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Fabricated for Dye-sensitized Solar Cells using polyethylene oxide - blended polymers based gel Electrolyte

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Abstract: Dye sensitized solar cell with a gel electrolyte has been fabricated using PEO- blended polymers. The different weight percentage (weight %) of blended polymers (2%, 4%, 6%,8%) has been synthesized by gel casting techniques. To improve the performance of the device blended polymers was added with different weight percentage of metal nanoparticles of carbon (5%,10%, 15%,20%,25%). The gel electrolyte with blended polymers was characterized by Differential Scanning Calorimetry (DSC). The PEO blended with 6% wt% polymer electrolyte has obtained the ionic conductivity value of 2.6×10^{-6} S/ cm^{-1} at room temperature, which was found to be maximum value compared with other wt%. The obtained gel electrolytes performance was analyzed by fabricating the device. DSSC assembled with the optimized 6% of blended polymers got an efficiency of 2.1 % and blended polymers with metallic nanoparticles improved the power conversion efficiency to 3.1% with the optimized weight of 20%.

Keywords: composite poly(ethylene oxide) PEO- blended polymers \ PEO- blended polymers with carbon filler electrolyte, Ionic conductivity, Efficiency, dye-sensitized solar cell.

1. INTRODUCTION

Dye-sensitized solar cells (DSSCs) brings an striking choice to conservative photovoltaic devices for conversion of solar energy to electrical energy at low cost and low environmental impact during the invention. Solar cell developed and classified in to four type **1. First generation solar cells** are make using crystalinity silicon wafer. It consist of required large area in single layer PN junction. It has a band gap of 11 eV. **2. Second generation solar cells** based thin film and reduce the mass of materials. **3.Thrid generation solar cells** photo electrochemical solar cell and dye sensitized solar cell the function of light absorption from charge carrier transport. Dye

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sensitized absorb the incident sunlight and exploits the light energy to stimulate vectorial electron transfer reaction. **4. Fourth generation solar cells** are polymer based solar cell, this technology relatively new. It is lightweight and flexible and customizable on the molecular level, lower potential for negative environmental impact. These cells with liquid electrolyte contain achieve in excess of 11% of solar alteration efficiencies below irradiation of AM 1.5 (1). A dye-sensitized solar cell consists of a dye-sensitized nano crystalline titanium dioxide film as working electrode, an electrolyte including a proper redox-couple and a platinum counter electrode. Energy transfer in a DSSC take places when the light is absorbed by a sensitizer and photo excitation of the dye outcome in the injection of electrons into the conduction band of TiO2 (2)(3). Consequently the dye is regenerate by the electron donor present in the electrolyte. The electrons in the conduction band are collected, flow through the external circuit to enter at the counter electrode, An outline of the operation of DSC is depicted in Fig. 1. The dye molecule is excited from ground state (D0) to excited state (D*) by sunlight, rapidly injects an electron into the conduction band (CB) of semiconductor porous film. The electrons in CB arrive at the counter electrode through the external circuit, and reduce the I^{3-} ions to I^{-} ions with the aid of Pt. Then, I^{-} ions donate electrons to dye cations to complete the cycle of dye regeneration.

