁻¹. When the amount of DEC for GP2 and GP3 samples increased, the conductivity

value showed an increment. GP3 (2.57 wt.% of DEC) has the highest conductivity of 11.19 mS cm^{-1} . This phenomenon showed that the DEC plasticizer helps with the mobility of ions in the gel polymer electrolytes.

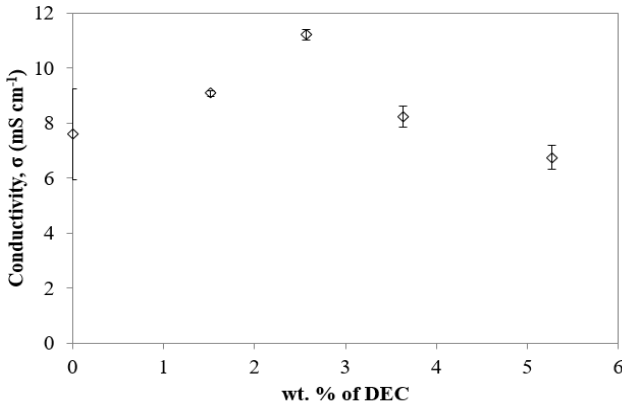


Fig. 2 Conductivity for all GPEs

Besides that, the plasticizer also improves the dissociation of ions in GPEs [15]. However, the graph indicated that the conductivity began to decrease 8.21 mS cm^{-1} for GP4 and 6.74 mS cm^{-1} for GP5. Decreasing conductivity showed that the amount of DEC was exceptionally high in the GPE system. $\log \sigma$ vs $1000/T$ has been plotted in Fig. 3. From the plotted graph, the conductivity is increase with different temperature.

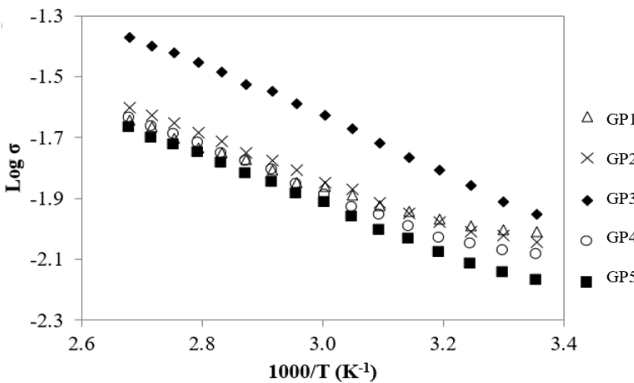


Fig. 3 $\log \sigma$ vs $1000/T$ for all GPEs

This graph obeys the Arrhenius rules, which explain that the conductivity will be elevated with increasing temperature. From Arrhenius rules, the minimum energy of chemical reaction begins or known as activation energy, E_a can be derived from the Arrhenius equation:

$$\sigma = \sigma_0 e^{\left(\frac{-E_a}{k_b T}\right)} \quad (4)$$

Where pre-exponential factor (σ_0), Boltzmann constant (k_b), and absolute temperature (T in Kelvin). Table 3 shows the activation energy (E_a) values for GPE samples containing DEC plasticizers with different compositions. GP3 (high conductance GPE) was the lowest activation energy (E_a) of 0.09 eV from the list. Despite this, GP5 (lowest conductivity) shows the highest activation energy (E_a) of 0.15 eV . It has been confirmed that a higher conductivity value showed low chemical reaction energy.

TABLE III
CONDUCTIVITY VALUES AND ACTIVATION ENERGY FOR ALL GPEs

GPEs	$\sigma_{RT} \text{ (mS cm}^{-1}\text{)}$	$E_a \text{ (eV)}$
GP1	7.58 ± 0.57	0.11
GP2	9.06 ± 0.11	0.10
GP3	11.19 ± 0.20	0.09
GP4	8.21 ± 0.39	0.14
GP5	6.74 ± 0.45	0.15

