



Polyacrylonitrile gel polymer electrolyte based dye sensitized solar cells for a prototype solar panel

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ABSTRACT

Polyacrylonitrile (PAN) based gel polymer electrolytes (GPE) were prepared using lithium iodide (LiI), 1-butyl-3-methylimidazolium iodide (BMII) and tetrapropyl ammonium iodide (TPAI). The LiI mass fraction in the electrolyte was varied while keeping the masses of other components constant in order to enhance the solar cell performance. The addition of 4.61 wt.% LiI in the GPE increased the electrolyte room temperature ionic conductivity from (2.32 ± 0.02) to (3.91 ± 0.04) mS cm^{-1} . The increase in conductivity with the addition of LiI salts was attributed to the increase in diffusion coefficient, mobility and number density of charge carriers as determined from Nyquist plot fitting. The incorporation of LiI salts in PAN-based GPE has enhanced the efficiency of the DSSC as expected. The best cell performance was obtained with an electrolyte containing 4.61 wt.% LiI sandwiched between a single mesoporous layer of TiO_2 soaked in N3 dye sensitizer and a platinum counter electrode, which showed a power conversion efficiency (PCE) of $(5.4 \pm 0.1)\%$ with a short circuit current density (J_{sc}) of (21.0 ± 1.1) mA cm^{-2} , an open circuit voltage (V_{oc}) of (0.48 ± 0.02) V and a fill factor (FF) of $(53.4 \pm 0.9)\%$. The DSSCs with 4.61 wt.% of LiI have been used to fabricate prototype solar panels for operating small devices. The panels were assembled using a number of cells, each having an area of $2 \text{ cm} \times 2 \text{ cm}$, connected in series and parallel. The panel, consisting of a set of eight cells in series which was connected in parallel with another set of eight cells in series, produces an average power conversion efficiency of $(3.7 \pm 0.2)\%$ with a maximum output power of (17.1 ± 0.9) mW.

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1. Introduction

At present, the world's power consumption is derived from fossil fuels, nuclear power, hydropower, solar power, wind power, etc. However, the solar energy is a more or less limitless freely available and environmentally friendly [1]. Basically, three methods are used to convert solar energy into usable forms of energy. Thermal power plants utilize heat energy in solar radiation, photo-electrochemical solar cells convert sunlight into electricity or stable chemical combinations that can be used as fuels, and solar cells convert sunlight into electricity [2]. The dye sensitized solar cells (DSSCs) that convert solar energy directly to electricity in an

environmentally friendly manner are very important devices to fulfill future energy requirements since electricity can be converted to other usable forms of energy very easily.

In general, silicon-based solar cells are conservative solar devices which can exhibit efficiency up to 25% in laboratories [3]. However, high production cost and use of toxic chemicals in the manufacturing process of these solar cells have compelled the search for low cost and 'greener' solar cells. The introduction and advancement of dye-sensitized solar cells (DSSCs) or "Grätzel cells" have attracted much attention due to their low cost, environmentally friendly operation, and easy fabrication. Progress in cell construction and efficiency has also been reported [4–6].

In DSSCs, the electrolyte plays an important role as a medium for charge transport. The electrolyte must contain a redox couple to transfer electrons from the counter electrode to the ionized dye in the photo-anode [4–8]. For an efficient charge transfer, the redox

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level should be compatible with the Fermi level of the TiO_2 film. In this work, the I^-/I_3^- couple was selected for use in the electrolyte due to the many advantages reported in the literature [4–6,9]. In addition to providing an appropriate redox level, the conductivity of the electrolyte, the presence of small cations that improves the current density and the presence of bulky cations that improves the cell potential are important to achieve optimum cell performances [10–12].

Electrolytes most commonly used in DSSCs are liquids, but nowadays gel polymer electrolytes (GPEs) are being explored [6,10–12]. The use of GPEs has minimized the problems of leakage, evaporation, and corrosion which are the main problems that arise in using liquid electrolytes. In addition, GPEs provide better contact with electrodes and higher ionic conductivity than those of solid polymer electrolytes and offer long-term chemical and physical stability for DSSCs. Many types of host polymers have been used for gel electrolytes in DSSC applications such as poly(ethylene oxide) (PEO) [13], poly(acrylonitrile) (PAN) [14], poly(vinylidene fluoride) (PVDF) [15], poly(vinylidene fluoride-hexafluoropropylene) (PVDF:HFP) [16], poly(ethylene glycol) (PEG) [17], poly(vinyl pyrrolidone) (PVP) [18], poly(methyl methacrylate) (PMMA) [19], poly(ethylene glycol) methyl ether methacrylate (PEGMA) [20–23] and poly(1-vinylpyrrolidone-co-vinylacetate) P(VP-co-VAc) copolymer [24] with reasonable light to electricity conversion efficiencies. In this work, PAN was chosen as polymer host for the preparation of gel electrolytes due to its relatively high dielectric constant of 5 [25] that assists to dissociate the doping salt into free ions which can increase the conductivity of the electrolyte [10–12]. The nitrile groups in the PAN polymer chain are advantageous because of their polar character which enables them to hold more free cations. In addition, PAN is compatible with polar substances [26] and can dissolve easily and become gel by trapping the polar solvent in their polymer matrix. These unique properties show PAN has a good potential as a host polymer in polymer electrolyte.

