

## Deep Eutectic Solvent (DES) Based on Choline Chloride and Mono-, Di-, Poly-Ethylene Glycol as KI/I<sub>2</sub> Electrolyte Solvents on DSSC Devices

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**Abstract:** Deep eutectic solvent (DES) has high viscosity and electrical conductivity values, so it can be used as an electrolyte solvent in dye-sensitized solar cells (DSSCs). This research was conducted to produce DES based on choline chloride (ChCl) and ethylene glycol (EG), diethylene glycol, and polyethylene glycol-400, which were then used as KI/I<sub>2</sub> couple redox electrolyte solvent to improve the DSSC performance. The synthesis was carried out by mixing each component in several variations of the mole fraction of ChCl ( $x_{\text{ChCl}}$ ) at 80 °C for 15 min, and then was characterized by their pH, freezing point, density, viscosity, and electrical conductivity. A mixture that meets the criteria as a eutectic solvent and has a freezing point of less than -18 °C with the highest electrical conductivity value is DES ChCl:EG with  $x_{\text{ChCl}}$  0.3 and  $x_{\text{ChCl}}$  0.4. Both DESs were then used as a solvent for KI/I<sub>2</sub>, combined with acetonitrile in various compositions. The electrolyte with the highest electrical conductivity value was KI/I<sub>2</sub> dissolved in ChCl:EG with  $x_{\text{ChCl}}$  0.3 solvent 6:4 v/v, and then employed in DSSC device. The best performance of DSSC ( $I_{\text{sc}} = 0.155 \text{ mA/cm}^2$ ;  $V_{\text{oc}} = 0.465 \text{ V}$ ;  $P_{\text{max}} = 0.719 \text{ W}$ ;  $\eta_{\text{max}} = 0.072\%$ ) was produced under a light intensity of 0.1 W/cm<sup>2</sup>.

**Keywords:** deep eutectic solvent; dye-sensitized solar cell; electrolyte; viscosity; electrical conductivity

### ■ INTRODUCTION

Dye-sensitized solar cell (DSSC) is a third-generation solar cell introduced by Professor Grätzel in 1991. DSSC was chosen as an alternative to convert solar energy into electrical energy because of its low production price, and the ingredients are easier to obtain. In addition, each component of the DSSC can be modified and optimized separately. The electrolyte is one of the main components whose performance must be optimized to increase the efficiency of DSSC. Electrolytes play an essential role in the electron transfer process in DSSC. The function of the electrolyte in DSSC is to replace the loss of electrons in the dye's valence band when the dye absorbs visible light [1]. The electrolyte used in DSSCs is generally a liquid containing an I<sup>-</sup>/I<sub>3</sub><sup>-</sup>. Anion I<sup>-</sup>/I<sub>3</sub><sup>-</sup> can be obtained from the compound KI/I<sub>2</sub>, which is generally dissolved in acetonitrile [2-3].

Acetonitrile is a widely used organic solvent because it has good solvation ability for polar and non-polar compounds. However, as an electrolyte solvent in DSSCs, acetonitrile has very low viscosity and electrical conductivity values. The viscosity of acetonitrile is 0.352 cP with an electrical conductivity of  $7 \times 10^{-3} \text{ mS/cm}$  [3]. The low viscosity value can result in leakage or instability, which will affect the electrolyte's quality and reduce the DSSC's efficiency. Therefore, it is necessary to use an alternative solvent with a higher viscosity and electrical conductivity value to replace acetonitrile as an electrolyte solvent to increase DSSC's efficiency. The solvent is known as deep eutectic solvent (DES).

DES is a binary or ternary mixture consisting of a hydrogen bond donor (HBD) and a hydrogen bond acceptor (HBA), which are strongly bound due to interactions with hydrogen bonds. The HBD and HBA

proper molar ratio will produce a eutectic mixture with a freezing point much lower than that of each pure constituent. DES is classified as an ionic liquid (IL), so its electrical conductivity value is relatively large. This is because when an IL is subjected to an electric field, a charge transport event or ion mobility will be associated with an empty space (hole). This ability affects the size of the electrical conductivity value in a liquid [4]. Because of its similarity with ILs, DES has started to be widely applied in electrochemical devices [5-7].