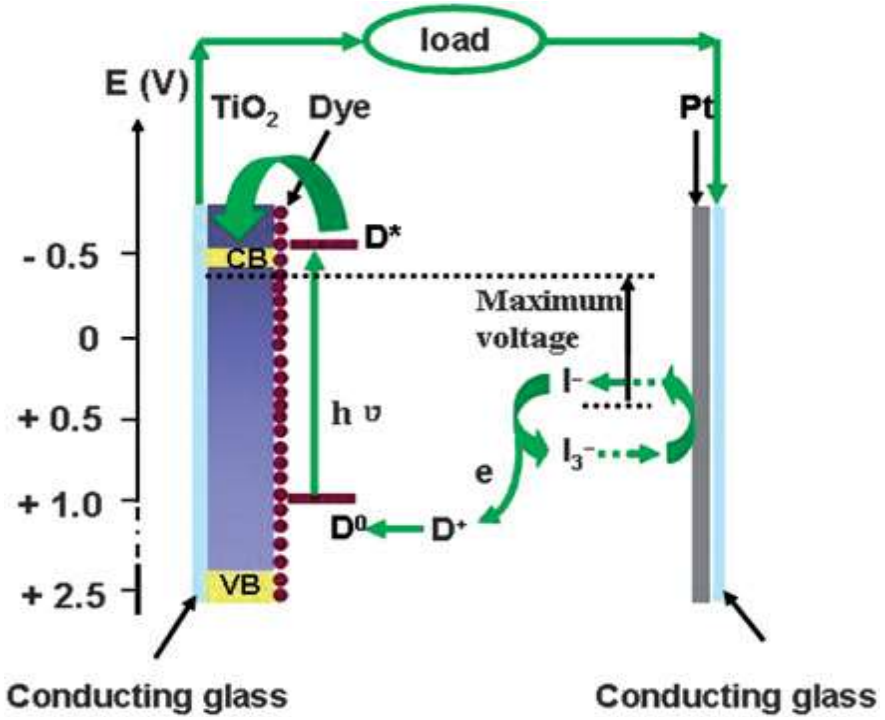

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Fig. 1

where they reason for the reverse effect of the redox intermediary. Thus, the photoelectrochemical cell is regenerative and the procedure ahead to direct alteration of sunlight into electrical energy(4) (5). The use of liquid electrolytes reason troubles such as outflow and volatilization of the fluid, desorption of the closed dyes, and the rust of the counter electrode and therefore restrictions the enduring act and commercial improvement of DSSC(6). numerous efforts have been made to find a appropriate substitute and polymer based electrolytes have revealed promising results. Poly(ethylene oxide) (PEO) has been intensively studied as solid polymer electrolyte because it is both chemically stable and polar, which means that it can readily dissolve salts and corrode the device. However, its ionic conductivity is not satisfactory for DSSCs, primarily because of its high crystallinity. In my research work carry out the energy conversion efficiency of DSSC employing PEO-blended polymer as polymer electrolyte was 2.1% . Many attempts have been made to improve ionic conductivity of PEO polymer electrolyte by incorporating nanoparticle fillers was introduced into a high molecular weight PEO to reduce crystallinity of the polymer. The energy conversion efficiency of DSSC using this polymer was 3.1% . In this work, the influence of carbon filler and blended polymer as additives in the composite PEO- blended polymers electrolyte on conductivity and photoelectric performance of quasi-solid-state dye-sensitized solar cells was studied. The optimized ratio of blended polymers with carbon filler additive is suggested based on the high conductivity of the composite polymer gel electrolyte as well as the overall energy conversion efficiency and stability of DSSCs.

2. MATERIALS AND METHODS

Fluorine doped tin oxide coated (FTO) was purchased from Solaronix, Switzerland. TiO_2 powder (P25) was a sample from Degussa, Germany. The absolute ethanol, acetone, acetonitrile,. N719 ruthenium(III) Dye was purchased from Solaronix S.A. (Switzerland) , iodine crystal, lithium iodide, and chloroplatinic acid hydrate ($H_2PtC_{l6}·H_2O$), blended polymer(PPG, PEG) were purchased from Sigma Aldrich. Poly ethylene oxide (MW=1000), Poly(ethylene glycol) (MW=8000) were obtained from Alfa Aesar India Pvt. Ltd., India (9).

2.1 Preparation of Polymer Electrolytes

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Composite polymer gel electrolyte consisting of PEO with a ratio of 2%, 4%, 6%, and 8% by weight was prepared in blended polymers and iodine iodide(I_2), lithium iodide as a solvent in acetonitrile added one by one in step wisely. Next another one electrolyte prepared in filler was added PEO-blended polymers with carbon additive (5%,10%,15%,20% and 25%) and then stirred for 12 hours.

2.2 Preparation of Electrodes

The nanocrystalline TiO_2 (Degussa P25) film was prepared by doctor-blade technique. The TiO_2 paste was prepared by mixing of commercial P25-TiO_2 powder with polyethylene oxide (PEO), 0.15g polyethylene glycone (PEG), distilled water finally and grind well using a mortar and pestle to obtain a colloidal paste. TiO_2 colloidal paste was coated on cleaned fluorine-doped tin oxide (FTO) glass substrate. After drying in air at room temperature, the TiO_2 film electrode was sintered at 500°C for 30 min. The sensitizing dye was adsorbed on the TiO_2 surface by immersing into 0.5 mM solution of N719 dye for room temperature at 24 hours. Afterwards, the TiO_2 film electrode was rinsed with ethanol and dried in the air. A platinum counter electrode was prepared by spin coating 0.1 mM solution of H_2PtC_{l6} in isopropanol on FTO glass substrate and then it was sintered at 420°C for 30 min.

