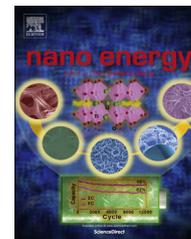




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FULL PAPER

# In-depth considerations for better polyelectrolytes as interfacial materials in polymer solar cells



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## Abstract

We perform a comprehensive study to achieve better polyelectrolytes (PEs) as electron-transport layers (ETLs) in polymer solar cells (PSCs). Three well-known PEs - PFN, PEIE, and WPF - are chosen as model systems and investigated with variations in their backbone structures and the state of the amine functionalities on their side chains. Respectively optimized PSCs using the three PEs exhibit different cell-performances, mainly owing to the diode characteristics of built-in potential and recombination strength. To identify how such deviated device-performances correlate with the structural features of PEs, the modulated interfaces of ITO/PEs and PEs/active layer are studied in detail. It is found that conjugated backbones and larger counter-anions on side chains can promote the modulation of ITO work functions (WFs) and that a large amount of protonated amines on PEs is beneficial for junction properties with a subsequent active layer. Additionally, our results indicate that interfacial dipole and electrical doping between the PE and active layer, in addition to WF modulation of the ITO cathode, are

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important for device efficiency. Accordingly, with the aid of the molecular features of PEIE, PEIE-PSCs exhibit excellent device efficiency and stability compared with PFN- and WPF-PSCs. In the PTB7-th:PC<sub>71</sub>BM system, a remarkable power-conversion efficiency of 9.97% is achieved with a single PEIE ETL.

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## Introduction

With the advent of next-generation energy sources, polymer solar cells (PSCs) have long maintained the top position among third-generation photovoltaics because of their unique properties, such as simple fabrication, light weight, and availability in flexible formats [1-5]. Presently, PSCs overcome the 10% hurdle of power conversion efficiency (PCE) even in single-junction devices owing to considerable efforts in the development of light-harvesting materials, the evolution of efficient device-structures, novel processing techniques, and the appropriate incorporation of interfacial materials [6-10]. Despite this encouraging progress, the commercialization of PSCs faces several challenges including long-term stability and compatibility with printing manufacturing for large-scale production. In light of this, the inverted structure has begun to replace the normal form of PSCs because of its superior ambient stability and compatibility with R2R processing using air-stable and printable metal electrodes (Ag and Au), along with its more efficient performances [4,11]. Also, the record-high PCEs of recent PSCs have been driven by the inverted structure [10,12,13].

For successful structural modification from normal to inverted systems, the interfacial characteristics between the active layer and respective electrodes must be controlled, unlike the case for existing interfacial modulations in the normal structure. Thus, considerable attention is directed to electron-transport materials allowing bottom-electrodes (generally indium tin oxide (ITO)) to serve as electron collectors. For electron-transport layers (ETLs), various materials have been investigated and proven effective for modulating the interfacial properties of the cathode and inducing a reliable rectifying characteristics of PSCs [9,14]. Among these, alcohol/water soluble polyelectrolytes (PEs) as ETLs have attracted great attention because PEs are easily able to control the work-function (WF) of the underlying electrode with simple and scalable solution-processing using eco-friendly solvents [14-17]. Additionally, PEs are versatile as their chemical structure can be modified. These PEs are generally featured by polar or ionic moiety, which renders them soluble to hydrophilic solvents such as water and alcohol and facilitates multilayer stacking with solution-processing. Also, the polar groups on PEs can induce interfacial dipoles to modify the WFs of adjacent electrodes and reduce the energy-barrier for efficient charge-extraction, which is essential for high-performing PSCs.

After poly(ethylene oxide) (PEO) emerged as an early version of a PE ETL [18], a structure based on a conjugated backbone with polar pendant groups has been the focus of intensive research toward better PE ETLs in PSCs [14]. Regarding conjugated PEs, because the polar moiety plays

a determining role in the formation and the strength of the interfacial dipole, a wide variety of strategies focusing on their side chains and ionic groups have been explored: the modifications in the length of the side chains, the number of ionic side chains, and the type of counter-ions and ionic functionalities (cationic, anionic and zwitterionic forms) [19-23]. On the other hand, the main backbones of conjugated PEs have been varied with polyfluorene, polycarbazole, and polythiophene derivatives for better interfacial characteristics because the charge-transport, energy band-gaps, thin-film morphology, and surface energy can be affected by the structures of the main backbones [24,25].

Using similar strategies, we previously introduced ammonium salts as cationic side chains with ethylene oxides onto polyfluorene chains (WPF) to induce large interfacial dipoles and tune the WF of an ITO electrode, yielding a high performance of inverted PSCs incorporating WPF as an ETL, along with favorable rectifying characteristics [15,26]. Regarding the polyfluorene structure, He et al. reported the conjugated PE (poly [(9,9-bis(3'-(N,N-dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)] (PFN)) and the encouraging performance of inverted PSCs with a 9.2% PCE [4]. Along with these conjugated frames, attention toward PE ETLs was extended to non-conjugated systems after the recent introduction of polyethylenimine ethoxylated (PEIE) as an ETL in PSCs. Advantages of PEIE include commercial availability, low price, and excellent compatibility with various organic devices owing to its high content of amine groups [17].

Thus far, both PFN and PEIE representing PEs of conjugated and non-conjugated frames have yielded high-performing PSCs, but it remains to be determined which PE frame is more effective as an ETL in the fabrication of PSCs. In addition, the fundamental mechanism for performance improvement of PSCs using PE ETLs is inconclusive, and concrete rules for the chemical design of PEs for an ideal ETL have not been outlined. To improve PEs as an ETL material, a clear understanding of the origins for their function as an ETL, and studies on the correlation between chemical structures and their performance are indispensable. Currently, numerous papers have attributed the ETL functions of PEs mainly to the formation of permanent dipoles on electrodes, which causes WF shifts [4,19,22]. Furthermore, the focus of the chemical design to control the strength of permanent dipoles differs among studies; for example, some place importance on the structure of the main backbones and others place importance on ionic side chains including counter-ions [14,25]. On the other hand, there are several reports indicating that the doping of organic semiconductors adjacent to PEs could be another main factor causing enhanced electron-transport in PSCs using PE ETLs. This is because of the anion-induced electron

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