



Ionic Liquid as Electrolyte in Photogalvanic Cell for Solar Energy Conversion and Storage

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Abstract: 1-Butyl-3-methylpyridinium bis(trifluoromethylsulfonyl)imide ($[c_4\text{3mpy}] [\text{NTf}_2]$) ionic liquid was used as electrolyte in photogalvanic cell. For photo-electrochemical conversion of solar energy to electrical energy, Rose Bengal, oxalic acid and ($[c_4\text{3mpy}] [\text{NTf}_2]$) was used as a novel system. The photopotential and photocurrent was 670.0 mV and 61.2 μA , respectively. The power of the cell at power point was 8.06 μW . The low values of the electrical output could be attributed to the fast mobility of the cation and aggregation motives. There are also several reasons related to the structure of the ionic liquid. The observed conversion efficiency was 0.077% and fill factor was 0.196. The storage capacity of the cell was 109.0 min. The effect of different factors affecting on electrical output of the cell was studied.

Keywords: Rose Bengal, Ionic Liquid, Conversion Efficiency, Storage Capacity, Photocurrent, Photopotential

1. Introduction

The coming deficiency of fossil fuel sources and environmental pollution force modern society towards the employment of renewable sources of energy. The most abundant energy resources on the earth surface are sunlight and water. The sunlight reaches on the earth in vast coverage that is enough to meet the global energy request. Solar energy is a cheap, clean, abundant and freely obtainable renewable non-classical source for power production. In photonic processes the photons of sunlight are used as the driving force in solar energy conversion. Photogalvanic cell (PG) technique provides a committed and unfamiliar technique for solar power generation and storage which contains dilute solution based dye sensitized solar power and storage means. There is no depletion of chemicals during charging and de-charging of these solar cells. These cells are completely dissimilar than other cells [1-3] such as galvanic or voltaic cells. Different PG cells containing diverse dyes, reductant and micelles have been studied [4-7] to enhance their electrical performance. At first, PG interested in using Pt electrode coated with Fe^{2+} as reducing agent. At the present

time, researchers used Pt, calomel electrodes and dyes such as toluidine blue [8, 9] methylene blue [10]. Also some literature used mixed dyes such as methylene blue and Azur-B [11], Aethioninee and azure-B [12], fluorescein [13], azure-A [14], Rose Bengal [15], etc., The used reductants as mannitol [16], oxalic acid [17], etc. and surfactants as sodium lauryl sulfate (NaLS) [18, 19], Tween-80 [5], etc.

Ionic liquids (ILs) which are molten salts with melting points at or below ambient temperature, they have some unique features such as good electrochemical and thermal stability, high ionic conductivity, non-volatility and non-flammability [20, 21]. Due to these properties, ILs have showed potential as safe electrolytes for being applied in high-energy-density lithium battery system [22-25]. The ILs, can be classified into two categories according to their electrochemical stability. The first category contains tetraalkylammonium, pyrrolidinium, piperidinium and quaternary phosphonium which have better electrochemical stability [26-28]. For example, ($[c_4\text{3mpy}] [\text{NTf}_2]$). The second category includes imidazolium and guanidinium ILs,

which have narrower electrochemical windows and higher cathodic limiting potentials compared with the first category.

The conductivity of an ionic liquid mainly depends on the mobility of its cation because in general the diffusion coefficients of ILs cations are higher than anions. Ionic liquids based on imidazolium and pyridinium cations have the highest ionic conductivity (~ 1 and 10^{-1} S/m, respectively) [29].

Our study interested in studying the electrochemical conversion of solar energy to electrical energy and storage using photogalvanic cell as well as studying the parameters affecting the electrical output.

2. Experimental

2.1. Materials

The structure of ([c₄3mpy] [NTf₂]) ionic liquid (IL) used in this study was shown in Figure 1. The ionic liquid (IL), Rose Bengal and Oxalic acid were purchased from Sigma-Aldrich. Sodium hydroxide was provided from Merck.

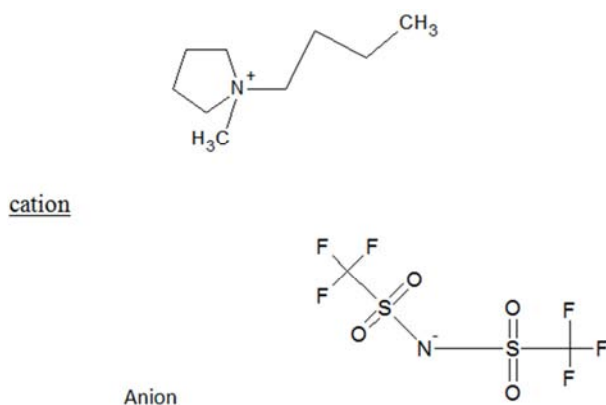


Figure 1. Structures of cation and anion of the ionic liquid used.