In recent years, choline iodide: glycerol and choline chloride (ChCl): glycerol-based DES have been used as part of the electrolyte composition for DSSC by Jhong et al. [8] and Boldrini et al. [9]. The reported DSSC showed an energy conversion efficiency of 1.70–3.88%. However, the efficiency values were still lower than DSSC using acetonitrile at the same conditions, namely 4.90%. This study aims to synthesize DES, which is then used as a solvent for KI/I<sub>2</sub> in DSSC devices. DES was synthesized by mixing ChCl with several hydrogen bond donor compounds such as ethylene glycol (EG), diethylene glycol (DEG), and polyethylene glycol (PEG). DES produced from a mixture of ChCl and EG has been studied to have a viscosity value of 36 cP and an electrical conductivity value of 7.61 mS/cm [10]. The DES viscosity value is 100× greater than that of acetonitrile. They have a 1000× more excellent electrical conductivity value compared to acetonitrile. Meanwhile, in this study, DEG and PEG were also used as HBD to study the effect of adding alkyl chains to the function of DES as an electrolyte solvent. The volume ratio between DES and acetonitrile, which can produce DSSC with the best efficiency, was also reported.

## ■ EXPERIMENTAL SECTION

### Materials

The materials used in this study include choline chloride (ChCl, ≥ 99%, Sigma Aldrich), EG (C<sub>2</sub>H<sub>6</sub>OH, ≤ 100%, Supelco), DEG (C<sub>4</sub>H<sub>10</sub>O<sub>3</sub>, 99%, Sigma Aldrich), PEG 400 (HOCH<sub>2</sub>(CH<sub>2</sub>OCH<sub>2</sub>)<sub>9</sub>CH<sub>2</sub>OH, ≤ 100%, Sigma Aldrich), acetonitrile (C<sub>2</sub>H<sub>3</sub>N, 99.8%, Sigma Aldrich), ethanol absolute (C<sub>2</sub>H<sub>5</sub>OH, ≥ 99.5%, Supelco), titanium(IV) oxide (TiO<sub>2</sub> anatase nanopowder, < 25 nm

particle size, 99.7% trace metals basis, Sigma Aldrich), potassium iodide (KI, ≤ 100%, Supelco), and iodine (I<sub>2</sub>, ≤ 100%, Supelco). Dye (Ruthenizer complex 535-bistba, N719), platisol T/SP (Pt paste), and transparent conductive oxide (TCO) glass with type fluorine-doped tin oxide (FTO) coated TEC-7 conductive glasses were purchased from Solaronix.

### Instrumentation

The instruments used in this study included analytical balances, ovens, THERMOLYNE furnaces, IKA C-MAG HS 7 hotplates, standard glassware, spatulas, thermometers, Iwaki Pyrex pycnometer, Oswald Iwaki Pyrex viscometer, Hanna conductivity meter, 500-Watt halogen lamp, solar power meter BTU W/m<sup>2</sup> tester handheld portable and scientific multimeter 6.5-digit Agilent 34461 A.

### Procedure

#### DES preparation

ChCl solid was weighed with a ChCl mole fraction ( $x_{\text{ChCl}}$ ) of 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, and 1.0, and then they were added into a beaker glass separately. EG was weighed according to the calculation with  $x_{\text{ChCl}}$  as above and placed into different beakers so that the total mass of the mixture of ChCl and EG was 5 g. The beaker glass containing ChCl and EG was then heated using a hotplate at 80 °C for 15–40 min to obtain dry ChCl solid (white crystals). The EG liquid was then poured into a beaker containing ChCl. The mixture was then stirred using a magnetic stirrer while heated at 80 °C for 15 min until a homogeneous mixture was obtained. The mixture was placed in the bottle at room temperature until further treatment [11]. The preparation of DES with DEG and PEG 400 was done with the same procedure. Homogeneous mixtures were selected for the characterization stage, including measurement of pH, freezing point, density, viscosity, and electrical conductivity.

#### Electrolyte preparation and characterization

Homogeneous mixtures (ChCl:EG, ChCl:DEG, and ChCl:PEG 400) were mixed with acetonitrile, respectively, with variations in the v/v composition of

the mixture (10:0, 8:2, 6:4, 4:6, 2:8 and 0:10) and the total volume of solvent was 10 mL. KI and I<sub>2</sub> weighed as much as 0.83 and 0.13 g, respectively. They were put into a beaker containing DES and acetonitrile, then wrapped in aluminum foil, and stirred using a magnetic stirrer for 30 min [12]. The electrolyte solution was then transferred into a dark vial. The electrolyte solution was observed for 24 h. The homogeneous electrolyte was further characterized by measuring density, viscosity, and electrical conductivity.