The PAN based single-salt gel electrolytes with quaternary ammonium iodides, R_4NI (R = ethyl, propyl, butyl, pentyl, hexyl), perform well in DSSCs with efficiencies of about 3 to 4.3% [27] as the bulky quaternary ammonium iodides can dissociate easily. The sluggish movement of the bulky cations results in lower cationic conductivity, thus the iodide ions contribute to a larger part of the conductivity. Small cations such as Li^+ , K^+ and Na^+ are also commonly used in electrolytes intended for DSSCs. KI-based gel electrolytes have been used widely since they give fairly good DSSC performance with efficiencies of the order of 5% [28,29]. Liquid and gel electrolytes with lithium iodide have also shown reasonably good efficiencies in DSSCs [30,31]. Even in the well-known first paper on DSSCs by O'Regan and Gratzel, it was reported that the incident photon-to-current conversion efficiency in the DSSC increased from 68% to 84% when tetrapropylammonium iodide (TPAI) in the electrolyte was replaced by LiI [5,32]. Thus the presence of small cations in the electrolytes for DSSC is important and is the main objective of this work. The other two salts have large cations i.e. TPA^+ and BMI^+ .

The ionic conductivity in gel polymer electrolytes can be improved by adding an ionic liquid. Hence, the efficiency of quasi-solid DSSCs can be enhanced by incorporating ionic liquids [33,34]. Ionic liquids are nonvolatile, nonflammable, thermally stable and have a high conductivity and a wide potential window [33,34]. Park et al. [35] have investigated the influence of Li^+ and 1,2-dimethyl-3-hexyl imidazolium ions in the electrolyte on the performance of DSSCs and concluded that Li^+ ions lower the open circuit voltage (V_{oc}) but increase the short circuit current density (J_{sc}). Some research papers report that the V_{oc} of DSSCs can be improved by incorporating bulky cations such as imidazolium ions in the electrolyte [33,34]. Since it has been shown that the interfacial electron injection rate in DSSCs increases with increasing cation

charge density and conversely the V_{oc} increases when the cation charge density decreases [15], thus the use of mixed cation systems is important to enhance the performance of DSSCs.

An efficiency enhancement in quasi-solid-state dye sensitized solar cells can be attained by incorporating a binary iodide salt into the electrolyte since different cations in the electrolyte play different roles. For instance, gel electrolytes with a mixture of two iodide salts consisting of large cations and small cations have been used to improve the performance of DSSCs by some authors [10–12,15,19,27]. In order to utilize all the advantages of different cationic species, triple iodides were used in the present work to prepare gel polymer electrolytes for DSSCs. After a thorough literature survey, TPAI, BMII and LiI were selected as suitable iodide salts to prepare gel polymer electrolytes for DSSCs expecting good efficiency owing to different functions of the cations in the electrolyte.

In an attempt to test the feasibility of using the GPE based DSSCs in a practical application, a prototype solar panel has been designed and fabricated. The construction procedure and the performance of this solar panel are also discussed in this paper.

2. Experimental

2.1. Gel Polymer Electrolyte Preparation

The chemicals used i.e. polyacrylonitrile (PAN), ethylene carbonate (EC), propylene carbonate (PC), lithium iodide (LiI), tetrapropylammonium iodide (TPAI), 1-butyl-3-methylimidazolium iodide (BMII) and iodine (I_2) were purchased from Sigma Aldrich. PAN, LiI and TPAI were vacuum dried prior to use. The gel polymer electrolytes were prepared by keeping the amounts of PAN (0.3 g), EC (1.245 g), PC (1.155 g), TPAI (0.1329 g), and BMII (0.0376 g) fixed and varying the amounts of LiI and I_2 as given in Table 1. For the preparation of GPEs, corresponding amounts of LiI, BMII and TPAI were first dissolved in appropriate amounts of EC and PC contained in a glass container. Then, the required amount of PAN was added to the salt solution and it was stirred at 383 K until homogeneous slurry was obtained. The slurry was then allowed to cool down to 313 K and iodine (I_2) chips were added to the slurry while maintaining the continuous stirring for few more minutes. The amount of I_2 added was 10 mole% of the total iodide salt added in the electrolyte. The resulting gel electrolytes were characterized and used for solar cell fabrication.

Table 1 Compositions of seven different gel electrolytes prepared with fixed amounts of PAN (0.3 g), EC (1.245 g), PC (1.155 g), TPAI (0.1329 g), and BMII (0.0376 g). This is the same composition as sample S1 but without LiI.

2.2. Differential Scanning Calorimetry (DSC)

DSC was carried out to determine the glass transition temperature (T_g) of the prepared gel polymer electrolyte. The electrolyte of weight ~ 5 mg was sealed in aluminum pans and the

Table 1

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Sample Name	LiI (wt.%)	LiI (g)	I_2 (g)
S1	0	0.0000	0.015
S2	1.32	0.0378	0.021
S3	2.61	0.0750	0.030
S4	3.29	0.0945	0.033
S5	3.95	0.1134	0.036
S6	4.61	0.1323	0.039
S7	5.28	0.1515	0.042

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