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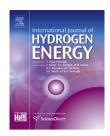
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The potential of polyurethane bio-based solid polymer electrolyte for photoelectrochemical cell application

M.S. Su'ait a,d, A. Ahmad a,*, K.H. Badri a, N.S. Mohamed b, M.Y.A. Rahman c, C.L. Azanza Ricardo d, P. Scardi d

- ^a School of Chemical Science and Food Technology, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia
- ^b Center for Foundation Studies in Science, University of Malaya, 50603 Kuala Lumpur, Malaysia
- ^c Institute of Microengineering and Nanoelectronics (IMEN), Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia
- ^d Department of Civil, Environmental and Mechanical Engineering, University of Trento, Via Mesiano, 77, 38123 Trento, Italy

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ABSTRACT

A photoelectrochemical cell was developed from bio-based polyurethane (PU), solid polymer electrolyte with lithium iodide as conducting material. At the initial stage, PU prepolymer was prepared via prepolymerization technique by reacting palm kernel oil-based monoester-OH (PKO-p) and 2,4'-methylene diphenyl diisocyanate (2,4'-MDI). The polyurethane electrolyte film was then prepared by inclusion of varying amount of lithium iodide (LiI) via solution casting technique. The formation of urethane linkages (NHCO backbone) and the chemical interaction between segmented polyurethane and lithium ion from LiI salts were confirmed by ATR-FTIR technique. Thermal studies carried out by TGA have proven the occurrence of polymer-salt complexation. Structural analysis by XRD has revealed that polyurethane electrolytes with 25 wt.% LiI reduced the semi-crystalline characteristics of plasticized polyurethane. The SEM morphological observation on the fractured film indicated the absence of phase separation. The ionic conductivity increased with the addition of 25 wt.% LiI resulted in the highest conductivity of 7.6 \times 10⁻⁴ S cm⁻¹. The temperature dependence conductivity of the electrolytes obeyed the Arrhenius law with the pre-exponential factor, σ_0 of 2.4×10^{-3} S cm⁻¹ and activation energy, E_a of 0.11 eV. A dye-sensitized solar cell of FTO/TiO2-dye/PU-LiI-I2/Pt give a response under light intensity of 100 mW cm $^{-2}$ indicated the photovoltaic effect with the $J_{\rm sc}$ of 0.06 mA cm $^{-2}$ and Voc of 0.14 V respectively. These properties exhibited promising potentials for photoelectrochemical cell giving the focus on bio-based polymer electrolyte.

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E-mail address: azizan@ukm.my (A. Ahmad).

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^{*} Corresponding author. Tel.: +60 389215439; fax: +60 389215410.

1. Introduction

Nowadays, the world is facing a major crisis in regards to sustainable, safe and environmental friendly energy resources. The finding of ionic conductivity in polymer material complex with salt gives a breakthrough to the development of solid-state electrochemical devices for energy storage such as rechargeable batteries, photoelectrochemical cell, supercapacitor and electrochromic windows [1]. A photoelectrochemical solar cell (PEC) is an electrochemical device that can generate electrical and electrochemical energy upon absorption of light by one or more of the electrodes. It is generally composed of a photoactive semiconductor electrode (either n- or p-type) and metal counter electrode, which are immersed in the suitable redox electrolytes [2,3]. Recently, Grätzel cell or commonly known as dyesensitized solar cell (DSSC), which is an extended of PEC devices developed by O'Regan & Grätzel [4] received more attention due to its high power conversion efficiency under cloudy and artificial light conditions, sustainable and low-cost starting materials and also simple fabrication technology. They are based on a molecular absorber based on that emits photogenerated electrons from an excited state upon illumination into photoactive semiconductor and receives ground-state electrons from a liquid redox carrier [4-6]. Ruthenium based complexes as photosensitizers have been dominant in DSSC application due to their favorable photoelectrochemical properties and high stability in the oxidized state, making practical applications feasible [7]. At present, DSSCs based on ruthenium(II)-polypyridyl complexes have overall power conversion efficiencies over 11% under standard air mass (AM) 1.5 illumination (light intensity of 100 mW cm⁻²) [8]. The high efficiencies of the ruthenium(II)-polypyridyl based DSSCs can be attributed to their wide absorption range from the visible to the near-infrared region. In addition, the carboxylate groups attached to the bipyridyl moiety lower the energy of the ligand π^* orbital [9]. Thus, this article utilized the standard dye sensitizer. From dye-loading data reported by Sacco et al. [10], it's easy to calculate that 1 g of N719 can sensitize 1.68 m² of TiO₂, that costs 158 EUR per gram by Solaronix company, whereas it will cost more for 1.68 m² of pure silicon. In addition, there are various types of organic sensitizers such as porphyrin based [11,12], coumarin [13,14], perylene based [15,16]. Various types of metal-salts based liquid electrolytes and ionic liquid are being used together with redox couple in DSSC such as iodide based, thiocyanate based, and Cobalt based electrolytes in various type of organic solvents such as acetonitrile, 3-methoxypropionitrile and N-methylpyrrolidone [4,6,11,17-19]. To date, the power conversion efficiency of the cells is still depending on the liquid electrolytes in order to improve its performance up to 12.4% by using Cobalt (II/III)-based redox species and Porphyrinsensitizer [11]. However, the presence of liquid electrolytes poses challenges in integration of large area modules, difficulty in implementing tandem architectures, sealing issues, possible desorption and photodegradation of the attached dyes, and the corrosion of Pt counter electrode which lead to lower lifetimes of the performance and practical use of the cells [20]. A more complicated situation arises when the photoanode is in contact with the volatilization of redox-electrolytes solution by affecting the charge distribution at the semiconductor/