#### **DSSC fabrication and performance analysis**

**FTO glass preparation.** The FTO glass was cut into a square of 2 × 2 cm<sup>2</sup>. The glass was then washed using 70% technical ethanol and sonicated for 30 min. Then, the glass was dried at a temperature of 100 °C for 1 h. The conductive side of the glass was determined [3].

**DSSC assembly.** An active side (1 × 1 cm<sup>2</sup>) of FTO glass was made by attaching transparent insulation. The TiO<sub>2</sub> anode and Pt cathode paste were deposited on different FTO glass. Photoanode was prepared by adopting the method of Rahmalia et al. [2]. The anode used was TiO<sub>2</sub> with a 5% TMK (NH<sub>4</sub>OH-treated metakaolinite which was previously produced by Rahmalia et al. [2]) mixture. Anatase TiO<sub>2</sub> weighing 2.3751 g and TMK weighing 0.1264 g were measured and then added to 15 mL of absolute ethanol, 800 μL of Triton-X, and 600 μL of acetylacetone. The mixture was stirred for 24 h until a paste was formed. The resulting anode paste was sonicated for 3 h. The anode was calcined at a temperature of 450 °C for 30 min, while the cathode was calcined at 400 °C for 5 min [3].

The anode layer was immersed in a sensitizer solution (1% ruthenium complex prepared by dissolving 0.005 g of ruthenium dye into 5 mL of ethanol) for 24 h [13]. Furthermore, the anode and cathode were assembled like a sandwich and locked using a binder clip. Then, a few drops of electrolyte were through the gap between the two glasses until the electrolyte solution covered and filled all parts of the glass. The cells formed are then sealed with glue [14].

**DSSC performance analysis.** The light source used in the DSSC performance analysis was a 500 W halogen lamp. The test was carried out with variations in light

intensity of 0.005–0.100 W/cm<sup>2</sup>, which was controlled by dimer and measured using a solar power meter. The values of cell short circuit current (I<sub>sc</sub>) and open-circuit voltage (V<sub>oc</sub>) were measured using the scientific multimeter Agilent 34461 A.

## ■ RESULTS AND DISCUSSION

### **DES Synthesis and Characterization**

The DES in this study is the result of a mixture of quaternary ammonium salt (HBA), namely ChCl and a HBD, namely EG, DEG, and PEG-400. The interaction occurs in the process of forming DES is an intermolecular hydrogen bond formed by the chloride anion group (Cl<sup>-</sup>) in the HBA and the hydroxyl group in the HBD. DES is also described as a mixture of Brønsted or Lewis acids and bases, so that the hydrogen bonds formed in all these systems could cause a significant freezing point depression [15].

The results of DES preparation show that a mixture of ChCl:EG and ChCl:DEG at x<sub>ChCl</sub> 0.1, 0.2, 0.3, and 0.4 were liquid and transparent, indicating the formation of DES in these compositions. The mixture with x<sub>ChCl</sub> 0.5–1.0 was crystallized. The acid dissociation, which releases hydrogen atoms, causes choline salts to form so that the mixture cannot change to the liquid phase at room temperature [15]. As for the ChCl:PEG 400 mixture, only the x<sub>ChCl</sub> 0.1 was liquid and transparent. Therefore, only the mixture with x<sub>ChCl</sub> 0.1 was indicated as DES.

The prepared DES was observed at room temperature for 24 h to evaluate their homogeneity. Several mixtures of ChCl:EG, ChCl:DEG, and ChCl:PEG 400 (as presented in Table 1) were continued in the characterization stage. They were a mixture that did not change, either phase changes or the formation of crystal deposits.

The pH condition affects solubility because solutes tend to be more soluble in solvents under acidic or neutral conditions than in alkaline conditions [16]. Therefore, pH measurements were carried out to determine whether the DES produced was acidic, neutral, or basic. Measurements showed that all three types of DES were detected at pH 6, so it can be concluded that the DES prepared in this study were weakly acidic.