C. DSSCs parameters

The current density versus voltage (J-V) graph has been plotted in Fig. 4 for DSSCs utilizing all GPE samples. The main parameters of DSSCs, such as short-circuit current density (J_{sc}) and open-circuit voltage (V_{oc}) have been classified and listed in Table 4. The fill factor (ff) and efficiency (η) have been calculated from equation 2 and 3. In Table 4, the short-circuit current density, J_{sc} was observed from 12 to $\sim 18 \text{ mA cm}^{-2}$. GP1 demonstrated the value of J_{sc} of 12.56 mA cm^{-2} while GP2 showed an increment of 16.13 mA cm^{-2} as DEC plasticizer has been added in this GPE sample. This phenomenon confirmed that the DEC plasticizer improved ionic conductivity in GPEs, and increased the short-circuit current density, J_{sc} [10]. The increase in J_{sc} associated to the improvement in ions mobility in the electrolyte GP3. Specifically, the short-current density, J_{sc} of DSSC was revealed by the charge mobility conductivity from dye-excited state move to the TiO_2 metal oxide layer conduction band and also from the platinum electrode to the ionized dye [21]. In this research, the dye-sensitized solar cell assembly with GPEs consisted of diethyl carbonate improved the charge carriers transport from a platinum electrode to the ionized dye, thus, enhancing J_{sc} from 14.10 mA cm^{-2} for GP1 sample to 16.13 mA cm^{-2} for the GP2 sample. Besides, GP3 has the highest J_{sc} , which shows a value of 17.58 mA cm^{-2} . Otherwise, GP3 has the highest conductivity value, also has the highest short-circuit current density, J_{sc} . For the GP4 and GP5 samples, the J_{sc} values decreased gradually to 14.37 mA cm^{-2} and 12.42 mA cm^{-2} , respectively. This phenomenon can be explained by the decrease of conductivity in the GPEs that reduced the short-circuit current density, J_{sc} . As seen in Table 3, the conductivity of GP4 and GP5 is reduced by about $\sim 40\%$ with the GP3 sample. This statement confirms that the J_{sc} for both GP4 and GP5 is reduced by about $\sim 40\%$ from GP3 sample.

The process of electron transfer from the excited level of dye to the TiO_2 conduction band has remained the same for all DSSC assembly with GPEs as the V_{oc} is $\sim 0.60 \text{ V}$. V_{oc} resolution suggested that the energy level for the Fermi level with the TiO_2 conduction band remains unchanged. This phenomenon is due to the charge carrier density in the gel polymer electrolyte that retained both energy levels, thereby sustaining the transfer of an electron from dye-excited state jump to the TiO_2 conduction band.

In Table 4, the DSSCs fill factors (ff) were in a good range between 0.62 and 0.64. Fill factor (ff) values suggested that all GPEs had a good contact between the photoanode and platinum metal electrode; electron transfer flowed in the circuit without any loss [10]. The efficiency of the DSSCs for the GP1 sample was only 5.51%. However, due to the existence of DEC plasticizer, DSSCs for GP3 have the highest efficiency of 6.42%.

In Table 5, the lists revealed the correlation between conductivity and efficiency for DSSCs with GPE samples. The value of efficiency follows the conductivity value. GP3 proved that the highest conductivity resulted in the highest efficiency of DSSCs. Other researchers have concluded that the existence of plasticizer had a positive impact on DSSCs efficiency [22-24].

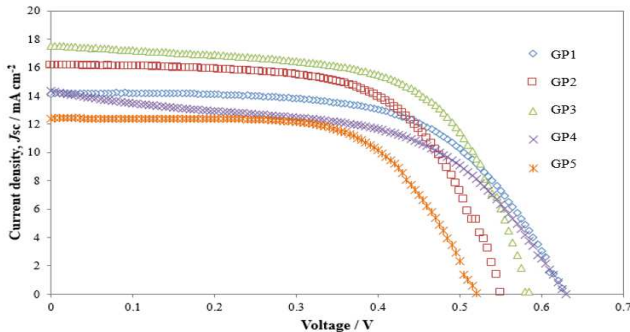


Fig. 4 J - V curves for all DSSCs with GPEs

TABLE IV
DSSCs PARAMETER FOR ALL GPES

DSSC	J_{sc} (mA cm^{-2})	V_{oc} (V)	Fill factor	η (%)
GP1	14.10	0.63	0.62	5.51
GP2	16.13	0.63	0.63	5.59
GP3	17.58	0.58	0.63	6.42
GP4	14.37	0.63	0.64	4.89
GP5	12.42	0.62	0.64	4.13

TABLE V
CONDUCTIVITY AND DSSCs EFFICIENCY FOR ALL GPES

GPEs	σ_{RT} (mS cm^{-1})	η (%)
GP1	7.58	5.51
GP2	9.06	5.59
GP3	11.19	6.42
GP4	8.21	4.89
GP5	6.74	4.13

IV. CONCLUSIONS

The PVA-based GPEs containing of EC-PC-DMSO-TPAI-KI-I₂, was prepared and characterized. XRD uncovered the identity of the GPEs. The broad peak of GPEs XRD confirmed that the samples were in the amorphous phase. The GP3 sample containing 2.57 wt.% DEC resulted in conductivity of $11.19 \pm 0.20 \text{ mS cm}^{-1}$ ($E_a = 0.09 \text{ eV}$). GP3 also displayed the DSSCs efficiency of 6.42%. Other GP3 DSSCs parameters; short-circuit current density, J_{sc} of 17.58 mA cm^{-2} , fill factor (ff) of 0.63 and open-circuit voltage, V_{oc} of 0.58 V were precisely obtained. Furthermore, GP3 showed the best composition in the GPE system, in which DEC plasticizer improved conductivity and performance of DSSC.

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