Figure 2. Set-up of photogalvanic cell.

2.2. Setup

PG cell consists of H-shape tube filled with known volume and concentration of the photosensitized dye, Oxalic acid, ionic liquid ([c₄3mpy] [NTf₂]) and sodium hydroxide. The total volume of the mixture was 25.0 ml. Different area ranged from 0.25-1.0 cm² of platinum electrode was dipped in the illuminated branch whereas the calomel electrode was immersed in the other dark branch. The terminals of the electrodes were connected to two Avometers and the whole cell was placed in the dark. The branch containing platinum electrode was illuminated using simulated visible light source. The lamp was placed in a silica tube surrounded with a water jacket to avoid any thermal effect on the cell. This means that the produced voltage and current is due to the solar light only. A water jacket is used to avoid thermal effect of the lamp, so that the produced voltage and current originate as a result of the photo-irradiation only. Different parameters such as; maximum potential (V_{max}), open-circuit potential (V_{oc}), maximum current (i_{max}) and short-circuit current (i_{sc}) could be measured after illumination the cell. From the analysis of the i - V curve, critical parameters of the cell as power at power point, p_{pp} , current at power point, i_{pp} , potential at power point, V_{pp} . The fill factor (FF) could be calculated using eq. (1), and the overall energy conversion efficiency (η) could be calculated using Eq. (2). Cell performance is studied in terms of half-life time ($t_{0.5}$).

$$\text{Fill factor (FF)} = V_{pp} \times i_{pp} / V_{oc} \times i_{sc} \quad (1)$$

$$\text{Conversion Efficiency \%} = [V_{pp} \times i_{pp} / I_s \times \text{Electrode area}] \times 100 \quad (2)$$

Where, I_s is the intensity of the incident light (mW/cm²).

3. Results and Discussion

3.1. Different Factors Affecting the Electrical Output

RB concentration

Table 1 shows the effect of photosensitizer on the electrical output of the cell. The results show that the optimum concentration of RB which produces the highest electrical output is 10.1×10^{-5} M. The increase in concentration of RB dye is accompanied with a decrease in electrical output of the cell. However, at low concentration of RB dye the Pt electrode receive a low number of electrons. Therefore, there is a low current in the external circuit. At higher concentration there will be less similarity in the absorption of light by the RB dye and hence the presence of different oxidized forms of RB dye could lead to the decrease in the output of the cell (RB^{•+} has normally three negative charges and the RB^{•+} is the mono-negative species so the decay of RB^{•+} is slower than RB^{•+}). The RB dye could be reduced by disproportion rather than loss of electron at Pt electrode. The dye molecules that exist and absorb photons near Pt have greater chance of electron donation to Pt electrode and a reversible dye electro-chemical process could be take place [30].

Table 1. Effect of Rose Bengal concentration on the electrical output.

[Rose bengal] × 10 ⁻⁵ M	Photopotential (mV)	Photocurrent (μA)	Power (μW)
10.75	453	39.2	17.75
10.47	560	42.3	23.68
10.35	632	56.5	35.70
10.10	670	61.3	41.00
9.84	601	52.6	31.61
9.52	514	46.5	23.90
9.00	475	41.3	19.61
7.80	429	38.5	16.51

Oxalic acid concentration

Table 2 shows the effect of reductant concentration on the photopotential and photocurrent on the cell. The suitable concentration which produces the highest electrical output was 1.9×10⁻³M. From this Table we can notice that the concentration below or higher the optimum concentration is accompanied by a decrease in the electrical output and hence the cell parameters. The fall of the power of the cell could be attributed to the presence of some electron scavenger species which arises from the hydrolysis of oxalic acid such as C₂O₄H⁻, C₂O₄²⁻. The photodegradation of oxalic acid which could occur due to the irradiation may result in the presence of some fragments. These fragments may decrease the produced power.

Table 2. Effect of reductant concentration on the electrical output.