**Table 1.** The DES composition was carried forward to the characterization stage

DES type	$x_{\text{ChCl}}$	$x_{\text{EG}}$
DES <sub>a1</sub>	0.1	0.9
DES <sub>a2</sub>	0.2	0.8
DES <sub>a3</sub>	0.3	0.7
DES <sub>a4</sub>	0.4	0.6
DES <sub>b1</sub>	0.1	0.9
DES <sub>b2</sub>	0.2	0.8
DES <sub>b3</sub>	0.3	0.7
DES <sub>b4</sub>	0.4	0.6
DES <sub>c1</sub>	0.1	0.9

Mixing two components at a certain mole fraction formed DES will cause a significant decrease in freezing point so that it will have a lower freezing point than each of its pure constituents [11]. The constituents of DES have different freezing points. The ChCl, EG, DEG, and PEG 400 compounds have a freezing point at 303 °C [15], -12 °C [17], -6.5 °C [18], and -4 °C [19], respectively. There was no phase change in DES<sub>a1</sub>-DES<sub>a4</sub> and DES<sub>b1</sub>-DES<sub>b4</sub>. They did not freeze when placed in the freezer at a temperature range of -15 to -18 °C for 24 h. This follows the definition of DES because it has a freezing point below the freezing point of the single substance, namely EG, and DEG, and is much lower than the freezing point of ChCl. Meanwhile, DES<sub>c1</sub> undergoes crystal formation sometime after being put in the freezer. It freezes after being placed for 5 min when a temperature of 0 °C. In this case, the mixture does not reach the eutectic point because it has a freezing point with a higher freezing rate than one of its pure constituents, in this case PEG 400. Therefore, in the next step, only DES<sub>a1</sub>-DES<sub>a4</sub> and DES<sub>b1</sub>-DES<sub>b4</sub> were used as electrolyte solvents.

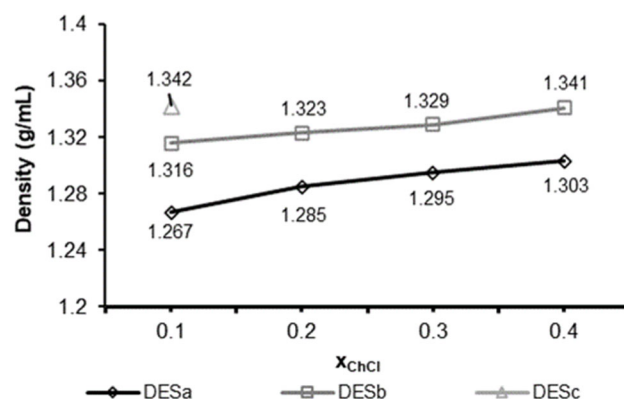
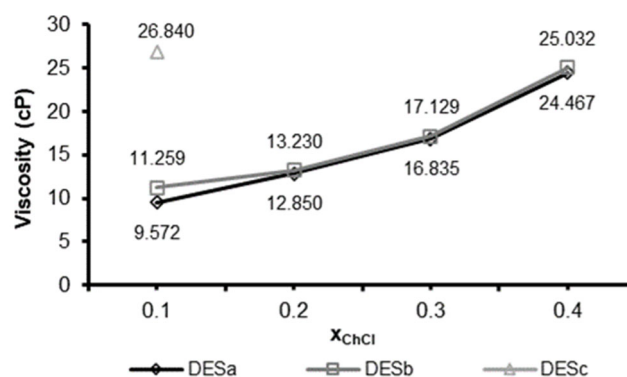
Density and viscosity have a close relationship as factors that affect mass transfer [11]. The type of HBD influences the density of DES. The density of DES will increase with the increase in the number of hydroxyl groups or the length of the HBD chain [4]. The DES constituents' molar ratio also affects the density value [20]. Fig. 1 shows the density measurement results of DES formed in this study.

Based on Fig. 1, it can be observed that the addition of alkyl chain lengths of constituent increased the DES

density value. The longer the alkyl chain, the greater the molecular weight. The increase in molecular weight affects increasing the density value [21]. The EG, DEG, and PEG-400 have a molecular weight of 62.07, 106.12, and 400.00 g/mol, respectively. The DES density value increased along with the increase in the  $x_{\text{ChCl}}$  in the mixtures. This was due to the contribution of the density of the pure constituents of the mixtures. ChCl, EG, DEG, and PEG-400 have a density of 1.19, 1.11, 1.12, and 1.13 g/mL. Mixing these compounds with ChCl causes an increase in density.

Like density, viscosity is often associated with a hydrogen bond network between HBA and HBD, corresponding to free volume [22]. The viscosity of the eutectic mixture is influenced by the chemical properties of the constituents (HBA and HBD), temperature, and water content [4]. The results of the viscosity measurement of DES in this study are presented in Fig. 2.

