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Polymer electrolytes for dye-sensitized solar cells prepared by photopolymerization of PEG-based oligomers

Federico Bella^{a,b,*}, Elena Daniela Ozzello^b, Adriano Sacco^a,
Stefano Bianco^a, Roberta Bongiovanni^b

^a Center for Space Human Robotics @Polito, Istituto Italiano di Tecnologia, Corso Trento 21, 10129 Torino, Italy

^b Department of Applied Science and Technology - DISAT, Politecnico di Torino, Corso Duca degli Abruzzi 24, 10129 Torino, Italy

ARTICLE INFO

Article history:

Received 14 December 2012

Received in revised form

18 June 2013

Accepted 25 June 2013

Available online 26 July 2013

Keywords:

Polymer electrolyte

Dye-sensitized solar cell

UV-curing

Crosslinking density

Electrochemical

impedance spectroscopy

ABSTRACT

We report on the preparation and characterization of novel polymer electrolyte membranes for quasi-solid dye-sensitized solar cells. New methacrylic–acrylic gel-polymer electrolytes were prepared by photo-polymerization of mono/di-functional monomers. The crosslinked films were self standing, transparent and flexible. They were swelled by an iodine–iodide solution, obtaining a stable gel, where the polymeric network acts as a cage to retain the liquid, preventing its evaporation. Such a system combines the cohesive property of a solid with the high ionic conductivity of a liquid. The evaluation of the structural and physical-chemical characteristics of the polymer, combined with the electrical characterization of the membranes by means of the electrochemical impedance spectroscopy, allowed us to investigate the structure/property relationship of the material. The electric characterizations of the solar harvester based on the gel-polymer electrolyte showed a maximum photovoltaic conversion efficiency of 4.41%. Moreover, a significant improvement in the durability of the device was demonstrated with respect to the liquid electrolyte-based counterpart.

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1. Introduction

In recent years, the increased demand for energy consumption is putting pressure on the whole human society. The increasing costs and the considerable environmental impact of fossil fuels have focused a strong interest on the development of technologies for the exploitation of renewable energies [1]. In this context, solar energy has been regarded as a potential candidate because of its wide availability and inexhaustibility on the human time scale.

One of the most promising approach for a new generation of solar devices is given by dye-sensitized solar cells (DSSCs), invented by O'Regan and Grätzel in 1991 [2]. A typical DSSC comprises a nanocrystalline semiconductor (i.e. titanium dioxide, TiO₂) layer electrode, a sensitizer adsorbed onto the semiconductor surface, an electrolyte solution with a dissolved iodide/triiodide (I⁻/I₃⁻) redox couple, and a platinum (Pt) counter-electrode. As visualized in Fig. 1, the excitation of the sensitizer upon irradiation is followed by the fast injection of the resulting electrons into the conduction band of the

* Corresponding author. Center for Space Human Robotics@Polito, Istituto Italiano di Tecnologia, Corso Trento 21, 10129 Torino, Italy. Tel.: +39 0110903448.

E-mail address: federico.bella@polito.it (F. Bella).

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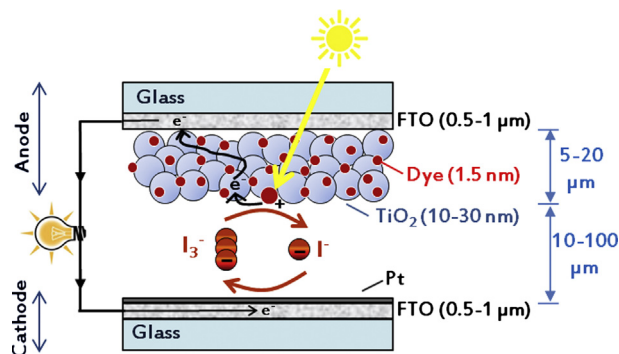


Fig. 1 – Schematic representation of the components and of the operating principle of a DSSC.

semiconductor, from where they reach the cell anode (i.e. a glass slice covered with a Fluorine doped Tin Oxide over-layer, FTO, which is a transparent conducting oxide). Dye regeneration occurs through electron donation from the I^- reducing agent. The resulting I_3^- ions are reduced in turn at the Pt counter-electrode, while electron migration from the anode closes the circuit. In recent years, DSSCs have been widely investigated because of their simple structure, easy production process and environmental friendliness [3]. At present, the efficiency of DSSCs has already surpassed 11% [4], achieving the maximum value of 12.3% at the end of 2011 [5]. Additionally, these devices can be coupled with photoelectrochemical cells in hybrid systems able to split water into hydrogen and oxygen using sunlight, alleviating the energy storage problem [6].

