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Efficiency enhancement of dye-sensitized solar cells with addition of additives (single/binary) to ionic liquid electrolyte

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Abstract. The effect of addition of single and binary additives on the performance of dye-sensitized TiO_2 solar cells based on electrolytes containing an ionic liquid (IL), 1,2-dimethyl-3-propylimidazolium iodide (DMPII) has been studied. Among the seven additives used, the addition of 2-(dimethylamino)-pyridine (DMAP) to IL resulted in best cell efficiency, which showed further enhancement with the addition of 5-chloro-1-ethyl-2-methylimidazole (CEMI) as second additive. The efficiency of the dye-sensitized solar cell (DSC) based on an electrolyte containing binary additives (DMAP and CEMI in equal molar ratios) has been found to increase by 62·5% from 4·35 to 7·07%. The dependence of different photovoltaic performance parameters (V_{oc}, J_{sc}, ff, η) of DSC upon temperature has been studied over a 30–120°C range and only a small decrease in conversion efficiency has been observed. The electrolyte containing binary additives (DMAP and CEMI) shows best cell performance up to 120°C.

Keywords. Dye-sensitized solar cell; additives; electrolyte; ionic liquid; efficiency.

1. Introduction

Since the report of O'Regan and Gratzel (1991), there has been a growing interest in dye-sensitized solar cells (DSCs) as an alternative to the conventional silicon-based solar cells because of their simple fabrication process and potential for low cost production. The DSCs efficiently use the dye's capacity to inject photo-excited electrons into the conduction band of wide bandgap nanocrystalline TiO_2. However, there are some limitations that restrict the commercialization of DSCs, such as (i) low photo-conversion efficiency, (ii) long term instability and (iii) no proven low manufacturing cost process. Among these problems, the long term instability is the major drawback for the commercialization of DSCs and is generally attributed to the evaporation and leakage of liquid electrolyte, if the cell is not properly sealed. In addition, the permeation of oxygen and water and their reaction with the electrolyte can also affect the cell performance. In order to solve these problems, electrolytes prepared from IL (room temperature molten salt), which is nonvolatile solvent and gel electrolytes containing an IL and suitable polymers, have been proposed (Wang *et al* 2002, 2003; Murai *et al* 2003). The problem of leakage of the liquid electrolyte can be addressed by using polymer gel electrolytes but the lower cell performance limits their use. Similarly, the volatility of the liquid electrolyte can be avoided by using electrolytes based on ILs, which have negligible vapour pressure along with high conductivity and wide electrochemical stability.

DSCs generally consist of a dye-coated wide bandgap nanocrystalline TiO_2, which is sandwiched between two transparent conducting oxide (TCO) coated glasses and a liquid electrolyte containing iodide/tri-iodide redox couple (Kang *et al* 2007a, b; Han *et al* 2008, 2009). The electrolyte is one of the main constituent of the cell, which provides internal ionic conductivity by diffusing iodide/tri-iodide within the TiO_2 layer and is an important factor in determining the performance of the cell (Kusama and Arakawa 2004a, b, c, d; Kusama *et al* 2005; Fukui *et al* 2006; Lee *et al* 2007; Lan *et al* 2008). The maximum efficiency of DSC based on liquid electrolytes containing organic solvents is reported to be up to 11% at 100 mA/cm^2 illumination. The cell performance of DSC with acetonitrile based electrolyte has been observed to drastically fall at temperature above 60°C because of the evaporation or leakage of acetonitrile due to its lower boiling point at 82°C (Green *et al* 2008).

The addition of different additives also affects the performance of DSC. The addition of pyridine based additives generally result in an increase in V_{oc}, *ff* and a decrease in J_{sc} which has been explained to be related to the size, ionization energy and dipole moment of the additive molecule (Kusama *et al* 2003; Kusama and Arakawa 2004a). The addition of allyl isonicotinate (AIN) has been reported to result in the negative shift of the conduction band edge of the TiO_2 and its presence in the electrolyte suppresses recombination of the injected electrons (Shi *et al* 2009). The improvement in the performance of DSC with the addition of thiazole, tri-

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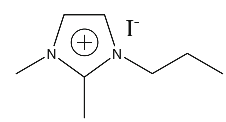
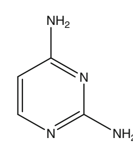
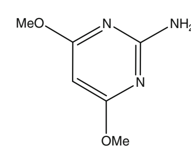
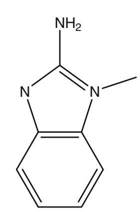
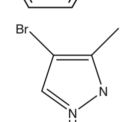
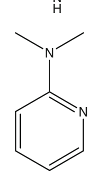
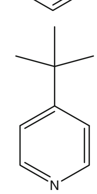
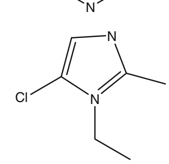
azole and benzimidazole type additives has been correlated with the calculated partial charge, size and dipole moment which is related to the donicity of the additives (Kusama and Arakawa 2004b, c, d). The electron donicity of the additives influences the interaction with the TiO_2 photoelectrode which affects DSC performance.