Fig. 2 shows that an increase in the  $x_{\text{ChCl}}$  causes an increase in the viscosity value. The increased viscosity

**Fig 1.** The relationship of  $x_{\text{ChCl}}$  and DES density**Fig 2.** The relationship of  $x_{\text{ChCl}}$  and DES viscosity

value due to adding the  $x_{\text{ChCl}}$  was caused by the high and extensive network of hydrogen bonds between each component resulting in a very viscous mixture [21]. High viscosity can cause an increase in the solvent diffusion coefficient so that the reaction rate will decrease [8]. However, the increase in viscosity also affects slowing the evaporation process to prevent leakage of the electrolyte system after DES was used as an electrolyte solvent in DSSC [23].

Fig. 3 shows the results of the DES electrical conductivity measurements produced in this study. Based on Fig. 3, it can be observed that the addition of the alkyl chain length of the constituent resulted in a decrease in the value of the electrical conductivity of DES. In general, the value of electrical conductivity is influenced by the viscosity of the solution, where the high viscosity of the solution will reduce the movement of free ions [11]. However, there was an increase in the electrical conductivity value when  $x_{\text{ChCl}}$  increased. This is because the addition of ChCl will result in the formation of more hydrogen bonds so that the intermolecular interactions due to the bond cause the electrical conductivity value to increase. Hydrogen bonding allows intermolecular delocalization, affecting increased electrical conductivity [24]. However, there was a drastic decrease in the electrical conductivity of  $\text{DES}_{a4}$ . It is probably due to the increase in bond density due to the increasing number of molecule interactions. The molecular interactions can cause an increase in viscosity, resulting in a decrease in electrical conductivity due to the hindered movement of

ions and electrons [25]. Based on the molecular structure, DEG has a longer alkyl chain than EG. However, in DEG, there is a lone pair of electrons in the ether group (R-O-R'). The lone pair of electrons acts as a medium that increases ionic mobility [26]. These factors allow the electrical conductivity of  $\text{DES}_{b4}$  not to decrease while  $\text{DES}_{a4}$  has decreased electrical conductivity.

### Electrolyte Preparation and Characterization

The DES used as an electrolyte solvent in this work was DES in the form of a homogeneous liquid that has the highest electrical conductivity value, namely  $\text{DES}_{a3}$ , with a density, viscosity, and electrical conductivity of 1.295 g/mL, 16.835 cP, and 9.940 mS/cm, respectively. In addition,  $\text{DES}_{b4}$  with a density, viscosity, and electrical conductivity of 1.341 g/mL, 25.032 cP, and 6.660 mS/cm, respectively. Meanwhile,  $\text{DES}_{c1}$  was not used as an electrolyte solvent because of its small electrical conductivity value of 0.213 mS/cm. There were several volume ratio variations in composition between acetonitrile and DES to dissolve KI and  $\text{I}_2$ . For further discussion, electrolytes resulting from dissolution with acetonitrile: $\text{DES}_{a3}$  10:0, 8:2, 6:4, 4:6, 2:8, and 0:10 v/v, are called  $E_{a1}$ ,  $E_{a2}$ ,  $E_{a3}$ ,  $E_{a4}$ ,  $E_{a5}$ , and  $E_{a6}$ , respectively. Meanwhile, the electrolyte resulting from dissolution with acetonitrile: $\text{DES}_{b4}$  10:0, 8:2, 6:4, 4:6, 2:8, and 0:10 v/v are referred to as  $E_{b1}$ ,  $E_{b2}$ ,  $E_{b3}$ ,  $E_{b4}$ ,  $E_{b5}$ , and  $E_{b6}$  respectively.

Fig. 4 shows that the absence of precipitates at  $E_{a4}$ ,  $E_{a5}$ ,  $E_{a6}$ ,  $E_{b4}$ ,  $E_{b5}$ , and  $E_{b6}$  indicated that DES could dissolve

