



Factors affecting the photovoltaic behavior of inverted polymer solar cells using various indium tin oxide electrodes modified by amines with simple chemical structures



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ABSTRACT

In a glass–indium tin oxide (ITO)/amine/regioregular poly(3-hexylthiophene) (P3HT):[6,6]-phenyl C₆₁ butyric acid methyl ester (PCBM)/poly(3,4-ethylenedioxylenethiophene):poly(4-styrene sulfonic acid) (PEDOT:PSS)/Au cell, which uses small molecule amine-modified ITO as the electron collection electrode, a light-soaking effect under irradiation of simulated sunlight was restrained considerably compared with in an ITO/P3HT:PCBM/PEDOT:PSS/Au cell containing bare ITO. That is, the time taken to arrive at a saturated V_{oc} from the initial V_{oc} became short when the ionization potential (I_p) of ITO reduced by the amine modification, and consequently both of its saturated V_{oc} and power conversion efficiency (PCE) improved. The I_p decreased with an increase in the number (N) of amino groups in a single amine molecule, because the basic amino groups can efficiently neutralize any acidic hydroxyl groups on ITO through a multipoint interaction. The superior performance of the cell containing the amine-modified electrode with large N was perhaps because the energy mismatch formed by a contact between ITO and acceptor PCBM reduced, and consequently the rate of electron collection at ITO increased.

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1. Introduction

Organic thin film solar cells have attracted considerable research attention as next-generation solar cells because of their light weight, flexibility, and low cost [1–8]. Conventional cells have usually been fabricated in a glove box filled with an inert gas such as N₂, and their performances have been evaluated in inert atmospheres or by encapsulating them [9–13]. Recently, many researchers have investigated inverted organic solar cells because of their stability in air [14–21]; specifically, we have developed polymer solar cells of this type that can be fabricated in air using air-stable materials such as gold and PEDOT:PSS in the hole collection sites along with titanium oxide and zinc oxide in the electron collection sites [22–28]. The metal oxides were chosen from the viewpoint of reducing the energy mismatch formed by a contact between transparent ITO electrode and acceptor PCBM, and promoting efficient electron collection.

Nüesch et al. investigated ITO that had been treated with acids and bases by ultraviolet and X-ray photoelectron spectroscopies (UPS and XPS) [29], observing both monolayer adsorption and a work function

shift. They reported that treatment by a base such as tetrabutyl ammonium hydroxide reduced the work function by 0.9 eV when compared with untreated ITO. Zhou et al. reported a universal method to produce low work function electrodes for organic electronics [30], i.e., they proposed electrodes that were modified by polymers containing simple aliphatic amine groups, such as polyethylenimine ethoxylated (PEIE) and branched polyethylenimine (PEI), as substitutions for low work function metals. Because these amine-modified electrodes can easily be prepared in air at room temperature, they have recently been used in inverted cells as electron collection electrodes [31–33]. Chen et al. reported an interesting work where amine-based fullerene derivatives formed a self-organized monolayer on ITO that was anchored by the amine parts, and this fullerene-modified ITO enhanced the electron collection layer function in the inverted polymer solar cells [34,35].

Because there are many commercially-available amine compounds with simple chemical structures, we have systematically investigated their use in inverted polymer solar cells using various amine-modified ITOs, while paying particular attention to the number (N) of amino groups in a single amine molecule. The ionization potential (I_p) of ITO effectively decreased by modifying ITO with small molecule amines of larger N, and better performance was obtained for the cell using such a modified ITO electrode, although a light-soaking effect was observed to a degree. The reasons behind the interesting results that were obtained in this work are discussed in detail in this paper. We believe that the

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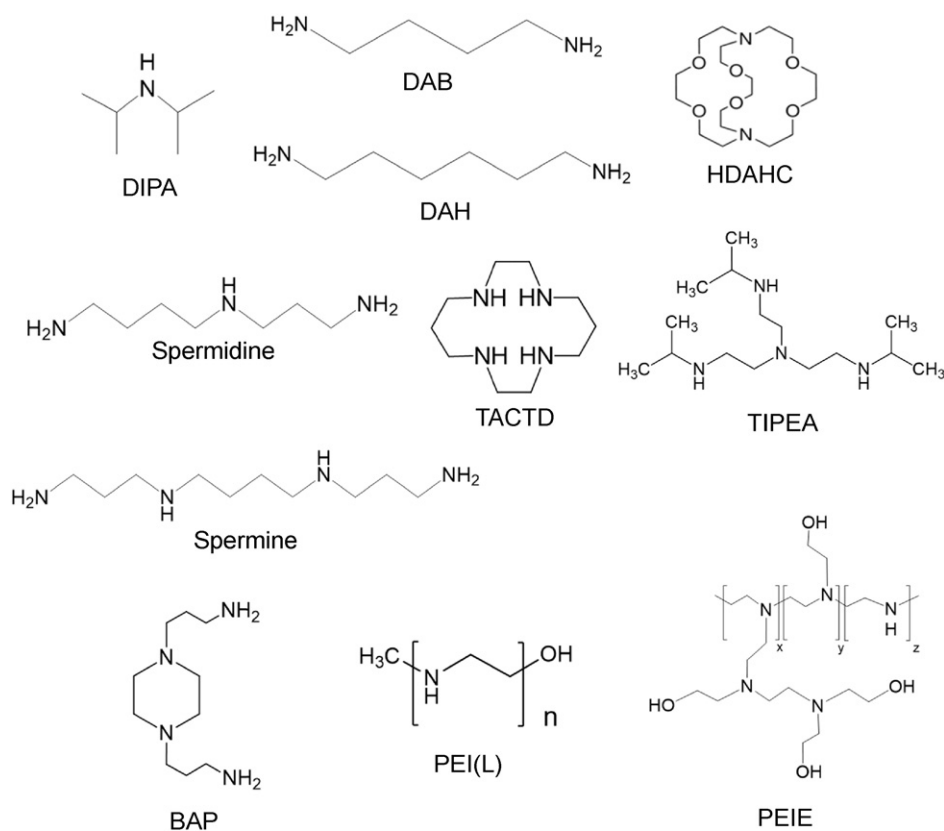