The main aim of the present study is to optimize the high temperature stable solvent based electrolyte by using binary additives and to study the effect of temperature on the performance of DSC. A mixture of ethylene carbonate (EC) and gamma butyrolactone (GBL), having boiling points at 248 and 204°C, respectively has been used as a solvent in place of acetonitrile. The single additive giving best cell performance is first identified and then the electrolyte composition has been further optimized with binary additives. The performance of DSC having an electrolyte with binary additives was compared with DSC using acetonitrile based electrolyte. The effect of operating temperature (30–120°C) on the photovoltaic parameters (V_{oc}, J_{sc}, *ff*, η) of the cells has also been investigated.

Table 1. Ionic liquid (IL) and different organic compounds used as additives.

Additives	Abbreviation	Chemical structure
Ionic liquid: 1,2-dimethyl-3-propylimidazolium iodide	DMPII	
2,4-diaminopyrimidine	DAP	
2-amino-4,6-dimethoxypyrimidine	ADMP	
2-amino-1-methylbenzimidazole	AMB	
4-bromo-3-methylpyrazole	BMP	
2-(dimethylamino)-pyridine	DMAP	
4-tert-butylpyridine	TBP	
5-Chloro-1-ethyl-2-methylimidazole	CEMI	

2. Experimental

2.1 *Preparation of TiO_2 photoelectrode and Ru(II) dye coating*

The TiO_2 paste (Ti-nanooxide D, Solaronix) was deposited on the conducting glass with a fluorine-doped tin oxide layer (FTO, TEC 8/2·3 mm, 8 Ω/□, Pilkington) by using screen-printing method. The resulting layer was calcined for 2 h at 470°C in a muffle furnace. This process was repeated three times or until a thickness of 15 μm was obtained. The area of the prepared porous TiO_2 electrode was 25 mm^2 (5 × 5 mm). Dye absorption was carried out by dipping the TiO_2 electrode in a 4×10^{-4} M *t*-butanol/acetonitrile (Merck, 1:1) solution of the standard ruthenium dye: N719 (Solaronix) for 48 h at 25°C. The photoelectrode was then washed, dried and immediately used to study performance of the solar cell.

2.2 *Fabrication of dye-sensitized solar cell*

Transparent counter electrodes were prepared by placing a few drops of 10 mM hydrogen hexachloroplatinate (IV) hydrate (99·9%, Aldrich) and 2-propanol solution on drilled FTO glass (TEC 8/2·3 mm, Pilkington). After calcining it at 450°C for 2 h, the counter electrodes were assembled with dye absorbed TiO_2 photoelectrodes. The two electrodes were separated by 25 μm surlyn and sealed by heating. The internal space was filled with electrolyte and the drilled hole was sealed with surlyn and coverglass.

2.3 *Photovoltaic characterization*

The photoelectrochemical properties of the DSCs were measured by using a computer-controlled digital source meter (Potentiostat/Galvanostat Model 273A, EG & G) and a solar simulator (AM 1·5, 100 mW/cm^2, Oriel) as a light source.

3. Results and discussion

The chemical structures of IL (DMPII) and seven additives used in the present study are given in table 1. The additives used have been selected on the basis of the performance of different electrolytes containing these additives in DSCs (Kusama and Arakawa 2004a, b, c, d; Kusama *et al* 2005). Single as well as binary additives have been used in the present study to improve performance of the electrolyte. The complete list of electrolytes used in the present study along with their composition and nomenclature is given in table 2. The 17 electrolytes ($E1$–$E17$) studied in the present case can be classified into the following three categories: (a) the electrolytes ($E1$–$E4$) containing different concentrations of IL, (b) the electrolytes ($E5$–$E11$) containing IL and different additives in 0·5 M concentration and (c) the electrolytes ($E12$–$E17$) containing IL and different binary additive mixtures in equal molar ratio.

The performance of the electrolyte, out of $E1$–$E17$, showing best cell performance has been compared with a reference electrolyte ($E18$) based on acetonitrile. The above three categories of electrolytes have been studied and the results obtained are discussed below.

3.1 *Effect of IL (DMPII) concentration*

The acetonitrile based electrolytes may not be suitable at higher temperatures due to its lower (82°C) boiling point. As one of the motive behind the present study is to develop electrolytes which are stable at higher temperatures, so the

Table 2. Composition of different electrolytes.

