



# Low-cost tetra ethylene glycol derivatives in polymer blend electrolytes for dye-sensitized solar cells with high photovoltaic conversion efficiencies

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## ABSTRACT

The present study reports on a novel polymer blend electrolyte system comprised of poly(ethylene oxide), poly(vinylidene fluoride) and poly(methyl methacrylate) (PEO/PVdF/PMMA) with potassium iodide, iodine and novel and cost effective organic compounds such as 1-(2-(2-(2-(benzoate)ethoxy)ethoxy)ethoxy)ethoxy)benzene (BEB) and 1-(2-(2-(2-(1H-pyrazol-1-yl)ethoxy)ethoxy)ethoxy)ethyl)-1H-pyrazole (PEP) for dye-sensitized solar cell applications. The influence of the synthesized organic compounds on the ionic and photovoltaic characteristics of the electrolytes was studied. In specific, the PEO/PVdF/PMMA/KI/I<sub>2</sub>/PEP blend electrolyte exhibited high ionic conductivity of  $6.3 \times 10^{-4} \text{ S cm}^{-1}$  and the corresponding DSSC demonstrated an enhanced conversion efficiency of 9%. The high photovoltaic conversion efficiency of the cell with the PEP-doped system can be attributed to the higher conductivities in the polymer electrolyte associated with the greater cross-linking and the increased I<sup>-</sup>/I<sub>3</sub><sup>-</sup> transportation along the interaction of the tetra ethylene glycol derivative, salt and the polymer matrix.

## 1. Introduction

Organic liquid electrolyte-based dye-sensitized solar cells (DSSCs) with high power conversion efficiencies (~11%) have emerged as a promising area of research in the field of energy storage [1–3]. However, the problems of leak-prone liquid electrolytes and iodine sublimation have decreased the stability of DSSCs. The past few decades have seen significant advancement in the research on solid polymer electrolytes as prospective replacements to the liquid redox couple (I<sup>-</sup>/I<sub>3</sub><sup>-</sup>) electrolyte in DSSCs. Polymer electrolytes have more advantages of overcoming internal shorting and electrolyte leakage at the electrode surface [4–10]. Recently, the concept of blending two different polymers with mutually exclusive physical and chemical properties was introduced to enhance their electrical conductivities. The strategy of preparing polymer blends from more than one or a few polymers is generally pursued to achieve products with customized physical properties and evades the need to prepare specialized polymer systems.

As is well-established, the blending of PVDF and PEO effects a change in the polymer chains decreases the crystallization phase in the polymer and ultimately contributes to improved ionic conductivities. However, the limitations of complicated synthesis and relatively low

mechanical strength of the two-polymer blends restrict their use for commercial purposes [11–15]. To overcome these limitations, blends composed of three different polymers were also attempted to further enhance the electrical property of the polymer membrane in an electrolyte solution [16]. In fact, several reports on two-polymer electrolyte blends for energy devices, particularly battery and dye-sensitized solar cells are available in literature [13–15]. On the other hand, the three polymer blending technique has been introduced very recently to arrive at customized photovoltaic properties for specific applications [16]. In specific, their physico-chemical properties have neither been fully understood nor their advantages for electrolyte applications in DSSCs been completely explored. Moreover, the usual trend of facilitating further enhancements in the physico-chemical properties of the polymer electrolytes by introducing nanoparticle fillers/plasticizers in three-polymer blends are yet to be pursued.

The strategy of incorporating plasticizer into the polymer electrolytes is attractive as the polymer-polymer chain interaction is decreased and the segmental motion of polymer is enhanced thereby improving the ionic conductivity as well as efficiency of dye-sensitized solar cells [16,17]. A majority of the research has reported the use of inorganic salts such as TiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> [18,19] as fillers and organic nitrogenous

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compounds as plasticizer that ultimately led to high conversion efficiency and life time [17,20]. Compared to using commercially available nanoparticle fillers that are expensive, the aspect of introducing plasticizers prepared by low-cost methods is attractive from the economic viewpoint; though such tasks are non-trivial.