Fig. 1. Chemical structures of amines used to modify the ITO surface and the associated abbreviations.

results of this study contribute to the technological advancement of inverted organic solar cells.

2. Experimental

2.1. Materials

4,7,13,16,21,24-Hexaoxa-1,10-diazabicyclo [8.8.8] hexacosane (HDAHC), linear polyethylenimine (PEI(L)), tris[2-(isopropylamino)ethyl]amine (TIPEA), 1,4-bis(3-aminopropyl)piperazine (BAP), polyethylenimine 80% ethoxylated solution (PEIE), 1,4,8,11-tetraazacyclotetradecane (TACTD), spermine, regioregular P3HT (Mw 87,000), 2-methoxyethanol, *o*-dichlorobenzene (*o*-DCB), and Triton-X 100 were purchased from Sigma-Aldrich Chemical Co., Inc. (Japan). Diisopropylamine (DIPA), 1,4-diaminobutane (DAB) and 1,6-diaminohexane (DAH) were purchased from Tokyo Chemical Industry Co., Ltd. Ethanol and methanol were purchased from Kanto Chemical Co., Inc. Spermidine was purchased from Wako Pure Chemical Industries, Ltd. PCBM was purchased from Frontier Carbon Corporation. A PEDOT:PSS (Clevios® P) 1.3 wt.% dispersion in water was purchased from H. C. Starck GmbH. All chemicals were used as received. The glass-ITO substrates (sheet resistance = 10 Ω/sq.) and Au wires were purchased from the Furuuchi Chemical Corporation. Fig. 1 shows the chemical structures of the amines that were used in this work and the associated abbreviations.

2.2. Preparation of amine-modified ITO

The glass-ITO electrode was cleaned by sonication in 2-propanol, washed in boiling 2-propanol, and then dried in air. The ITO substrate was covered using a masking tape (“E-MASK® TP200” protective film, Nitto Denko Corp.) to prevent any additional modification of the amines. To prepare the amine-modified ITOs, we used water as the solvent for DIPA, DAB, DAH, spermidine, HDAHC and spermine, ethanol as

the solvent for TIPEA and BAP, 2-methoxyethanol as the solvent for PEIE and PEI(L), and methanol as the solvent for TACTD. The amine solution was dropped on the ITO substrate and was then spin-coated at 5000 rpm for 1 min. When alcohols and water were used as the solvent, the substrates were dried at 120 and 150 °C for 10 min on a hotplate, respectively. Additionally, to remove any extra amines from the ITO, the same operation was performed using a solvent instead of the amine solution. These experimental operations were carried out in air.

2.3. Fabrication of inverted polymer solar cells

An *o*-DCB solution containing 16.7 mg mL⁻¹ of P3HT and 13.3 mg mL⁻¹ of PCBM (weight ratio = 5:4) was spin-coated on the amine-modified ITO substrate. Subsequently, a PEDOT:PSS dispersion in water containing 0.5 wt.% of Triton-X 100 as a surfactant was spin-coated on the hydrophobic P3HT:PCBM layer, and then the PEDOT:PSS film formed on the P3HT:PCBM was dried at room temperature for

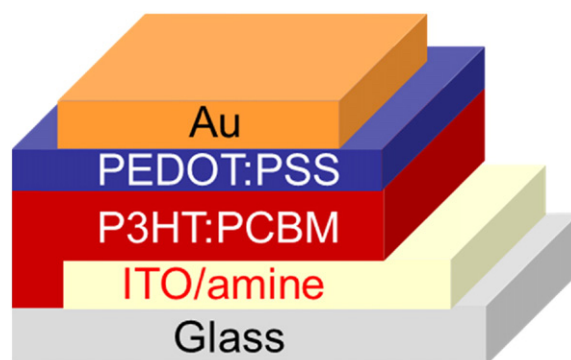


Fig. 2. Schematic structure of an inverted polymer solar cell with an amine-modified ITO electrode.

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