



Properties of inverted polymer solar cells based on novel small molecular electrolytes as the cathode buffer layer



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ABSTRACT

Novel small-molecule electrolytes were designed and synthesized for use in the cathode interlayer in bulk-heterojunction polymer solar cells (PSCs). The synthesized materials consist of polar quaternary ammonium bromide with the addition of multiple hydroxyl groups, which are N,N,N,N,N-hexakis(2-hydroxyethyl)butane-1,4-diaminium bromide (**C4**) and N,N,N,N,N-hexakis(2-hydroxyethyl)hexane-1,6-diaminium bromide (**C6**). The materials generate a favorable interface dipole through the quaternary ammonium bromide. In addition, the multiple polar hydroxyl groups increased the interface dipole magnitude. The power conversion efficiency of the devices with the interlayer was up to 9.20% with a J_{sc} of 17.22 mA/cm², a V_{oc} of 0.75 V, and an FF of 71.3%. The PCE of devices with an interlayer show better long-term stability than a device without an interlayer. Our strategy shows that it is possible to enhance the efficiency of PSCs by simple approaches without complicated syntheses.

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1. Introduction

Recently, bulk-heterojunction (BHJ) polymer solar cells (PSCs) have attracted attention due to their high potential for application as clean energy sources, as they are lightweight, flexible, and capable of large-area fabrication [1–4]. The power conversion efficiencies (PCEs) of PSCs have improved rapidly. PCEs of over 10% have been reported, due to tremendous efforts made in areas such as the synthesis of new conjugated polymers/oligomers [5,6] and interface engineering [7]. Charge transport and collection properties at both electrode interfaces are crucial factors for improving the efficiency of PSCs. Significant work has been undertaken to improve interfacial properties, such as alcohol/water soluble conjugated polymer electrolytes (CPEs) [8–17], non-conjugated polymers [18–23], alcohol/water soluble conjugated [24–27] or non-conjugated small molecules (SMs) [28–30], and polar solvent treatment [31–35]. These materials enable the fabrication of a multilayer device without destroying a pre-coated organic semi-conducting layer by orthogonal solubility in the processing solvents. PSCs with a thin film of these materials as an interlayer (IL) at

the cathode interfaces show dramatically improved performance, relative to the devices manufactured without these materials as an interfacial layer. It has been reported that the ionic functionalities, at the end of side chains on the conjugated polymer backbone, induce the formation of favorable interface dipole, which leads to a decrease in the work function of the cathode. Non-conjugated polymers or SMs [28–35] can also reduce the work function of the cathode due to the formation of an interface dipole. In comparison with polymeric materials, SMs have advantages such as ease of synthesis and purification. In addition, SMs do not have batch-to-batch variation, or broad molecular weight distribution.

In this research, we designed and synthesized non-conjugated SM organic electrolytes, which consist of polar quaternary ammonium bromide with the addition of multiple hydroxyl groups. The structure of SM electrolytes used in this research, N,N,N,N,N-hexakis(2-hydroxyethyl)butane-1,4-diaminium bromide (**C4**) and N,N,N,N,N-hexakis(2-hydroxyethyl)hexane-1,6-diaminium bromide (**C6**), are shown in Fig. 1. In order to observe the effect of hydrophobicity of the on the interfacial properties, we introduced different lengths of alkyl chains in between **C4** and **C6**. The thin layer of SMs in this research generated a favorable interface dipole with the quaternary ammonium bromide. In addition, the magnitude of the interface dipole was increased by the multiple polar hydroxyl groups at the end of **C4** and **C6**. Inverted PSCs (iPSCs), with

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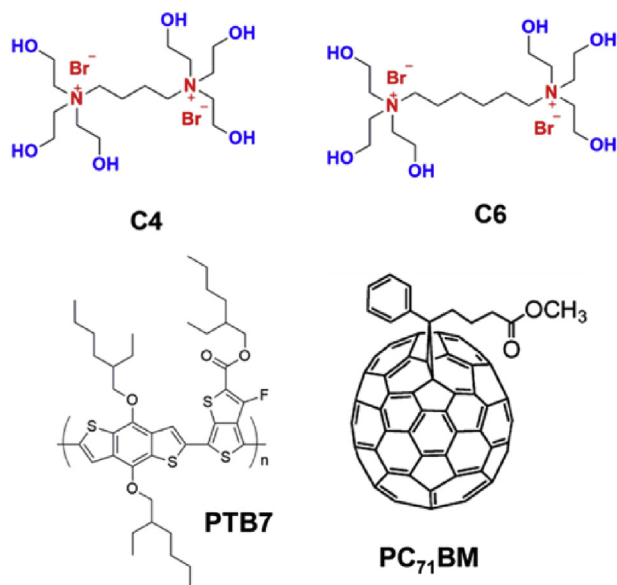


Fig. 1. Chemical structure of **C4**, **C6**, and materials in this research.

a structure of ITO/ZnO/**C4** or **C6**/PTB7:PC₇₁BM(1:1.5)MoO₃/Ag, were fabricated to investigate the effect of the IL on photovoltaic properties. A thin layer of **C4** and **C6** as the IL between the ZnO layer and the active layer in the iPSC dramatically improved the short circuit current density (J_{sc}), and the fill factor (FF), resulting in a PCE improvement from 7.41% to 9.20%, which is a 24.2% relative enhancement. The open circuit voltage (V_{oc}) data of the device with the ILs are almost same as the data of the reference device. Enhancements of the PCE of the devices were primarily due to the improvement of the J_{sc} . The ILs facilitate charge extraction from the

active layer to the cathode due to their favorable interface dipole, which reduces the energy barrier at the cathode interface, i.e., transition from a Schottky to an Ohmic contact.