2.3 Fabrication of DSSC

A dye-sensitized solar cell was fabricated by sandwiching the polymer gel electrolyte in between a dye-adsorbed TiO_2 film electrode and a platinum counter electrode. The two pieces of FTO glass were clamped on the opposite sides by two metal clips. The DSSC was then measured its open circuit voltage (VOC), short circuited current density (ISC) and fill factor (FF). The working performance of the DSSCs was tested by recording the current density voltage (J-V) curves with a Keithley 2400 Source Meter (Oriel) under illumination of simulated AM 1.5 solar light coming from a solar simulator (Oriel-91193 equipped with a 1000 W Xe lamp and an AM 1.5 filter). The electrochemical impedance spectroscopy(EIS) was carried out on a symmetric thin-layer cell composed by sandwiching the composite polymer electrolyte between two platinized FTO electrode. The measurement was performed by using Autolab Potentiostat Chiang Mai J. Sci. 2011; 38(2) 225 PGSTAT 302 with FRA module at the zero bias with the frequency range of 0.05 Hz to 1 MHz. The DSSCs were illuminated by simulated sunlight from

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the Xenon arc lamp under AM 1.5 (100 mW cm-2) and measured by a Keithley 236 source-measure unit. The active area of the cell was 1 cm^2.

3. RESULTS AND DISCUSSION

3.1 Ionic Conductivity study

Ionic conductivity of the blended Polymer based gel electrolyte were measured using electrochemical impedance spectroscopy (EIS) under AC amplitude of 10 mV and the frequency range from 100 to 10^5 Hz . The gel electrolyte was sandwiched between two FTO glasses separated as the contact electrode was measure the Ionic conductivity of these gel electrolytes were calculated by using the following formula:

$$\sigma = L/R^* A \qquad \text{_____ (1)}$$

where ''σ'' is the ionic conductivity, R is the bulk resistance, L is the thickness of the sample and A is the area of the given sample. The ionic conductivity of the gel electrolytes was measured and calculated in **table 1**. the variation of ionic conductivity as a function of blended polymers content in the gel electrolyte as shown in Fig.2. The ionic conductivity of gel electrolyte increases with increase in the blended polymers content and maximum conductivity of 1.7 $x10^{-3}$ S/cm was achieved for 6 wt% of blended polymers. The conductivity was found to decrease when blended polymers content was increased more than 6wt% in the gel electrolyte.

Table 1. Ionic conductivity Measurement: blended polymers

PEO-Blended polymer %	R Ω	L Mm	A Cm	Conductivity σ (S/cm)
2	65.01	0.0133	0.16	1.2 $x10^{-3}$
4	56.31	0.0133	0.16	1.4 $x10^{-3}$
6	47.92	0.0133	0.16	1.7 $x10^{-3}$
8	53.12	0.0133	0.16	1.5 $x10^{-3}$

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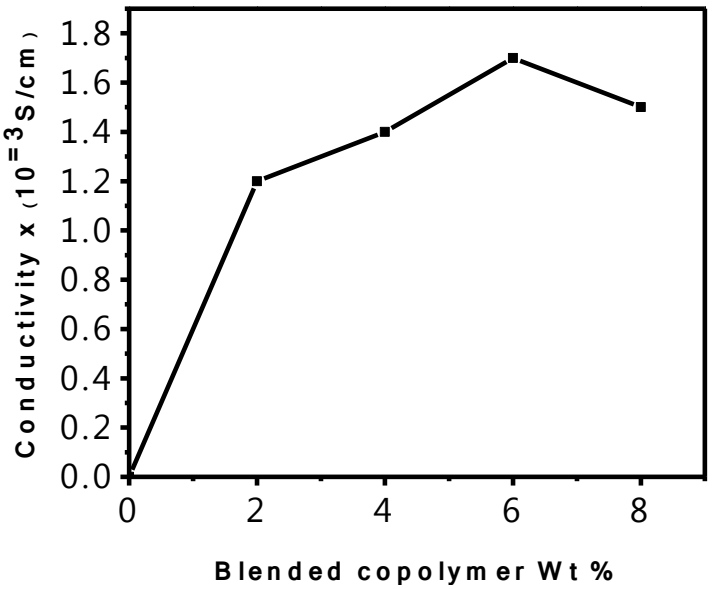

Fig. 2. Ionic conductivity of gel electrolyte as a function of different wt% of blended polymers.