#No.	Solvent	Redox couple	Electrolyte
E1	EC and GBL	0·1M LiI + 0·05M I_2	0·1M DMPII
E2	″	″	0·2M DMPII
E3	″	″	0·3M DMPII
E4	″	″	0·4M DMPII
E5	″	″	0·3M DMPII + 0·5 M DAP
E6	″	″	0·3M DMPII + 0·5 M ADMP
E7	″	″	0·3M DMPII + 0·5 M AMBI
E8	″	″	0·3M DMPII + 0·5 M BMD
E9	″	″	0·3M DMPII + 0·5 M DMAP
E10	″	″	0·3M DMPII + 0·5 M TBP
E11	″	″	0·3M DMPII + 0·5 M CEMI
E12	″	″	0·3M DMPII + 0·5 M DMAP + 0·5 M DAP
E13	″	″	0·3M DMPII + 0·5 M DMAP + 0·5 M ADMP
E14	″	″	0·3M DMPII + 0·5 M DMAP + 0·5 M AMBI
E15	″	″	0·3M DMPII + 0·5 M DMAP + 0·5 M BMD
E16	″	″	0·3M DMPII + 0·5 M DMAP + 0·5 M TBP
E17	″	″	0·3M DMPII + 0·5 M DMAP + 0·5 M CEMI
E18	Acetonitrile	″	0·7M DMPII + 0·125M TBP

binary solvent mixture containing EC (bp = 248°C) and GBL (bp = 204°C) in 30:70 volume ratio has been used in place of acetonitrile and 0·1 M LiI + 0·05 M I_2 has been used as the redox couple. The IL (DMPII) in different molar concentrations was added to the binary solvent mixture and the redox couple. The I–V measurements were performed on DSCs with electrolytes ($E1$–$E4$) containing different concentrations of DMPII, and the solar cell performance is given in figure 1. The open-circuit voltage (V_{oc}) increases with an increase in the molar concentration of DMPII from 0·1 to 0·3 M, but decreases for electrolyte containing 0·4 M DMPII. It can be due to the presence of a large number of ions at higher concentration of IL, which hinders ion diffusion. The cell based on electrolyte $E3$, containing 0·3 M DMPII, shows a maximum value of V_{oc} (0·608 V). The short-circuit photocurrent density (J_{sc}) increases with an increase in the concentration of IL and a maximum value of 16·10 mA/cm^2 has been obtained for the electrolyte $E4$, containing 0·4 M DMPII and 15·92 mA/cm^2 for electrolyte $E3$, containing 0·3 M DMPII. The fill factor (ff) of the solar cell is maximum, 0·493 for $E2$, whereas solar energy conversion efficiency (η) is maximum (4·35%) for $E3$. Different solar cell parameters (V_{oc}, J_{sc}, ff, η) have been calculated for DSCs based on these electrolytes and are plotted in figure 2 as a bar diagram. Thus, electrolyte $E3$ having composition, EC + GBL (30:70) + 0·1 M LiI + 0·05 M I_2 + 0·3 M DMPII, shows best solar

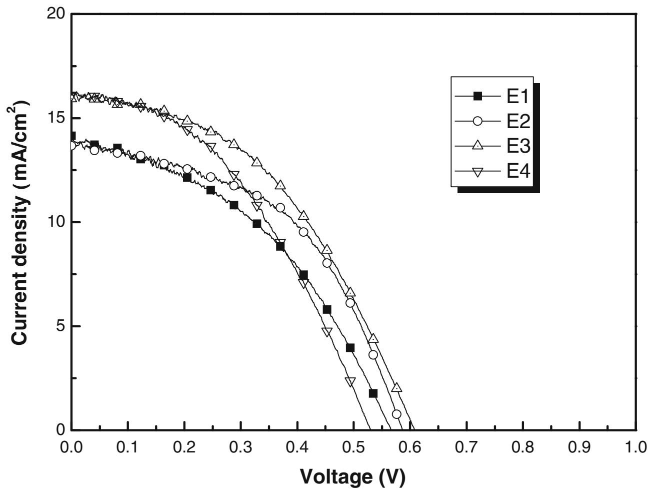


Figure 1. Current density versus voltage curves for dye sensitized TiO_2 solar cells with electrolytes ($E1$–$E4$) containing different concentrations of DMPII. Light intensity, 100 mW/cm^2, AM 1·5 .