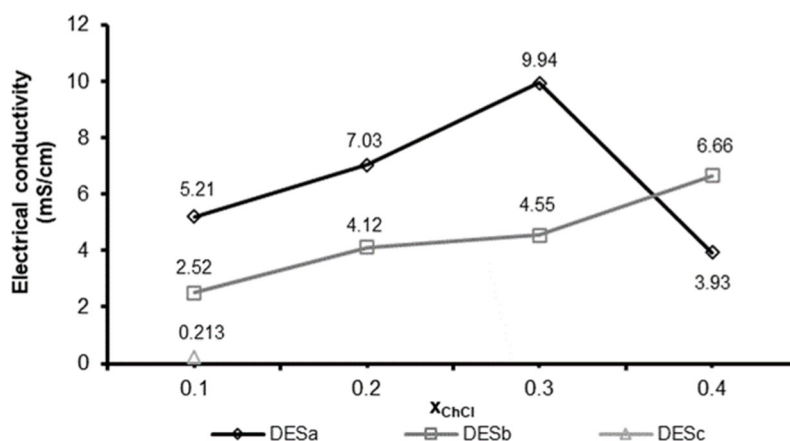


Fig 3. The relationship of  $x_{\text{ChCl}}$  and DES electrical conductivity

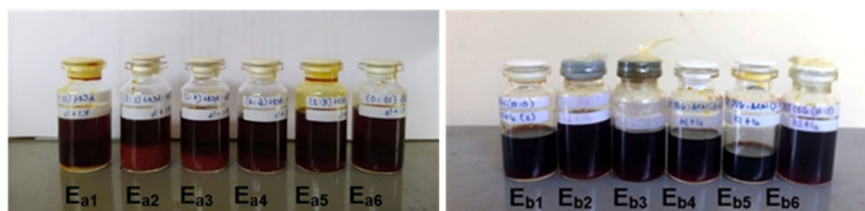


Fig 4. KI/I<sub>2</sub> electrolyte with acetonitrile:DES<sub>a3</sub> (left) and acetonitrile:DES<sub>b4</sub> (right) as solvent

the KI/I<sub>2</sub> redox couple better than acetonitrile. Hydrogen bonding in the solvent increases the solvent polarity or polarizability, which can cause higher solubility [27]. A white precipitate exists at E<sub>a1</sub>, E<sub>a2</sub>, E<sub>a3</sub>, E<sub>b1</sub>, E<sub>b2</sub>, and E<sub>b3</sub>. The precipitate is a residual KI compound that cannot be completely dissolved in the solvent. KI compounds have low solubility in organic solvents compared to their solubility in water [28].

Table 2 shows the variations in the volume ratio between acetonitrile and DES affecting the electrolyte's density, viscosity, and electronic conductivity. The relatively high density and viscosity of DES cause an increase in the viscosity of the electrolyte. The electrolyte using 100% acetonitrile as solvent had a density and viscosity of 0.875 g/mL and 0.796 cP, respectively. The presence of DES in the mixture causes the density and viscosity of the electrolyte also to increase.

An interesting thing happened in the observation of electronic conductivity. The electronic conductivity of the electrolyte increased with the addition of DES up to a volume ratio of acetonitrile:DES of 6:4 v/v (E<sub>a3</sub> and E<sub>b3</sub>).

The high viscosity of DES is thought to be the cause of the low electrolyte's electrical conductivity value when using more DES in the mixture. Although DES has a much higher electrical conductivity value than acetonitrile, when it is used as a KI/I<sub>2</sub> solvent, the high viscosity of DES can affect the redox system in the electrolyte so that the electrolyte with pure acetonitrile solvent has a higher electrical conductivity value. However, the right combination of these two solvents produces an electrolyte with the highest electrical conductivity value and optimum viscosity so that the mobility of electrons in the redox system is not disturbed and the electrolyte is not easily vaporized reducing the potential for leakage of the electrolyte system in DSSC.

#### DSSC Fabrication and Performance Analysis

DSSC is a medium used to observe the function and capabilities of KI/I<sub>2</sub>-based electrolytes with a combination of DES and acetonitrile as a solvent. DSSC performance analysis was performed on cells using E<sub>a3</sub>, E<sub>b3</sub>, and E<sub>a1</sub> separately. The DSSC performance tested