[Oxalic acid] × 10 ⁻³ M	Photopotential (mV)	Photocurrent (μA)	Power (μW)
1.3	505	47.6	24.03
1.5	560	51.8	29.01
1.7	611	56.2	34.33
1.9	670	61.2	41.00
2.1	581	55.6	32.30
2.5	525	49.8	26.15

([c₄mpy] [NTf₂]) concentration

It was observed that electrical output of the cell increases with increasing the concentration of IL reaching a maximum value. Further increase in concentrations, photopotential, photocurrent and power decreased. The results are reported in Table 3.

Table 3. Effect of electrolyte concentration on the electrical output.

([c ₄ mpy] [NTf ₂]) × 10 ⁻³ M	Photopotential (mV)	Photocurrent (μA)	Power (μW)
1.0	520.0	49.50	25.74
2.0	561.0	52.70	29.56
3.0	670.0	61.20	41.00
4.8	535.0	51.50	27.55
5.5	490.0	45.40	22.24
30.0	363.8	30.50	11.09

In a description of classical electrolyte solutions, the mobile charge carrier is related to its diffusion coefficient *D* through the Nernst–Einstein equation:

$$A = \frac{z^2 e_0 F D}{k_B T} = \frac{z^2 N A e_0^2 D}{k_B T}$$

where, *z* represents the valence of the charge carrier, *e*₀ the elementary charge, *N*_A the Avogadro number, *k*_B the Boltzmann constant and *F* the Faraday constant.

Franca et al., 2009 found that,

- 1 The diffusion of cation is greater than the diffusion of anion indicates a higher degree of intermolecular grouping of the anions, leading to lower mobility.
- 2 Favored interaction of the anion with the five-membered ring of the cation of the ionic liquid.
- 3 Homonuclear, cation-anion heteronuclear aggregation motives.

From these findings we can conclude that as the concentration of IL increase to a certain limit, there will be a low mobility, and increase the viscosity which decrease the mobility of the ions and so decrease the electrical output.

Change in pH

Table 4 shows the effect of change in pH on the cell. The pH was varied according to the used system. Our study was done by adjusting the pH ranges from 12.98-13.89. The results show that the efficiency of the photogalvanic depends on the pH of the system. The electrical output of the photogalvanic cell was improved at pH 13.4. It can be observed from the Table 4 that below or higher the pH 13.4 there is decrease in the electrical output of the cell. The reason could be attributed to the protonation- deprotonating of RB dye. At very high pH more than 13.4 there is excess of OH⁻ available in the system which could acts as electron scavenger and hence decrease the number of electrons reaches the Pt electrode. The increase in hydroxyl radical could also attack the benzene ring of RB and destroy the structure of RB dye.

Table 4. Effect of different pH on the electrical output.

pH	Photopotential (mV)	Photocurrent (μA)	Power (μW)
13.89	372.6	30.0	11.170
13.74	402.0	35.2	14.150
13.62	561.0	47.5	26.64
13.52	623.0	52.4	32.64
13.40	670.0	61.2	41.00
13.26	556.0	45.6	25.35
13.11	442.0	33.5	14.81
12.98	418.0	29.5	12.33

Table 5. The performance of photogalvanic cell.

Parameters	Observed results
Open circuit potential (<i>V</i> _{oc})	850 mV
Short circuit current (<i>i</i> _{sc})	75.2 μA
Maximum photocurrent (<i>i</i> _{max})	80 μA
Time of illumination	32 min
Storage capacity (<i>t</i> _{1/2})	109 min
Conversion efficiency	0.077%
Fill factor	0.196
Current at power point (<i>i</i> _{pp})	29.7 μA
Potential at power point (<i>V</i> _{pp})	271.7 mV
Power at power point (PP)	8.06 μW
Maximum power	41.00 μW

Diffusion length

Figure 3 shows the effect of diffusion length on the

photoinduced potential, photoinduced current and power. The diffusion length was studied using cells with different distances between the two electrodes; 2, 3, 6 and 8 cm. It was observed that there is increase in the photopotential, photocurrent and consequently the power of the cell as the diffusion length increase. We can notice that the photopotential increases from 498 to 670 mV as the diffusion length increases from 2 to 8 cm. i.e. The photopotential increases by 182 mV, the increase in photocurrent is about 15.6 μA and the increase in power is about 18.3 μW . The increase in photocurrent and power with the diffusion length is not significantly high as the photopotential. This confirms that the rate determining step is the recycling reaction of the semi- or leuco dye and the oxidized compound of the reducing agent in the dark chamber [31].