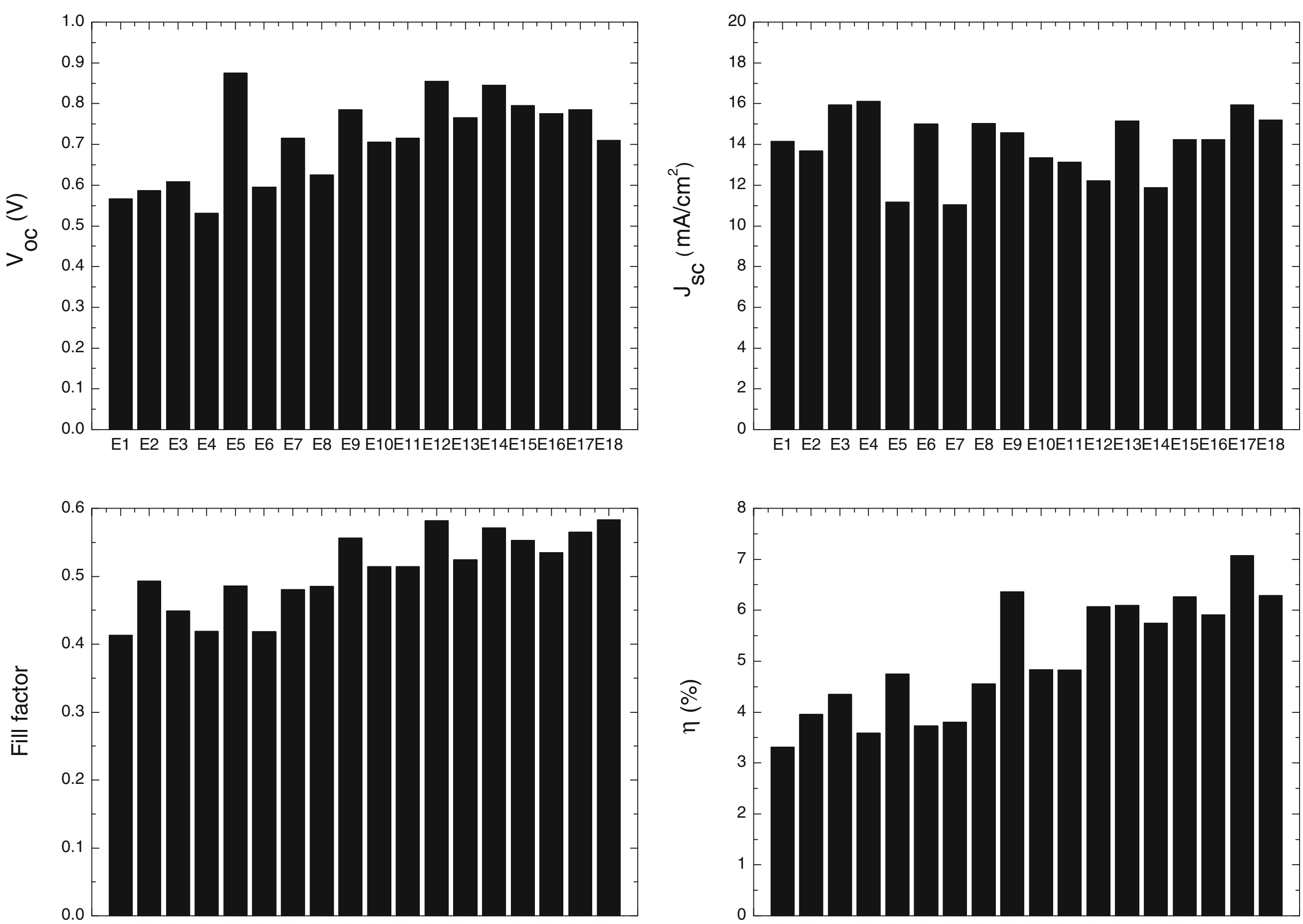


Figure 2. V_{oc}, J_{sc}, ff and η of DSCs containing various electrolytes ($E1$–$E18$). Light intensity, 100 mW/cm^2, AM 1·5.

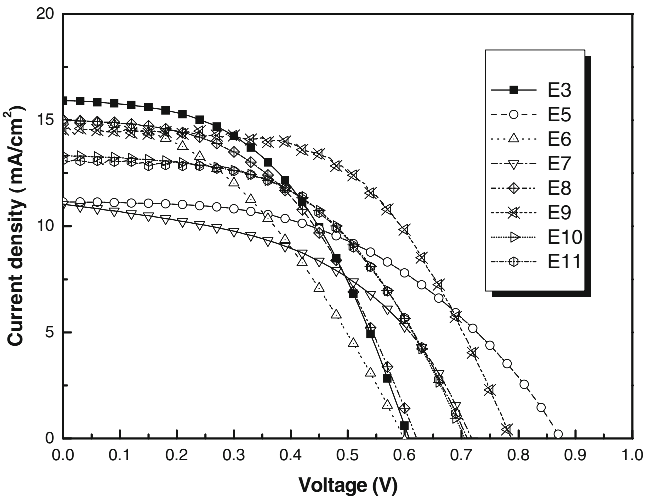


Figure 3. Current density versus voltage curves for dye sensitized TiO_2 solar cells with electrolyte ($E3$) containing different additives ($E5$–$E11$). Light intensity, 100 mW/cm^2, AM 1·5.

cell performance and this electrolyte has been used in further studies.