3.2 Differential scanning calorimetric studies were carried out using neat PEO, PEO in acetonitrile, blended polymer , blended polymer in acetonitrile and PEO with blended polymer in acetonitrile along with 6 wt% are presented in Fig. 3. Pure PEO and PEO in acetonitrile have showed the transition melting temperature (Tm) at around 64.4 °C and 61.9°C respectively. On gelation blended polymer with the Tm observed for PEO is absent and a weak transition at around 40.7 °C , blended polymer in acetonitrile 37.7°C was observed. This is attributed to the loss of crystallinity of PEO (7). In addition to that the percentage of the crystallinity of neat PEO and PEO with 6% blended polymer can be calculated by taking the ratio of change in enthalpy (DHm) of sample to that of 100% crystalline PEO (~193 Jg^{-1}) (8)(9). The crystallinity of neat PEO was found to be 68.7%, while the crystallinity was reduced up to 0.53% PEO in acetonitrile and 0.74% for PEO with 6% blended polymer respectively. This indicates the amorphous nature of this material due to gelation that resulted in the increase in ionic conductivity of the gel electrolyte.

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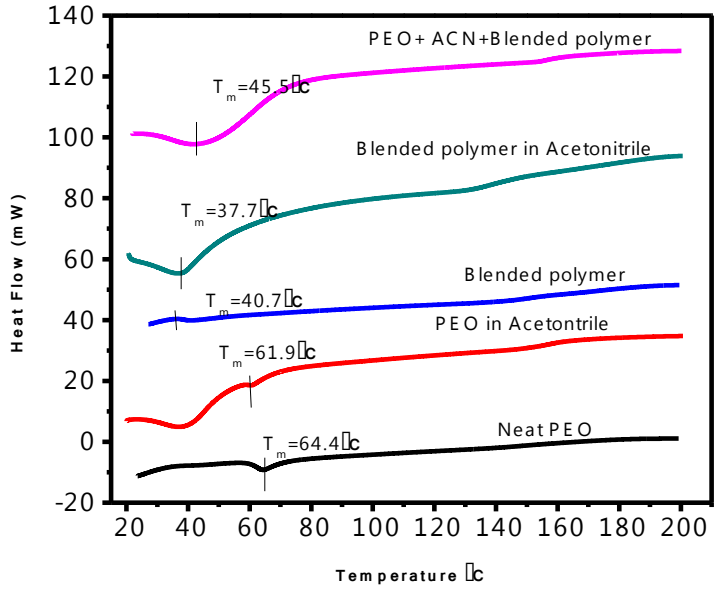

Fig. 3. Differential scanning calorimetrythermograms

The charge transport mechanisms of EIS measurements were carried for G-DSSCs under constant illumination of 100 mW/cm^2 with the optical filter of AM 1.5 G under open circuit condition **Fig. 4(a).** It can be seen from the figure that it comprises two semicircles are provided in the figure as an inset in accordance with the reported literature(10) (13) . Rs, Rct1, and Rct2 refer to the total series resistance, charge transfer resistance at platinum CE and electrolyte interface, recombination resistance at TiO2/Dye and electrolyte interfaces, respectively. The corresponding data are extracted from the EIS spectra and listed in the **Table 2.** Rct1 corresponds to the charge transport resistance at Pt and PEO-blended polymer electrolyte interface where the reduction of tri iodide species takes place. It can be seen from the Table. 2 that PEO- Blended Polymer based G-DSSC shows the lowest Rct1 (34.77) among these G-DSSCs, this resulting in the best I– production at the counter electrode. Hence, Jsc value was higher for PEO- Blended Polymer based G-DSSC. This low Rct_1 also manifests in the FF values (Table 3). Rct_2 corresponds to charge recombination resistance at TiO2/Dye and electrolyte interface. For filler carbon based G-DSSC the Rct_2 of 35.54 was observed. To the low charge transport resistance in TiO_2 and in TiO_2/Dye/electrolyte interface. Hence, higher efficiency was observed (14,15). Nano carbon material was used as a filler for polymer based gel electrolyte and the corresponding G-DSSC was fabricated. The J-V characteristics for the DSSC based on carbon fillered gel