electrolyte interface, and thereby also the potential distribution [21]. To overcome the problem, polymer electrolyte membranes provided alternative materials to be utilized in DSSC. There are various families of classic polymer electrolytes such as gel, plasticized, ionic rubber polymer electrolytes and ion conducting polyelectrolytes. In addition, there are another two main families of hybrid inorganic-organic polymer electrolytes which is mono-phase and multi-phase polymer electrolytes as reported by Di Noto et al. [22]. Extensive studies have been conducted elsewhere in order to improve quasi-solid-state/ solid-state PEC efficiency by replacing liquid electrolytes with gel/quasi-solid polymer electrolytes [23-28], hole-transport materials [29], p-type semiconductor [30,31] and ionogels based ionic liquid [32]. Recent study by Bella et al. [25] shows solar energy conversion efficiency of quasi-solid-state DSSC at 5.41% using (35:65) BEMA:PEGMA polymer membrane swelling in liquid electrolyte NaI/I2 in acetonitrile. Joseph et al. [26] reported the quasi-solid-state DSSC containing the hybrid TEOSpoly(ethylene glycol) (PEG) gel electrolyte with an overall solar energy conversion efficiency of 4.1%. In addition, Kim et al. [27] discovered power efficiency of 4.2% by using poly(vinylidene fluoride-co-hexafluoropropylene) (PVdF-HFP) quasi-solid-state polymer electrolytes using the same LiI/I redox-electrolytes solution.

Recently, polyurethane (PU) has attracted attention as promising polymer electrolytes candidate in the future [23,24,33-36]. PU has a unique multiphase structure formed from soft and hard segment of the polymeric chain contributed by the polyols and isocyanates respectively. The soft segment of the PU can act as a polymeric solvent to solvate the cations and the hard segment can be functionalized to maintain a wide electrochemical stability to permit the fabrication of the polymer electrolytes in electrochemical devices [24]. Moreover, the strong adhesions properties of PU allow the electrode interfaces to stick together during the PEC assemble [37]. PU is synthesized by polymerization reactions between polyester polyols and diisocyanate. Conventional polyester polyols are synthesized by direct polyesterification of high-purity diacids and glycols from virgin raw materials. They are distinguished by the choice of monomers, molecular weight, and degree of branching. Whereas, bio-based polyester polyols are derived from vegetable oils that provide better elastomeric properties, flexible and easy to be mould. It offers physical properties that cannot be obtained by petrochemical-based polyether polyols such as PEG, including superior solvent, abrasion, and low resistance [38]. Research on bio-based PU synthesis from palm kernel oilbased monoester-OH (PKO-p) has been studied extensively by Badri and co-workers [37,39,40]. However, there is no report related to the potential of palm based PU bio-polymer electrolytes for solid-state electrochemical devices application. On the other hand, diisocyanate with two or more functional groups are required for the formation of polyurethane. The aliphatic isocyanate group is less reactive than the aromatic. Aliphatic isocyanates are used only if special properties such as light stable coatings and elastomeric properties are required for the final product. Even within the same class of isocyanates, there is a significant difference in reactivity of the functional groups based on steric hindrance. In the case of 2,4'-methylene diphenyl diisocyanate (MDI), the isocyanate group in the para

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