In the light of these considerations, we hereby report on a three-polymer blend composed of PEO, PVDF and PMMA with the advantageous presence of fluorine and oxygen that tends to enhance ion transport properties and lower the rate of re-combination along the interface of the semiconductor/polymer electrolyte contact area of the DSSC. Poly (Methyl methacrylate) (PMMA) is a tough, rubbery and moderately hard polymer that tends to be highly miscible with other polymers. Also, PMMA is known for its stability under severe service conditions. The addition of a third polymer in the blend introduces apparently high polymer chain movement in the solution and thereby increases entropy and hence the amorphous polymer phases [17]. In addition, this three-polymer blend electrolyte was doped with a plasticizer synthesized by a simple and low-cost method using tetra ethylene glycol to improve the mechanical, structural and electrical properties of the electrolyte and hence the photovoltaic properties of the corresponding dye-sensitized solar cell (DSSC).

## 2. Experimental details

### 2.1. Material synthesis

#### 2.1.1. Reagents and chemicals

Poly(ethylene oxide), Poly(vinylidene fluoride) and poly(methyl methacrylate) tetraethylene glycol, pyrazole and tetraethyleneglycol dichloride procured from Aldrich were used in the present study. Whereas the chemicals namely, iodine ( $I_2$ ), potassium Iodide (KI), potassium tertiary butoxide, dimethyl formamide (DMF), tetrahydrofuran and benzoyl chloride, were purchased from Merck.

## 3. Synthesis of plasticizers

### 3.1. Synthesis of 1-(2-(2-(2-(1H-pyrazol-1-yl)ethoxy)ethoxy)ethoxy)ethoxy)-1H-pyrazole (PEP)

An amount of 1.5 g of potassium tertiary butoxide was dissolved in 30 ml of tetrahydrofuran (THF) in a 100 ml round bottom flask and stirred well with 0.58 g of pyrazole for about half an hour. To this solution was added 0.77 g of tetraethyleneglycol dichloride and refluxed at 80 °C for 1 day. On completion of the reaction, the solvent was evaporated at room temperature followed by washing with water and brine solution. The crude product was obtained by the addition of chloroform. Finally, the pure colorless solid compound obtained (yield ~20%) after column chromatography was characterized by  $^1H$  NMR and  $^{13}C$  NMR using  $CDCl_3$  solvent. The column chromatography was carried out by using n-hexane and ethyl acetate effluents in the molar ratio 90% and 10% respectively. The schematic diagram of the above compound is shown in Fig. 1(a). The proton NMR peaks in  $\delta$  are characterized as 7.41–7.44 m 4H and 6.15–6.16 m 2H, 4.22–4.26 t 4H, 3.75–3.78 t 4H, 3.47–3.55 m 8H (Fig. 1b).  $^{13}C$  NMR peaks at 139.3, 130, and 105.3 are ascribable to the pyrazole ring peaks and 70.6, 70.4, 69.9, 52.04 to the aliphatic carbon (Fig. 1c).

### 3.2. Synthesis of 1-(2-(2-(2-(2-(benzoate)ethoxy)ethoxy)ethoxy)ethoxy)benzene (BEB)

An amount of 0.4 g of tetraethylene glycol was taken in a 100 ml round-bottom flask with 1.14 g potassium tertiary butoxide in 30 ml of tetrahydrofuran (THF). The solution was stirred for an hour followed by the addition of 1.12 g benzoyl chloride dropwise at 0 °C and room temperature stirring of the mixture continued for 3 h. Once the reaction was completed, a rotary evaporator was used to remove THF. The crude

