

# Switchable polarity in polymer solar cells using conjugated polyelectrolyte



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## ABSTRACT

We report polarity-switchable polymer solar cells that use poly [(9,9-bis((6'-(*N,N,N*-trimethylammonium)hexyl)-2,7-fluorene)-alt-(9,9-bis(2-(2-(2-methoxy-ethoxy)ethyl)-9-fluorene))dibromide polyelectrolyte (WPF-6-oxy-F). By introducing WPF-6-oxy-F as a polarity-controlling layer, we selectively achieved polarity switching in the device operation. When we deposited the WPF-6-oxy-F film on the top of an active polymer layer, the device operated conventionally; holes moved to the transparent indium tin oxide (ITO) electrode. However, the device showed switched polarity when we changed the position of insertion of the WPF-6-oxy-F film to the top of the transparent ITO electrode. Then, the electrons moved to the transparent ITO electrode, leading to an inverted device.

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## 1. Introduction

Plastic solar cells (PSCs) made from semiconducting and metallic polymers have a number of potential advantages, including lightweight, flexibility, and fabrication by printing/coating methods that enable low-cost manufacturing [1–3]. Although the device performances have steadily improved, further improvements in efficiency are required for large-scale commercialization [3,4]. Since conventional PSCs are based on a sandwich structure that uses a metal–insulator–metal (MIM) configuration [5–7], the device performances are very sensitive to the electrical properties of the metal electrodes [8,9]. Specifically, the built-in fields (BIFs) arising from the Fermi-level difference between the anode metal and the cathode metal in the PSCs plays an important role in charge collection [10]. The BIFs accelerate free charges separated at the interfaces of the electron donor and the electron acceptor, to the electrodes, and consequently, it directly affects the internal quantum efficiency (IQE) of the devices [2]. In addition, since the positive charges must move to the electrode with the higher work function (WF) because of the direction of the applied field, the BIF determines the polarity of the device [8].

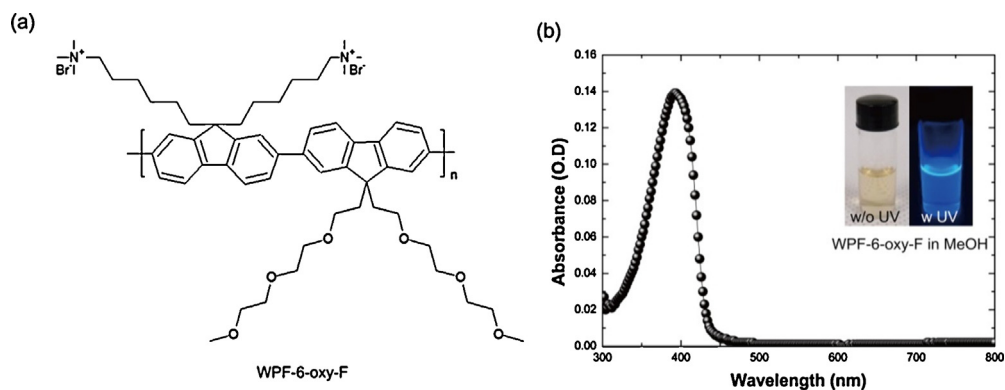
Controlling the device polarity provides several useful advantages in device fabrication. It enables the conversion of the device

structure from a conventional structure consisting of the transparent electrode as the anode to an inverted structure with the transparent electrode as the cathode, which is reversible. While the conventional device shows higher device efficiency, the inverted device leads to an enhancement in the device stability [11,12]. Moreover, it is possible to select better transport channels for electrons and holes in anisotropic bulk heterojunctions (BHJ) of an electron-donating conjugated polymer and electron-accepting fullerene derivatives in PSCs by controlling the polarity [13,14]. In principle, one can simply alter an electrode to modulate the BIF properties. However, it is difficult to secure suitable electrode materials. Most of the transparent electrodes used as anodes, such as indium tin oxide (ITO), indium zinc oxide (IZO), and aluminum-doped zinc oxide (AZO) show almost the same WF values of 4.7–4.8 eV, and low-WF metals used as cathodes such as Ca, Ba, and Li are unstable in air and H<sub>2</sub>O [15,16]. Therefore, it is crucial to develop alternative approaches to BIF modification without replacing the electrode materials. In this work, we report a simple, printable, and efficient approach to modify the BIF of the device without changing the metal, leading to polarity variation of the device using the water-soluble poly[(9,9-bis((6'-(*N,N,N*-trimethylammonium)hexyl)-2,7-fluorene)-alt-(9,9-bis(2-(2-(2-methoxy-ethoxy)ethyl)-9-fluorene))dibromide polyelectrolyte (WPF-6-oxy-F) as the polarity-controlling layer.