3.2 *Effect of single additive*

The IL electrolyte $E3$, with best solar cell performance, has been used as the base electrolyte in this part of the study. The seven additives, listed in table 1, were added to $E3$ in 0·5 M concentration and the I–V measurements of the DSCs containing these electrolytes ($E5$–$E11$) were recorded and are given in figure 3. The values of V_{oc}, J_{sc}, *ff* and η for these electrolytes have also been calculated and are given in the bar diagram (figure 2). The addition of different additives to the IL electrolyte $E3$ results in a decrease in J_{sc} and an increase in the V_{oc} of the solar cell. The addition of alkyl amino pyridine as additive has been reported to decrease the size of ionization energy and as a result, V_{oc} of the cell increases (Kusama and Arakawa 2004a, b, c, d). All the electrolytes ($E5$–$E11$) containing an additive have a lower J_{sc} value than the cell without an additive ($E3$). Electrolyte containing AMB showed a much lower (11·15 mA/cm^2) value of J_{sc} than other additives. The highest (15·02 mA/cm^2) and lowest (11·04 mA/cm^2) J_{sc} has been observed for electrolytes containing BMP and AMB, respectively. The change in J_{sc} observed by using different additives is related to the interface between the TiO_2 and the electrolyte. The additives generally increase the desirable electron transfer reaction and decrease the efficiency loss steps like the back electron transfer reaction (Kusama and Arakawa 2004b; Shi *et al* 2009). Among the seven additives used, DAP has the highest V_{oc}, 0·875 V, which is comparable to the maximum V_{oc} of 0·9 V for a cell consisting of TiO_2 electrode and I^-/I_3^- redox system, but one of the lowest J_{sc}, 11·15 mA/cm^2. The enhancement in *ff* of the solar cell and the solar energy conversion efficiency is highest for electrolyte containing DAP. The maximum η (6·36%) has been observed for this electrolyte $E9$, which is higher by 46·2% than for the base electrolyte $E3$ (4·35%) containing no additive. The enhancement of the solar cell performance by the pyridine additives is generally due to the donating properties of the nitrogen lone pair, whereas in case of additives based on benzimidazole, the decrease in J_{sc} is related to its size. The use of dialkylamine additives results in higher J_{sc} and the addition of DAP and DMAP result in a minimum decrease in photocurrent density and J_{sc} of 15·00 and 14·56 mA/cm^2 has been observed for electrolytes containing these additives, respectively. The effect on the photovoltaic parameters observed by changing the chemical nature of the additives is related to the electron donating ability and the hydrogen bonding of the additives. The changes in these parameters are also evident from the bar diagram given in figure 2. On the basis of above results it has been observed that the electrolyte $E9$ containing DMAP ($E3 + 0·5$ M DMAP) shows best solar cell performance and this electrolyte has been used in further studies.

3.3 *Effect of binary additives*

It has been observed from literature that generally a distinct group of single additives is used to enhance the performance of electrolytes and the additive resulting in optimum properties is then used in DSC. However, in the present study, after optimizing the performance of the electrolytes containing a single additive, binary additive mixtures were also used to study their effect on the performance of DSCs. The electrolyte ($E9$) containing 0·5 M DMAP, which showed best cell performance has been used as the base electrolyte and the remaining six additives (table 1) were added to $E9$ in 0·5 M concentration. The compositions of all the electrolytes ($E12$–$E17$) containing binary additives in equal molar ratio are given in table 2. The I–V characteristics of solar cells based on these electrolytes are given in figure 4 and also

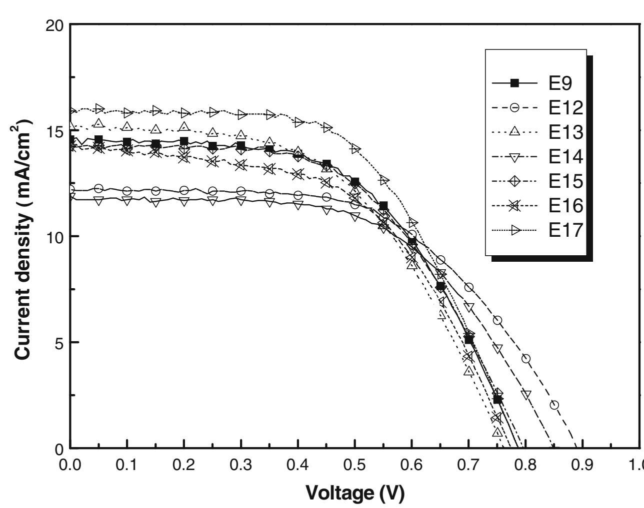


Figure 4. Current density versus voltage curves for dye sensitized TiO_2 solar cells with electrolyte ($E9$) containing different binary additives ($E12$–$E17$). Light intensity, 100 mW/cm^2, AM 1·5.

in the bar diagram (figure 2). The highest V_{oc}, 0·855 V, has been observed for electrolyte $E12$ containing additives DAP and DMAP along with the highest value (0·582) of *ff* of the solar cell. However, the J_{sc}, 12·20 mA/cm^2 and efficiency, 6·07% is quite lower than for the electrolyte $E9$ having base composition. The electrolyte $E17$ containing the additives CEMI and DMAP shows highest J_{sc} (15·92 mA/cm^2) along with maximum solar energy conversion efficiency, 7·07%. The minimum J_{sc} (11·88 mA/cm^2) and η (5·74%) has been observed for electrolyte $E14$, containing AMB and DMAP. Thus the use of suitable binary additive mixtures has been observed to enhance the J_{sc} and η to a higher value than that obtained with the single additives. The maximum J_{sc} of 15·92 mA/cm^2 and efficiency, 7·07%, has been obtained for DSC containing electrolyte $E17$, which contains binary additives (DMAP and CEMI) in equal molar concentration.

