



# Efficient dye sensitized solar cell and supercapacitor using 1-ethyl 3-methyl imidazolium dicyanamide incorporated PVDF–HFP polymer matrix



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## ABSTRACT

Electrode and electrolytes are always playing a key role in developing good efficient devices. This paper reports the synthesis, characterization and application of low viscosity ionic liquid (1-ethyl 3-methyl imidazolium dicyanamide) incorporated into polymer matrix (PVDF–HFP). The samples were characterized using various experimental tools. Complex impedance spectroscopy shows that conductivity of polymer host increases with increasing ionic liquid (IL) concentration attains maxima and then decreases. Optical microscopy and scanning electron microscopy (SEM) reveals the reduction of crystalline matrix of polymer host by adding IL. The primarily fabricated devices using maximum conductivity film affirms that this material is promising candidate for dye sensitized solar cell (DSSC) and supercapacitor application.

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## Introduction

Polymer electrolyte can be defined as materials consisting of ionic salts dissolved in an appropriate polymer, which enable ionic conductivity at room temperature. It consists of an inorganic salt dissolved in a polymer host. Because of their various applications solid polymer electrolyte have becoming very popular in electrochemical devices such as thin-film batteries, fuel cells, dye sensitized solar cell, supercapacitor, sensors etc. [1–4].

Polymer electrolytes present a number of desirable properties, such as simplicity of manufacture in a wide variety of shapes and sizes, ability to form thin films, flexibility, light weight, elasticity, longer life and greater safety, since non-corrosive or explosive liquids can leak out. In contrast to the cases of brittle glassy or crystalline solid conductors, polymer materials can accommodate volume changes, which make them particularly suited for

applications together with intercalation materials, such as the anode and cathode in a rechargeable battery [5,6].

The essential condition of any materials to be used in electrochemical devices is high ionic conductivity ( $10^{-2}$ – $10^{-3}$  S/cm). Such an ionic conductivity is generally not appear in insulating polymer's like polyethylene oxide (PEO), polyvinyl alcohol (PVA), polyvinylidene fluoride (PVDF), poly(vinylidene fluoride-hexafluoropropylene) (PVDF–HFP) etc. Different approaches have been adopted by researchers in modifying polymer host matrix [7]. Recent approaches indicate towards a new kind of dispersoid materials i.e. ionic liquid (IL) to be used frequently in many area as solvents, plasticizers and as a high conducting electrolyte [8–11].

The role of IL's as electrolyte are now spreading gradually. Good conducting polymer electrolyte matrix have many advantageous properties like high ion's concentration, wider electrochemical stability, non flammability etc. which make them suitable candidate for electrochemical devices albeit its liquid nature is main problem. Keeping this problem as positive concern we have try to dispersed these IL's into polymer host matrix and develop a new kind of electrolyte. We have just concentrated our IL–polymer solid electrolytes study towards energy devices namely DSSC and supercapacitor knowing the facts that IL's area are vast. IL's are available in market depending on our own choice. Our work mostly

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focuses towards low viscosity IL's since conductivity and viscosity are inversely proportional to each other.

In present paper we have synthesized a new IL–polymer electrolyte comprising a low viscosity IL (1-ethyl 3-methylimidazolium dicyanamide, viscosity 17 cP) into PVDF–HFP matrix. We have tested our IL–polymer electrolyte for two energy devices i.e. DSSC and supercapacitor.

## Experimental

### Materials

poly(vinylidene fluoride-hexafluoropropylene) (PVDF–HFP,  $M_w = 500,000$ ), ionic liquid (1-ethyl 3-methylimidazolium dicyanamide), multiwalled carbon nanotubes (MWCNT) were purchased from Sigma-Aldrich, USA. The other chemicals like potassium iodide (KI), iodine ( $I_2$ ), acetonitrile were purchased from Qualikems Fine Chem Pvt. Ltd., Vadodara, India.

