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Quasi-solid-state dye-sensitized solar cells assembled with polymeric ionic liquid and poly(3,4-ethylenedioxythiophene) counter electrode



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ABSTRACT

Poly(3,4-ethylenedioxythiophene) nanofibers (PEDOT-NF) with high catalytic activity were synthesized and employed as a counter electrode in dye-sensitized solar cells (DSSCs). A polymeric ionic liquid (PIL) was used as a gelling agent and an iodide source for making a highly conductive gel polymer electrolyte. A quasi-solid-state DSSC assembled with this PIL-based gel polymer electrolyte and PEDOT-NF counter electrode exhibited high conversion efficiency of 8.12% at 100~mW cm $^{-2}$.

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1. Introduction

Dye-sensitized solar cells (DSSCs) are promising alternatives to silicon solar cells due to their low cost, easy fabrication and relatively high conversion efficiency [1,2]. Recently, impressive conversion efficiency of 12% has been achieved in a DSSC with a liquid electrolyte [3]. However, leakage or evaporation of liquid electrolyte is a critical problem that limits the long-term operation and practical use of DSSCs [4]. To overcome these problems, considerable efforts have been made to replace liquid electrolytes with gel polymer electrolytes that exhibit high ionic conductivity and improved stability [5–8]. In these gel polymer electrolytes, both cations and anions are mobile, which polarizes the electrolyte and increases the resistance of I⁻/I₃ ions [9]. The use of a single-ion conductive gel polymer electrolyte in which cations are immobilized while anions are highly mobile is therefore highly desirable. A polymeric ionic liquid (PIL) is a singleion conductor in which cationic or anionic centers are constrained to the polymer backbone. Although extensive research has been devoted to PIL-based electrolytes [10–15], their ionic conductivities are too low for practical applications. Therefore, a PIL-based gel polymer electrolyte with high ionic conductivity is required to achieve high conversion efficiencies in a DSSC. Pt has been used widely as a counter electrode in DSSCs because of its high conductivity and catalytic activity for the reduction of I₃⁻ ions. However, the high cost of Pt has driven the development of other highly efficient and low-cost counter electrodes. Recently, poly(3,4-ethylenedioxythiophene) (PEDOT) has attracted a great deal of attention as a counter electrode due to its high conductivity, catalytic activity, low cost, ease of synthesis and environmental stability [15–20]. However, there are few reports describing highly efficient quasi-solid-state DSSCs based on PIL-based electrolytes and PEDOT counter electrodes. One such study by Kawano et al. [15] reported low conversion efficiency of 1.0–1.5% for DSSCs using PIL-based solid polymer electrolyte and PEDOT counter electrode. Such a low efficiency arose from high resistance of the cell due to the use of solid-state polymer electrolyte and plain PEDOT electrode.

This report details highly efficient quasi-solid-state DSSCs assembled with a PIL-based gel polymer electrolyte and a PEDOT nanofiber (NF) counter electrode. In the PIL, the iodide anions migrate easily, while the imidazolium cations are immobilized. The PIL was important not only for increasing the concentration of I^- ions but also in forming the stable gel polymer electrolyte. The resulting DSSC exhibited high conversion efficiency of 8.12%, which was higher than that of the cell with liquid electrolyte and Pt counter electrode.

2. Experimental

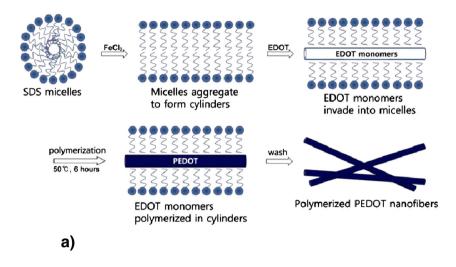
2.1. Synthesis of PMAPII and PEDOT-NF

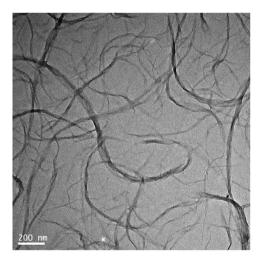
Poly(1-methyl 3-(2-acryloyloxypropyl) imidazolium iodide) (PMAPII) was synthesized as previously reported [21]. 3-lodopropyl acrylate was obtained by the reaction of 3-iodo-1-propanol and acryloyl chloride with triethylamine in tetrahydrofuran for 24 h at 0 °C. A quaternization reaction of 3-iodopropyl acrylate and *N*-methylimidazole

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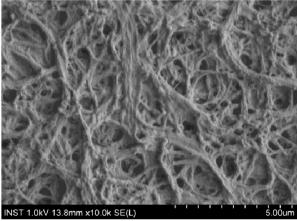
was performed in ethanol 45 $^{\circ}$ C for 24 h, and the obtained MAPII monomer was polymerized with azobisisobutyronitrile in dimethyl sulfoxide at 80 $^{\circ}$ C for 12 h. After polymerization, the resulting solution was added drop by drop to diethyl ether, and the obtained polymer was then washed successively with diethyl ether to remove

impurities such as residual monomer and initiator. PEDOT-NF was synthesized by chemical oxidative polymerization, shown schematically in Fig. 1(a). An aqueous solution of sodium dodecyl sulfate (SDS) and FeCl₃ was stirred for 1 h at 50 °C to produce micelles with a cylindrical structure. EDOT monomer was then slowly added









c)

Fig. 1. (a) Schematic illustration showing the synthesis of PEDOT-NF, (b) TEM image of the synthesized PEDOT-NF and (c) FE-SEM image of the PEDOT-NF counter electrode.

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