For a comparison of the results reported in above sections (3·1, 3·2 and 3·3) for electrolytes containing IL, single and binary additives, respectively the $I–V$ characteristics have been plotted in figure 5 for electrolytes $E3$, $E9$ and $E17$ which show best cell performance in each category. The V_{oc}, J_{sc}, *ff* and η values for these electrolytes are already available in the bar diagram given in figure 2. From the results given in figure 5, it has been observed that the photovoltage (V_{oc}) of the electrolyte $E3$, 0·608 V, increases to 0·785 V in $E9$ with the addition of DMAP as the single additive and the V_{oc} does not change with the addition of the second additive, CEMI and electrolyte $E17$ containing the binary additive mixture of DMAP and CEMI also shows the same value of V_{oc} of 0·785 V. The J_{sc} of cell with electrolyte $E3$ is 15·92 mA/cm^2, which decreases to 14·56 mA/cm^2 for cell with electrolyte $E9$ and then increases to 15·92 mA/cm^2 for the cell with electrolyte $E17$. Thus the electrolyte $E17$ containing binary additives show the same current density as observed in the cell with the base electrolyte ($E3$) and the decrease in J_{sc} with the addition of single additive, DMAP, is compensated with the addition of second additive CEMI. The *ff* of the solar cell increases from 0·449 for $E3$ to 0·556 for $E9$, with the addition of DMAP and to 0·565 with the addition of CEMI as the second additive in E17. The most significant change has been observed in η which increases by 46·2% from 4·35% for $E3$ to 6·36% for $E9$ with the addition of a single additive DMAP and by 11·2% to 7·07% for $E17$ with the addition of second additive CEMI. Thus an increase of 62·5% has been observed in the cell efficiency with the addition of binary additives to the IL. The above results show that the addition of binary additives in equal molar concentration results in an improvement in the value of different photovoltaic parameters (V_{oc}, J_{sc}, *ff*, η) of the solar cell. The electrolyte containing binary additives shows better solar cell performance than for electrolyte containing a single additive.

The electrolytes based on acetonitrile are generally used in DSCs, as they show better cell performance at room temperature. The $I–V$ characteristics of the electrolyte $E17$ having best cell performance has been compared with acetonitrile based electrolyte ($E18$) and the results are plotted in figure 6. It has been observed that the cell based on electrolyte ($E17$) containing binary additives shows relatively higher values of V_{oc}, J_{sc} and η than for the cell with electrolyte ($E18$) based on acetonitrile at room temperature.

3.4 *Effect of temperature*

The stability of electrolyte ($E17$) containing binary additives has been checked by studying the $I–V$ characteristics for the cell with electrolyte $E17$ and $E18$ in the 30–120°C temperature range. The variation of photovoltaic parameters (V_{oc}, J_{sc}, *ff* and η) with temperature have been plotted in figure 7 for both the cells. The effect of temperature on the

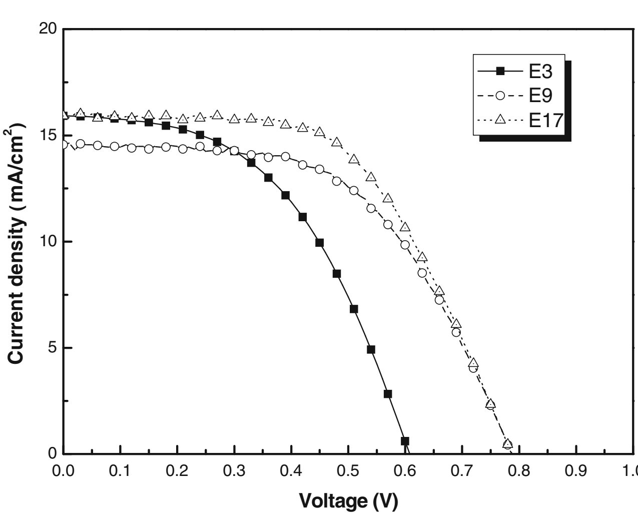


Figure 5. Current density versus voltage curves for dye sensitized TiO_2 solar cells containing different electrolytes ($E3$, $E9$ and $E17$). Light intensity, 100 mW/cm^2, AM 1·5.