Table 2. Density, viscosity, and electrical conductivity of electrolytes

Ratio volume Acn:DES	Electrolyte type code	Density (g/mL)	Viscosity (cP)	Electrical conductivity (mS/cm)
10:0	E <sub>a1</sub>	0.875	0.796	14.140
8:2	E <sub>a2</sub>	0.923	3.865	22.240
6:4	E <sub>a3</sub>	0.955	8.639	25.200
4:6	E <sub>a4</sub>	1.043	12.679	15.350
2:8	E <sub>a5</sub>	1.192	16.372	12.190
0:10	E <sub>a6</sub>	1.325	18.466	11.850
10:0	E <sub>b1</sub>	0.875	0.796	14.140
8:2	E <sub>b2</sub>	0.996	5.653	15.780
6:4	E <sub>b3</sub>	1.028	10.712	22.500
4:6	E <sub>b4</sub>	1.086	16.068	18.900
2:8	E <sub>b5</sub>	1.205	22.425	8.050
0:10	E <sub>b6</sub>	1.368	26.682	6.710

includes short-circuit current ( $I_{sc}$ ), open-circuit voltage ( $V_{oc}$ ), maximum power ( $P_{max}$ ), and maximum energy conversion efficiency ( $\eta_{max}$ ). The effect of light intensity on the values of  $I_{sc}$ ,  $V_{oc}$ ,  $P_{max}$ , and  $\eta_{max}$  can be seen in Fig. 5.

The  $V_{oc}$  value is generated due to the difference in charge between the anode and cathode. The potential difference that occurred was expressed as a voltage value. Fig. 5 shows that the greater the light intensity was given, the greater the  $V_{oc}$  value also increased. Increasing the light intensity higher than  $0.06 \text{ W/m}^2$  does not significantly increase the  $V_{oc}$  value. Nunno et al. [29] reported the causes and effects of the voltage drop phenomenon. According to their research, the voltage drop is affected by several things, for example, unacceptable termination systems, hot spots, small-sized conductors, and non-uniform conductor materials. These factors will affect the system's increasing local temperature and resistance, causing a decrease in the voltage value [29]. In our research, an increase in light intensity also increases the local temperature of the system so that the  $V_{oc}$  value does not increase. It even experiences a slight decrease, an indication of a voltage drop.

Besides affecting the value of  $V_{oc}$ , light intensity also impacts the value of  $I_{sc}$ . This can happen because  $I_{sc}$  is related to the number of photons absorbed, affecting the electron excitation process from the dye used. An increase in light intensity is accompanied by an increase in the value of  $I_{sc}$  [30]. The  $P_{max}$  continues to increase with increasing light intensity and reaches its highest point at a light intensity of  $0.1 \text{ W/cm}^2$ . The highest maximum power is generated from DSSC with  $E_{a3}$  which is equal to  $0.719 \text{ W}$ .

The  $\eta_{max}$  value can be determined after obtaining each cell's  $V_{oc}$  and  $I_{sc}$  values. In this study, the value of energy conversion efficiency indicates the ability of DSSC to convert solar energy into electrical energy regardless of the fill factor value. DSSC using  $E_{a3}$  has a higher energy conversion efficiency value than DSSC using  $E_{b3}$  and  $E_{a1}$ . These results are directly proportional to the magnitude of the electrical conductivity value that is owned by the  $E_{a3}$  solvent. The greater the electrical conductivity value, the higher the movement or mobility of electrons, which is then interpreted as an electric current. Based on the Ohm's Law, voltage (V) is directly proportional to current (I) but inversely proportional to