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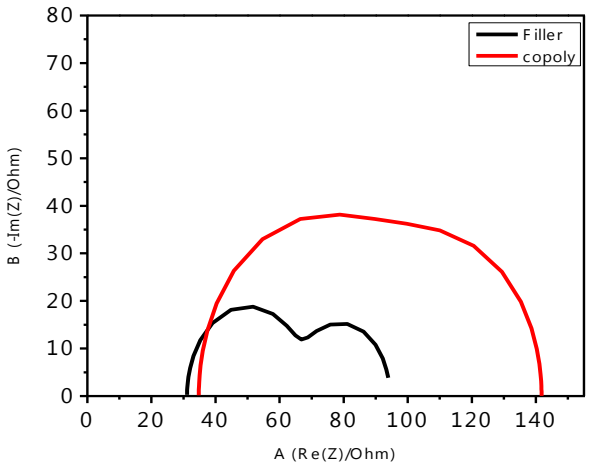
electrolyte along with the blended polymer gel electrolyte is presented in Fig. 5(a),(b) and the corresponding data are listed in the **Table 3.** The AFM 3D image (Fig. 2a) of gel-electrolyte film with carbon shows that the filler particles have the strong influence on the surface modification of the polymers and reduces the crystallinity (11)(12) and forms mechanically stable network that creates voids and free spaces for the effective percolation of I^- and I^{3-} ions, rendering good charge transport characteristics.



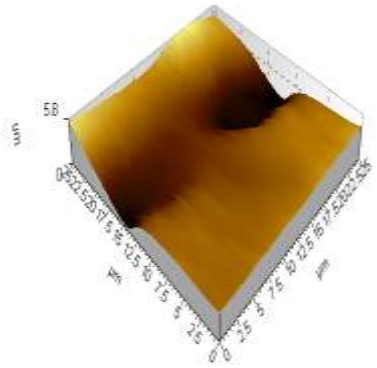


Fig. (a) Nyquist impedance Zpit curve Fig. (b) AFM

Fig. 4 (a). Electrochemical impedance spectra of G-DSSCs and the corresponding equivalent circuit (inset) under constant illumination of 100 mW/cm2 with the optical filter of AM 1.5 G under open circuit condition (b) Filler gel electrolyte AFM

Table.2 EIS analysis of the gel state DSSC under constant illumination of 100 mW/cm^2 with the optical filter of AM 1.5 G under open circuit condition

Sample	Conductivity σ (S/cm)	Balance resistance Rs Ω	Rct_1 Ω	Rct_2 Ω	Diffusion coefficient ω	Efficiencies η (%)
PEO-Blended Polymer	1.7 x10^{-3}	4.68	34.77	58.99	48.02	2.1
PEO-Blended	2.6 x10^{-3}	3.13	31.11	35.54	27.74	3.1

Polymer with filler						

3.3 Performance of Dye-sensitized Solar Cells

In order to investigate the photovoltaic performance of assembled DSSCs using different weight % of blended polymers electrolytes, the J-V characterization was done using solar simulator with the stimulated radiation of AM 1.5G. J-V (a) PEO- blended polymers and (b) \ PEO- blended polymers with carbon filler electrolytes is shown in Fig.3. From the obtained JV characteristics the photovoltaic performance of the as fabricated solar cells is calculated using following equations:

$$FF = P_{max} / J_{sc}\, V_{oc} \quad (4)$$

The photo-conversion efficiency (%) of DSSC is calculated using:

$$\eta = J_{sc}\, V_{oc}\, FF / P_{in} * 100\ (\%) \quad (5)$$

where, FF is the fill factor, P_{max} is the maximum power, J_{sc} is the short circuit current density (mA/cm^2), V_{oc} is the open circuit voltage (V), η is the photo-conversion efficiency (%), P_{in} is the incident light power, J_{max} (mA/cm^2). The calculated photovoltaic parameters are summarized in **Table 3.** The power conversion efficiency of 2.1% with J_{sc} value of 4.231 mA/cm^2, V_{oc} value of 0.767 V, and FF value of 0.666 has been obtained for the blended polymers electrolyte with the weight percentage of 6%. To further improve the device performance, electrolyte was modified by adding metal nanoparticles of carbon to the existing 6% weight of electrolyte. The photo-conversion efficiency of 3.1 % with J_{sc} of 1.120 mA/cm^2, V_{oc} of 0.768 V and FF of 0.578 is obtained for 20 mg carbon loaded blended polymers electrolyte. From Fig. 5 (a) and (b), it is evident that there is a significant increase in the performance of blended polymers and modified blended polymers with carbon electrolyte. JV curves start increasing from 2 weight percentage to 6 weight percentage with the efficiency of 2.1% at 8 weight percentage the performance of the JV drops which may be due to Similarly for the modified polymer the performance of the JV curves increases from 5 mg loading to 20 mg and at 25 mg the JV drops which may be due to 3.1% The observed trend may be due to the increase in the amorphous nature of the blended polymers electrolyte on the addition of carbon filler that improved the penetration of polymer

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electrolyte into the porous TiO_2 electrode. Hence, the reduced interfacial recombination of electrons have led to high Jsc and fill factor values of the cell . The stability of the gel electrolyte and carbon filled G-DSSCs was studied at room temperature over a period of time for the sealed devices. The efficiency of the carbon filled G-DSSC did not change even after 600 hours and retained 85 % of the initial efficiency, whereas the DSSC using gel electrolyte decreased to 70% of the initial value during the same period. These results confirm that gel electrolyte can be used for the fabrication of a stable quasi solid state DSSC

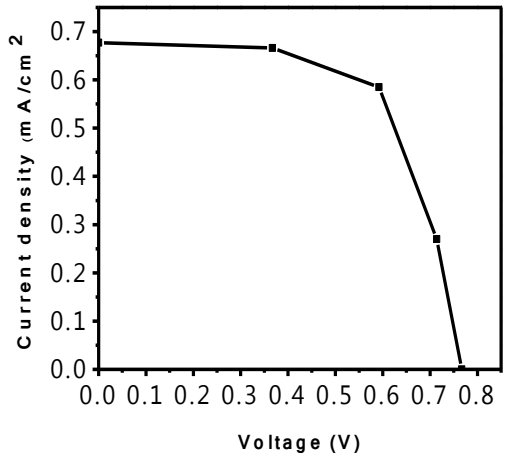

Fig. 5. (a) PEO- Blended Polymer efficiency

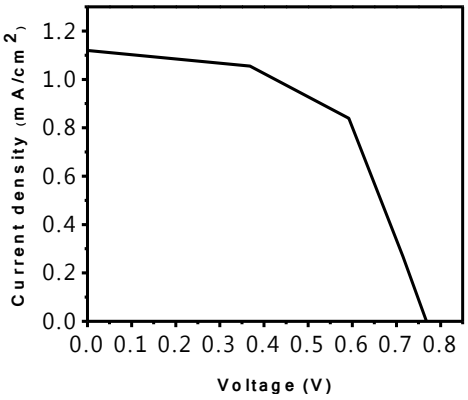

Fig.5. (b) PEO- Blended Polymer with carbon filler efficiency

Table. 3. Detailed photovoltaic performances of the two gel state DSSCs

Sample	Weight %	Voc V	Isc mA	Jsc (mA/cm^2)	FF	Efficiencies η (%)
PEO- Blended Polymer	6	0.767	6.77	4.231	0.666	2.1
PEO- Blended Polymer	20	0.768	1.120	7.000	0.578	3.1

With carbon						

4. CONCLUSIONS

The Quasi-solid state DSSCs were fabricated successfully by using PEO-blended polymer based gel electrolyte with N719 dye as a sensitizer. The presence of blended polymer moiety in the present gel system is helpful in the dissociation of added alkali salts, leading to higher ionic conductivity. Gel electrolyte with carbon show high ionic conductivity and JV characteristics compared with the other gel systems. A photo-conversion efficiency of 2.1 % was achieved for PEO-blended polymer G-DSSC under 100 mW/cm2, AM 1.5 G. EIS study reveals that charge transport resistance at Pt/ electrolyte interface is lower, for blended polymer based device, than that of the other devices. Furthermore, the introduction of carbon metal as filler in to the PEO- blended polymer gel system can improve the efficiency upto 3.1 %. The device fillred with carbon nanoparticles is highly stable for more than 600 hours and retains 95% of the initial PCE under ambient conditions.

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