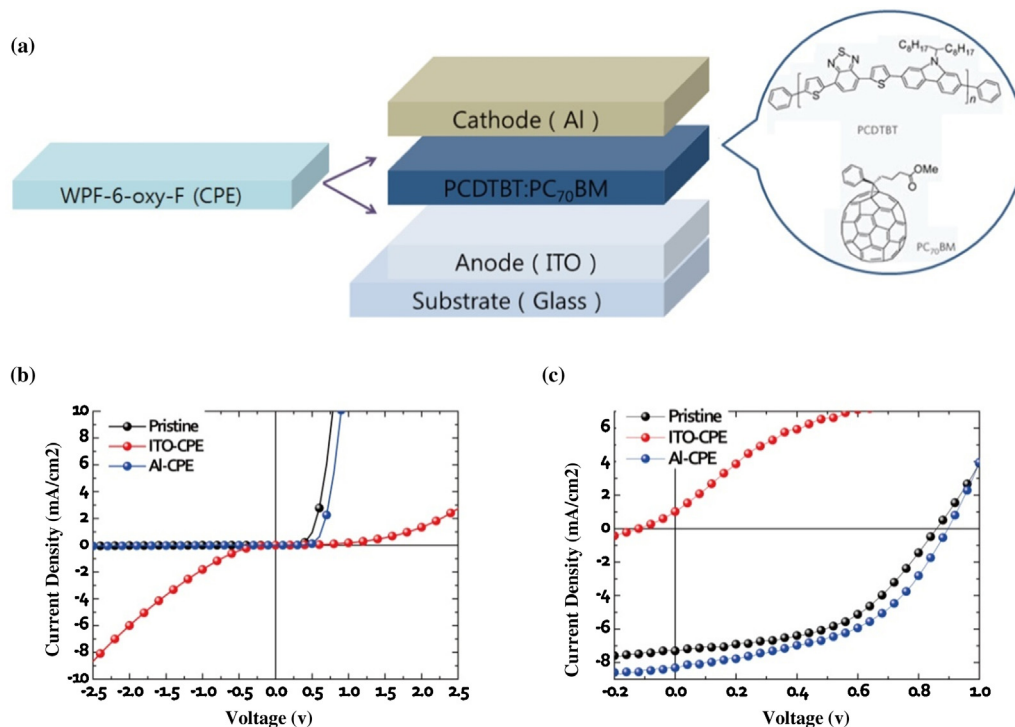
Recently, several materials have been investigated for modifying the interface between the polymer and the metal electrode [17–21]. In particular, conjugated polyelectrolytes (CPEs) with ionic

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**Fig. 1.** Molecular structure (a) and absorption spectrum (b) of poly [(9,9-bis((6-(N,N,N-trimethylammonium)hexyl)-2,7-fluorene)-alt-(9,9-bis(2-(2-methoxyethoxy)ethoxy)ethyl)-9-fluorene)]dibromide polyelectrolyte (WPF-6-oxy-F).



**Fig. 2.** Device structure (a), current–voltage characteristics under dark condition (b) and under AM1.5G irradiation condition (c). (For interpretation of the references to color in text, the reader is referred to the web version of this article.)

side groups have been successfully used as an interfacial layer in PSCs. The overall efficiencies, including the open-circuit voltage ( $V_{oc}$ ) [22,23], short-circuit current ( $J_{sc}$ ), and the fill factor (FF), have been enhanced by the introduction of the CPE interfacial layer. Because the CPE can induce interfacial dipoles because of its ionic group, the BIF in the device can be expected to be modified, leading to the switching of the polarity. Here, we have successfully demonstrated polarity-switchable PSCs by using the water-soluble WPF-6-oxy-F CPE as the polarity-controlling layer. By introducing the WPF-6-oxy-F into the conventional PSC with the structure of ITO electrode/BHJ layer/Al, we selectively achieved polarity switching during device operation. When we deposited the WPF-6-oxy-F film on top of the BHJ active layer, the device operated conventionally and the holes moved to the transparent ITO electrode. However, the device switched polarity when we cast the WPF-6-oxy-F film on top of the transparent ITO electrode and the electrons moved to the transparent ITO electrode.

## 2. Results and discussion

Fig. 1 shows the molecular structure and the optical properties of WPF-6-oxy-F. The WPF-6-oxy-F CPE was synthesized in our group. Although the backbone structure of the WPF-6-oxy-F is identical to that of blue-emitting polyfluorene (PF) [20,21,24], WPF-6-oxy-F consists of unique ionic or polar side groups that can induce interfacial dipoles, as shown in Fig. 1(a). WPF-6-oxy-F has several advantages as an interfacial layer material. WPF-6-oxy-F exhibits a relatively large Stokes shift: it emits blue color during photoluminescence (PL) after absorbing ultraviolet (UV) (see the Fig. 1(b)) [25–27]. Therefore, WPF-6-oxy-F can lead to further enhancement in the absorption of the active layer with absorbance in the visible range. Further, because WPF-6-oxy-F is soluble in water, it can prevent damage of the underlying organic soluble active layer during film casting. This enables the fabrication of a multilayer device [28–30]. In addition, since electrons and holes are delocalized along

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