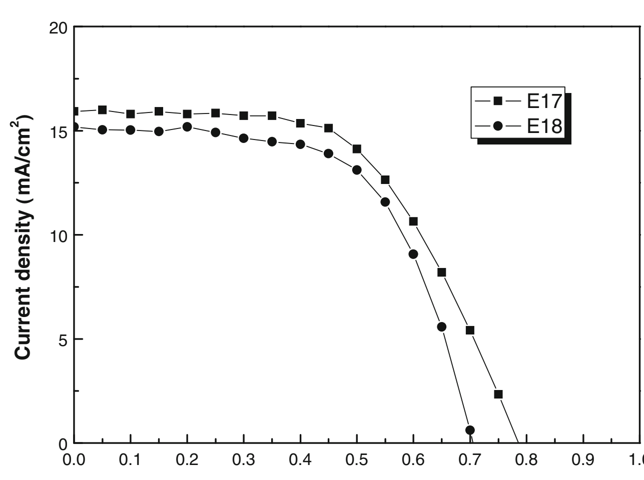


Figure 6. Current density versus voltage curves for dye sensitized TiO_2 solar cells with $E17$ and $E18$ electrolytes. Light intensity, 100 mW/cm^2, AM 1·5.

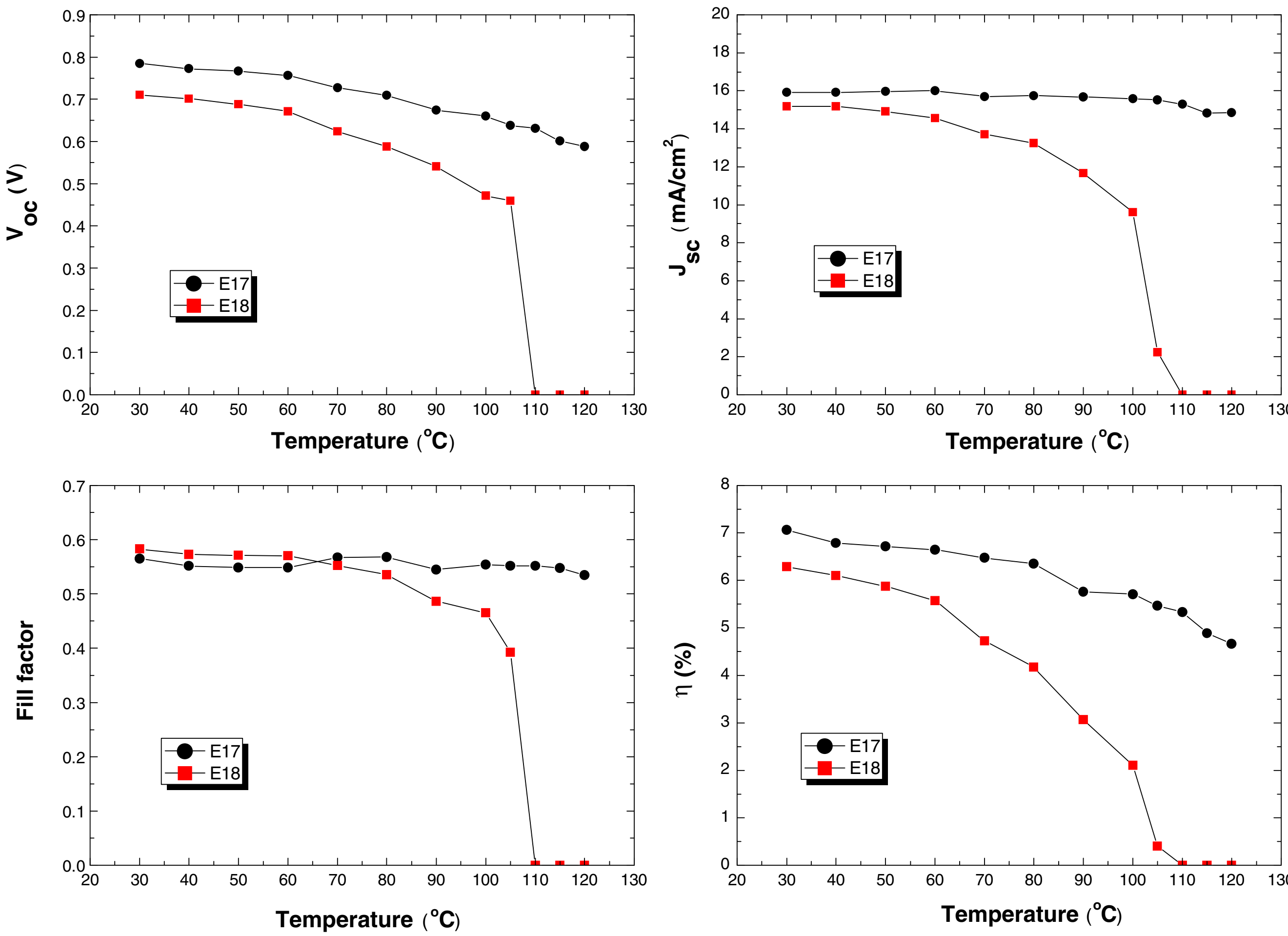


Figure 7. Variation of photovoltaic parameters (V_{oc}, J_{sc}, *ff*, η) with temperature for dye sensitized TiO_2 solar cells containing $E17$ and $E18$ electrolytes. Light intensity, 100 mW/cm^2, AM 1·5.