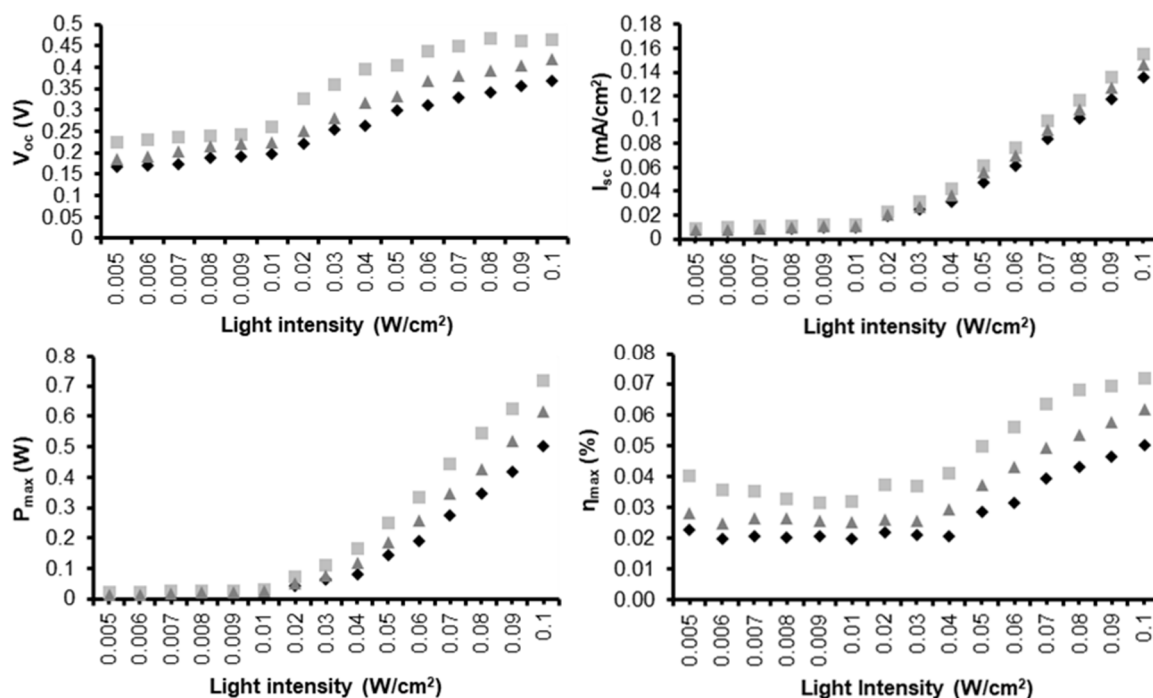


Fig 5. Performance parameters of DSSC with KI/I<sub>2</sub> electrolyte system with combination of acetonitrile:DES solvent (◆ E<sub>a3</sub>, ■ E<sub>b3</sub>, ▲ E<sub>a1</sub>)

**Table 3.** DSSCs performance

Parameter	Electrolyte used in DSSC		
	E <sub>a1</sub>	E <sub>a3</sub>	E <sub>b3</sub>
I <sub>sc</sub> (mA/cm <sup>2</sup> )	0.136	0.155	0.147
V <sub>oc</sub> (V)	0.369	0.465	0.419
P <sub>max</sub> (W)	0.502	0.719	0.616
η <sub>max</sub> (%)	0.050	0.072	0.062

resistance (R) [31-32]. The greater the electrical conductivity value, the resistance or resistance value will decrease, while the current and voltage values will increase.

In addition, based on the density and viscosity parameters in Table 2 shows that E<sub>a1</sub> has the lowest density and viscosity values, followed by E<sub>a3</sub> and E<sub>b3</sub>. The density and viscosity values of the electrolyte that are too small can result in leakage in the electrolyte system at DSSC. However, an increase in relatively high density and viscosity values in the E<sub>b3</sub> electrolyte causes a decrease in fluid velocity and electrolyte diffusion, which affects charge transport in electrolytes [12]. Therefore, the E<sub>a3</sub> electrolyte with the best density, viscosity, and conductivity parameters generates the highest energy conversion efficiency.

Table 3 shows that the DSSC using E<sub>a3</sub> performs better than the DSSC using E<sub>b3</sub> or E<sub>a1</sub> because the acetonitrile:DES<sub>a3</sub> based solvent has a higher electrical conductivity value. In addition, in the DSSC fabrication process, it was also observed that E<sub>a3</sub> and E<sub>b3</sub> had a longer shelf life in the cell state that had not been covered with glue when compared to E<sub>a1</sub>, which experienced faster evaporation. This can happen because E<sub>a3</sub> and E<sub>b3</sub> have a higher viscosity value than E<sub>a1</sub>.

## ■ CONCLUSION

The potential of deep eutectic solvents as solvents for KI/I<sub>2</sub> redox pairs has been successfully developed to substitute acetonitrile. The suitable volume composition of a mixture of DES and acetonitrile can produce electrolytes with higher viscosity and electrical conductivity than pure acetonitrile. The optimal composition was obtained at the volume ratio of acetonitrile: DES ChCl:EG with x<sub>ChCl</sub> 0.3 (η = 16.84 cP; κ = 9.94 mS/cm). These characteristics then cause the increased performance of DSSC in converting light

energy into electrical energy. Evaporation and solvent leakage in cells can be prevented, and the electron transport process in the electrolyte system can be improved. To determine the charge transport mechanism, further research will be needed on IPCE measurements and impedance data.

## ■ AUTHOR CONTRIBUTIONS

Adhitya: conceptualization, methodology, formal analysis, writing-original draft, Winda Rahmalia, Intan Syahbanu, Gusrizal, and Adhityawarman: supervision, resources, project administration, writing-review, editing, and validation. All authors have read and agreed to the published version of the manuscript.

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