2. Results and discussion

To investigate the effect of ILs on photovoltaic properties, iPSCs with the structure ITO/ZnO or IL/PTB7:PC₇₁BM/MoO₃/Ag (as shown in Fig. 2 (a)) were fabricated. The current density vs. voltage curves of the iPSCs under AM 1.5G simulated illumination with an intensity of 100 mW/cm² and in the dark are shown in Fig. 2 (c). The device performances are summarized in Table 1. The V_{oc} and FF of the device based on ITO/**C4** and ITO/**C6** were smaller than the device based on ITO/ZnO while the J_{sc} of the devices based on ITO/**C4** and ITO/**C6** increased slightly from 14.65 mA/cm² to 14.95 and 15.33 mA/cm², respectively. Thus, the PCE of the devices based on ITO/**C4** and ITO/**C6** are 6.03 and 6.65%, respectively, which are lower than the device based on ITO/ZnO (7.35%).

Kelvin probe microscopy (KPM) measurements were performed to investigate the effective work function of IL coated ITO and the ITO/ZnO surface and to prove the formation of an interface dipole at the surface of the substrate. The work function of **C4** and **C6** coated ITO with IL decreased from 5.0 eV (the work function of ITO) to 4.76 and 4.86 eV, respectively. These values are greater than that of ITO/ZnO (4.4 eV). The energy barrier height between the work function of IL coated ITO and the LUMO level of PC₇₁BM is larger than that of ITO/ZnO (as illustrated in Fig. 2 (b)). Thus, the performance of devices based in IL coated ITO are lower than those of devices based on ITO/ZnO. The work function data from the KPM measurements support the findings that the performance of a device with IL is lower than that of a device based on ITO/ZnO. The series resistance (R_s) values of the devices based on IL coated ITO are lower than that of devices based on ITO/ZnO. The leakage current (inset of Fig. 2 (c))

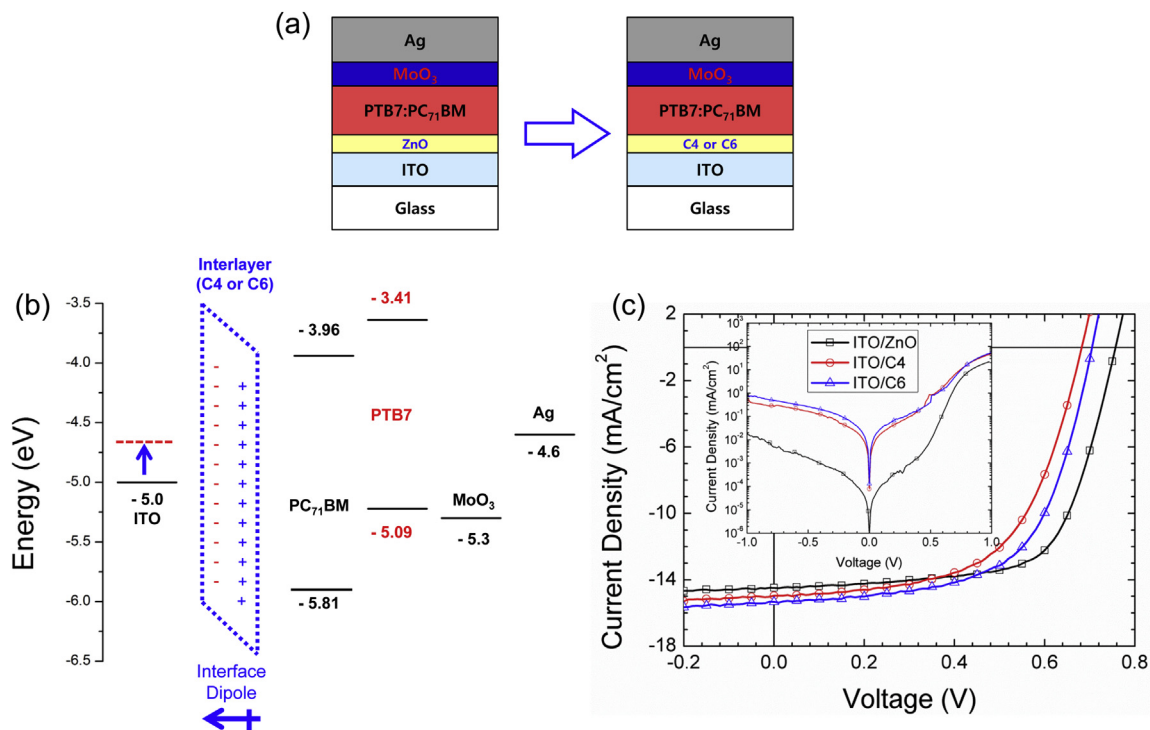


Fig. 2. Schematic illustration of (a) the device structures based on ITO/ZnO and ITO/**C4** or **C6**, (b) the energy level diagram of materials in this research, and (c) current density–voltage curves of iPSCs under AM 1.5G simulated illumination with an intensity of 100 mW/cm² (inset: in the dark condition; square: with ITO/ZnO; circle: with ITO/**C4**; triangle: with ITO/**C6**).

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