The appropriate selection and optimization of the materials constituting the cell components is essential both for obtaining high photovoltaic performance and to ensure their durability over time. In this context, one of the main criticality of DSSCs is related with the use of an organic solvent-based liquid electrolyte [7]. In fact, when dealing with liquids, a robust and hermetic sealing of the device is difficult, therefore liquids can evaporate and leak; moreover they can alter the adsorbed dye, dissolving water and oxygen. To overcome these critical issues, room temperature ionic liquids [8], gelled liquids [9], hole-transporting materials [10], gel-polymers [11] and polymers [12] have been recently proposed as electrolytes for quasi-solid and solid DSSCs. Actually the combination of non-volatility, good performance, flexibility on the structure design, chemical and electrochemical stability, makes the polymer-based electrolytes the best candidates to ensure high and durable sunlight conversion efficiencies of the corresponding DSSCs [13,14].

Polymer electrolyte membranes can be obtained by trapping a liquid electrolyte in a polymer as a host matrix [15]. The resulting material possesses simultaneously the beneficial aspects of liquid electrolytes (high ionic conductivity, diffusive transport and interfacial contact properties) and the cohesion typical of solids; the overall effect is a prolonged durability of the quasi-solid devices compared to liquid ones [16]. However, many experimental steps are usually required for the preparation of polymer-based electrolytes: solubilization and mixing of reactants, heating ramps, catalyzed

processes, separations and purifications steps [17]. It is easy to imagine that the set of all these steps heavily increases the production times and the cost of the final device, both on laboratory scale, and on a hypothetical industrial plant.

To overcome these problems, a powerful and innovative strategy involves the preparation of quasi-solid electrolytes by the process of photoinitiated polymerization. This technique induces the polymeric network formation by means of a rapid transformation of liquid monomers into solid membranes, with tailored physical–chemical, functional and mechanical properties. UV-induced polymerization is a solvent-free process, carried out at room temperature in few minutes, therefore it guarantees the saving of energy and time [18].

The main class of UV-curable monomers and oligomers used for the preparation of gels and membranes for electrochemical and photoelectrochemical devices is that of acrylates and methacrylates. In this context, we have successfully proposed the photopolymerization of these oligomers for the preparation of electrolytes for Li-ion batteries [19]; more recently, these materials were also considered by Passerini et al. for the same application [20]. Ryou et al. carried out an initiator-free photopolymerization of acrylates/thiol-based reactive mixtures, obtaining very flexible gel-polymer electrolytes for lithium secondary batteries [21]. Wang et al. included fluorine and sulfonic acid lithium in UV-cured networks, reaching a liquid electrolyte uptake of 144% [22]. As regards other applications in electrochemistry, acrylate/methacrylate-based UV-cured polymers were used as self-plasticized membrane for Na^+ and K^+ ion selective electrodes [23], crosslinked film with outstanding photochromic and electrochromic performances [24], and corrosion protection coatings on galvanized steel [25].

Being the promising physical-chemical and electrochemical properties of these UV-cured polymers, we decided to extend our experience on these materials to new horizons, i.e. the preparation of photo-crosslinked membranes able to trap a fair amount of iodide/iodine-based liquid electrolyte for quasi-solid DSSC application. Very recently some research groups have prepared and tested UV-cured polymer electrolytes in this field, and light-to-electricity conversion efficiencies around 3–4% were obtained [26,27]. However, the polymerization process, the characterization of the polymer electrolyte, and the membrane transport properties were not thoroughly discussed. In this paper, we propose polymer electrolyte membranes obtained by copolymerizing poly(ethylene glycol) (PEG)-based methacrylic/acrylic reactive oligomers, whose photo-curing process has been investigated by real time Fourier Transform Infrared Spectroscopy (FTIR) technique. The characterization of materials (degree of crosslinking, thermal stability, mechanical properties) and devices will be presented, and a clear correlation between the structural and electrochemical properties of the polymer electrolyte membranes and the photovoltaic performance of the corresponding cells will be discussed in detail. In particular, the elaboration of dynamic mechanical thermal analysis will allow us to justify the different photoelectrochemical behavior observed for the UV-crosslinked polymer electrolytes prepared in the presence of various amounts of reactive oligomers. These data will be cross-compared with the investigation of charge transfer impedance at the membrane/

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