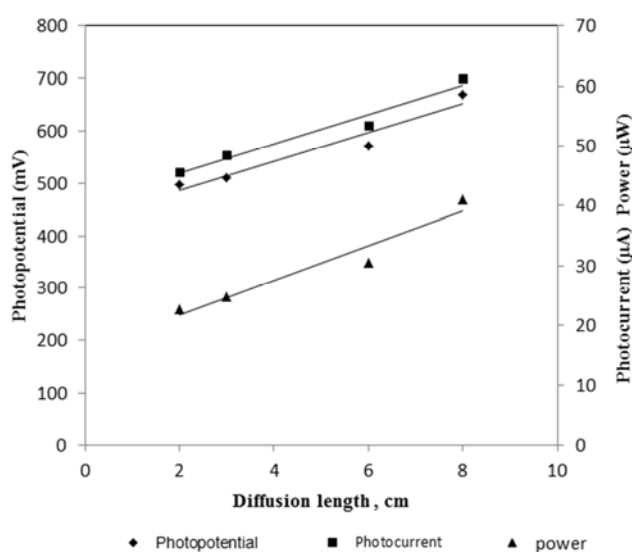


Figure 3. Effect of variation of diffusion length of the cell on the electrical output.

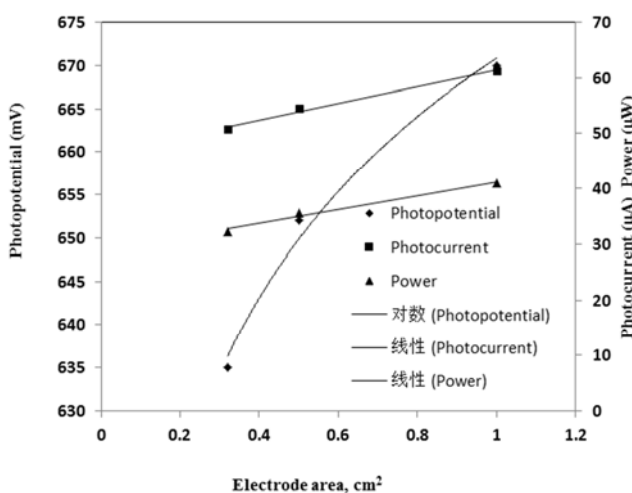


Figure 4. Effect of variation of electrode area of the cell.

Electrode area

Figure 4 shows the effect of variation of Pt electrode area on the electrical output of the photogalvanic cell. This study was carried out using different electrode area ranges from

0.3-1 cm^2 . The results confirmed that with the increase in the electrode area, the electrical output of the cell was increased. The photocurrent and power increase linearly as the electrode area increase. The photopotential increases logarithmically. This indicates that the photopotential tend to reach equilibrium value as electrode area increase. On the other hand, the increment in photocurrent and power depends on the electrode area. The dye molecules are transported towards the charged illuminated electrode by diffusion and a static layer is gradually developed near the electrode surface. The formation of charge transfer complex between the dye and IL could be formed on the electrode surface which could be a charge transfer type with increased donor capacity [32]

3.2. Data Analysis

The Current-Voltage (i-V) characteristics of the photogalvanic cell containing Rose Bengal-Oxalic acid - ([c43mpy] [NTf₂]) system is represented in Figure 5. From i-V (current-potential) the following parameters can be obtained:

- i_{sc} (Short-circuit current): the measured cell current at zero applied potential.
- V_{oc} (Open-circuit voltage): the potential measured of the cell when the current is zero.
- M_{pp} (Maximum power point): the point where the maximum power is generated.
- FF (Fill factor): the ratio of the maximum power to the short and open circuit values.

The value of fill factor was 0.196 and the power at power point of cell (pp) was 8.06 μW .

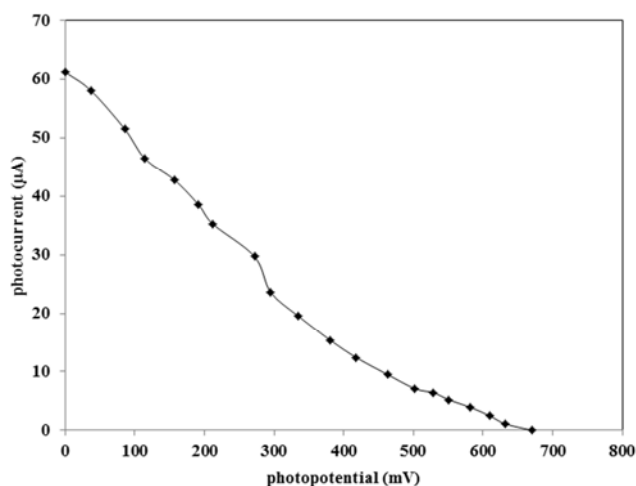


Figure 5. Current-voltage curve of the cell.