### Synthesis of IL–polymer solid electrolyte films

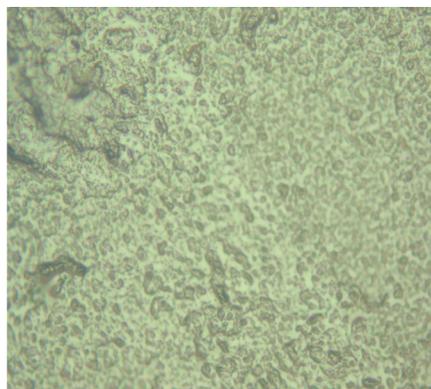
The film was prepared by solution cast technique. In this method the desired amount of host polymer (PVDF–HFP) and IL's were dissolved in a common solvent (acetonitrile). As a common procedure, a fixed amount of PVDF–HFP (~500 mg) was completely dissolved in acetonitrile. The stoichiometric amounts of IL are then added into polymer matrix and the mixture was stirred for 4–6 h. After complete dissolution the viscous IL–polymer electrolyte solution was poured in glass petridishes. After ambient drying, solvent was evaporated and free self standing IL–polymer salt complex films were obtained. These films were further dried under vacuum to remove remaining traces of solvent (if any). The obtained films were characterized by different characterization techniques. The room temperature ionic conductivity measurements, electrochemical performances of DSSC and supercapacitor were carried out with the help of a computer controlled ZEHNER (IM6) electrochemical workstation. The optical microscopy was recorded using a Leica DM 4000 M while scanning electron microscopy (SEM) micrographs were taken using HITACHI instruments (HITACHI SU 3500).

## Results and discussions

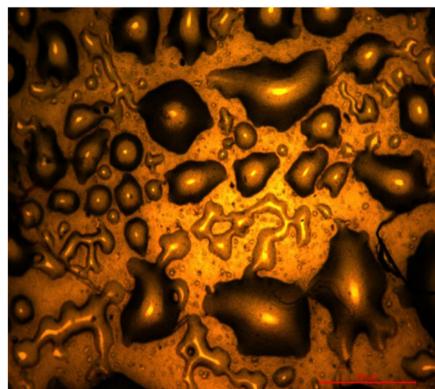
### Electrical conductivity

Ionic conductivity of IL–polymer electrolyte samples were calculated using formula:

$$\text{Conductivity } (\sigma) = G \times \frac{L}{A}$$



(a)



(b)

Fig. 1. Optical micrograph of (a) pure PVDF–HFP and (b) IL incorporated PVDF–HFP polymer electrolyte films.

Table 1

Room temperature ionic conductivity vs IL concentration in solid IL–polymer electrolyte system.

Composition (IL in wt %)	Conductivity ( $S\text{cm}^{-1}$ )
0	$1.20 \times 10^{-6}$
4	$5.53 \times 10^{-5}$
8	$1.51 \times 10^{-4}$
12	$5.30 \times 10^{-4}$
16	$1.10 \times 10^{-3}$
20	$2.90 \times 10^{-3}$
25	$3.33 \times 10^{-3}$
30	$2.20 \times 10^{-3}$
40	$1.80 \times 10^{-4}$

where  $G$  is the conductance of sample,  $L$  is the thickness of sample and  $A$  is the area of sample in contact. The evaluated ionic conductivity values with varying IL concentration are tabulated in Table 1. It is clearly observed that with increase in wt% of IL, the conductivity increases upto 25 wt% IL and then decreases. We could not get free standing films after 40 wt% IL concentration and hence we stuck our study till 40 wt%.

The increase in ionic conductivity with increasing IL concentration is correlated with free number of charges (cations and anions) provided by IL since conductivity is governed by well known equation:

$$\sigma = n \times q \times \mu$$

where  $n$  is charge carrier,  $q$  is the charge present on ions and  $\mu$  is the mobility of ions. So with increasing IL concentrations the concentration number of charge carriers ( $n$ ) increases which leads to increase in conductivity with increase in salt concentration. This trend should be throughout for increase in IL concentration (till 25 wt%) but there is decrease in conductivity afterwards which is controversial to above statement. This decrease is due to decrease in number of charge carrier ( $n$ ). As concentration of IL increases above certain limit there is increase in steric hindrance or steric crowding of ions which strikes each other and it causes barriers in its motions of ions going towards respective electrode, which leads to decrease in mobility of ions ( $\mu$ ).

### Optical microscopy

To check the surface morphology by adding IL into host polymer matrix we have recorded the optical micrographs of IL–polymer electrolyte films. Fig. 3 shows the recorded micrograph of pure PVDF–HFP polymer. Pure PVDF–HFP matrix shows well known porous morphology (Fig. 1a). From Fig. 1b, it was clear that doping of ionic liquid into polymer matrix (PVDF–HFP) produces a matrix

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