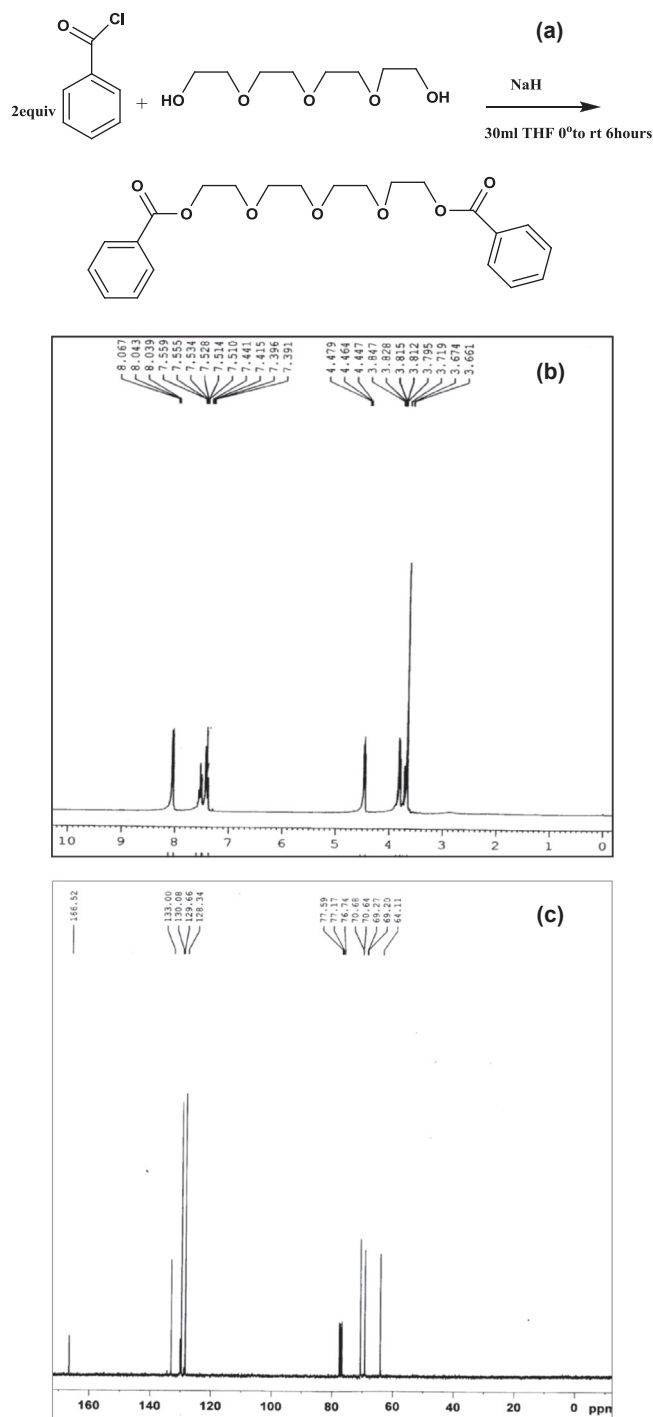


Fig. 1. (a) Schematic representation on the synthesis of PEP. (b)  $^1H$  NMR of PEP (c)  $^{13}C$  NMR of PEP.

sample was then washed using aqueous sodium bicarbonate solution and brine solution, respectively, in order to separate the crude product by the addition of chloroform. The schematic diagram of the final product obtained after purification by column chromatography using hexane elution is provided in Fig. 2(a). The column chromatography was carried out by using the effluents, namely, n-hexane and 10% ethyl acetate in the percentage ratio of 90% and 10%, respectively. The pure compound was found to be a colorless liquid and was characterized by  $^1H$  NMR and  $^{13}C$  NMR using  $CDCl_3$  solvent. The proton NMR peaks of  $\delta$  are 8 m 4H, 7.51–7.56 m 2H, and 7.3–7.4 m 4H and 4.44–4.46 t 4H, 3.81–3.83 m 4H and 3.66–3.79 m 8H (Fig. 2b).  $^{13}C$  NMR peaks in  $\delta$  of

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