The storage capacity of the cell was determined in term of $t_{1/2}$, i.e., the time essential to fall the electrical output (power) to its half value at power point in the absence of illumination. It was found that the cell can be used in dark for 109.0 min after illumination for 32.0 min. Figure 6 shows the storage capacity of the photogalvanic cell.

The conversion efficiency of the system containing Rose Bengal as photosensitizer is calculated using the electrical output at power point and the power of incident radiation.

The conversion efficiency of the photogalvanic cell is determined as 0.077% using the following formula:

Conversion efficiency = $[(V_{pp} \times i_{pp}) / (I_s \times \text{Electrode area cm}^2)] \times 100\%$

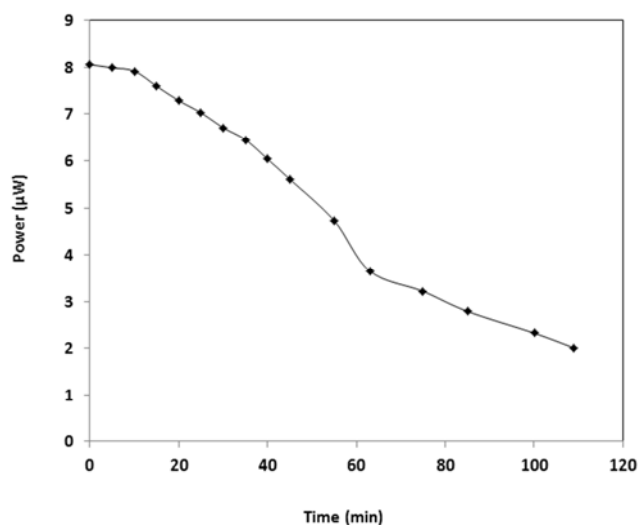


Figure 6. Storage capacity of the photogalvanic cell.

The overall performance of the Rose Bengal–Oxalic acid ([c₄mpy] [NTf₂]) system is summarized in Table 3.

Comparing the obtained data with recent published work [18, 33] shows higher values of the cell parameters of Rose Bengal–Oxalic acid–Tween 80 system; CE 0.258%, I_{sc} is 119.5 μA; P_{pp} is 79.58 μW and t_{0.5} was 230 min. Whereas, Rose Bengal–Oxalic acid–CTAB system; CE 0.067%, I_{sc} 75.0 μA, P_{pp} 41.3 μW and t_{0.5} was 175.0 min.

In the present work, we observe a fall in storage capacity to half its value (109 min upon using the ionic liquid ([c₄mpy] [NTf₂]) while it was recorded 230 min using CTAB [15]. This could be attributed to the fact that covalent ionic liquid cannot effectively allow the passage of charge. The formal charges or electronegativity effects within the compounds create dipoles which allow some charge (though very little) to be stabilized and transported. Therefore, they are poor conductors of electricity but have good storage efficiency.

From these results we can conclude that, the order of electricity production is:

Non-ionic surfactant > cationic > covalent ionic liquid

The surfactants increase the cell operation through (i) overcoming the back electron transfer, (ii) increase electron transfer to photosensitizer and (iii) improving the solubility [34] and stability of the sensitizer molecules [35]. It also observed that the photogalvanic cell with ionic liquid system has conversion efficiency and storage capacity, 0.077% and 109.0 min, respectively. These values are relatively higher in comparison to previously published system [8] containing malachite green–arabinose–NaLS (0.059% and 32.0 min). It is also observed that previously reported photogalvanic cells [5, 35] with surfactants have relatively lower storage capacity compared with our system; EDTA–thionine–azur B (59.0 min), EDTA–safranin–tween-80 (20.0 min), EDTA–

safranin–O–DSS (80.0 min), methylene blue–xylose–NaLS (55.0 min) systems.

4. Conclusions

The photo-electro conversion of solar energy to electrical energy and storage by photogalvanic cell considers a very simple technique to produce power. Data analysis of our study reveals that the conversion efficiency was 0.077% and storage capacity was 109.0 min using 1-Butyl-3-methylpyrrolidiniumbis (trifluoromethylsulfonyl) imide as electrolyte in this system. Different factors affecting the electrical output of the cell as; photosensitizer concentration, change in pH, reductant concentration, diffusion length and electrode area were studied. The study showed that the order of electricity production is: non-ionic surfactant > cationic > covalent ionic liquid. As is no external circuit, the cell will keep light energy stored.

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