performance of DSC based on electrolyte containing an IL, propyl-methyl-imidazolium iodide (PMII) has been reported earlier over the 5–50°C temperature range only (Berginc *et al* 2007). In the present case, variation of the efficiency of solar cell with temperature shows only a small decrease up to 55°C for electrolytes based on acetonitrile. However, a further increase in temperature results in a sharp decrease in η which could be due to the evaporation of acetonitrile due to its lower boiling point. Thus electrolytes based on acetonitrile are not suitable for use in DSC at temperatures above 60°C. In comparison, the efficiency of DSC containing electrolyte ($E17$) based on binary additives and with EC and GBL as binary solvent mixture does not show much decrease in efficiency at temperatures up to 120°C. The electrolyte ($E17$) can be used in DSC at higher temperatures without any appreciable decrease of efficiency. The decrease in efficiency at higher temperatures is generally due to the recombination reactions which occur at the interface with the TiO_2 or between the front transparent conducting oxide (TCO) and the electrolyte, where the photo-generated electrons react with tri-iodide (Berginc *et al* 2007). The recombinations are generally expected to be more pronounced at higher temperatures.

The effect of operating temperature (30–120°C) on the photovoltaic parameters of DSC has been studied and the variation of photovoltaic performance parameters with temperature is given in figure 7. The open circuit voltage (V_{oc}) decreases with temperature and the temperature coefficient (dV_{oc}/dT) has been found to be $-2{\cdot}23$ mV/°C and it generally depends upon the recombination probability. The decrease in V_{oc} can be explained in terms of the shift of the standard reduction potential (E_o) of the I^-/I_3^- redox couple. E_o shifts in the negative direction with increasing temperature, because of the relationship $dE_o/dT = -0{\cdot}186$ mV/K, where K is the temperature in Kelvin, resulting in a decrease in V_{oc} with increasing temperature according to the relation $|V_{oc} = V_{FB} - E_o|$, where V_{FB} is the flat band potential of the TiO_2 surface (Zaban *et al* 1998; Kang *et al* 2000). Assuming that V_{FB} remains the same, the negative shift of E_o can decrease the V_{oc} value of DSC with increasing temperature of the cell (Pleskov and Gurevich 1986; Lee *et al* 2011).

J_{sc} does not show any decrease with temperature. It suggests that an increase in diffusion coefficient with increase in temperature, which enhances J_{sc}, has been compensated by an increase in the probability of recombination, which decreases J_{sc}. The temperature effect on the fill factor is also very small and *ff* shows only a small decrease with temperature. The conversion efficiency, which depends upon V_{oc}, J_{sc} and *ff* of the cell, has been observed to show a small decrease with an increase in temperature. However, in comparison for the cell containing electrolyte ($E18$) based on acetonitrile, the decrease in V_{oc}, J_{sc}, *ff* and η is much larger and all the parameters approach zero value at 100°C. Thus the small change in all photovoltaic performance parameters with temperature for the cell containing binary additives show the stability of electrolyte at higher temperatures. This is important as the temperature of the solar cell under actual operating outdoor conditions often exceeds 50°C, so good performance at higher temperatures is desirable for practical applications of DSCs. However, further studies are required to study the performance of DSC at higher temperatures and for prolonged periods along with effect of the concentration of additives.

4. Conclusions

The effect of addition of single and binary additives to the electrolyte of DSCs based on an IL, 1,2-dimethyl-3-propylimidazolium iodide has been investigated. The addition of single additive to IL results in an increase in V_{oc} along with a decrease in J_{sc}. The efficiency of the cell at room temperature has been observed to increase by 46·2% from 4·35 to 6·36% with the addition of DMAP. For optimizing the performance of the electrolyte, second additive was added to electrolyte containing DMAP and the best cell performance was observed for DSC based on electrolytes containing binary additive mixture of DMAP and CEMI in equal molar ratio. A 11·1% increase in the conversion efficiency, from 6·36 for electrolyte with single additive to 7·07% for electrolyte containing binary additives has been observed. The performance of the best cell was also compared with DSC containing a standard acetonitrile based electrolyte and DSC with electrolyte containing binary additives was found to show better cell performance. The variation of photovoltaic performance parameters (V_{oc}, J_{sc}, *ff*, η) with temperature over the 30–120°C range also shows that the cell based on binary additives has better performance than the cell based on acetonitrile. Thus, the addition of binary additives lead to electrolytes with better cell performance in the 30–120°C range and hence are suitable for use in DSCs.

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