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Work function optimization of vacuum free top-electrode by PEDOT:PSS/PEI interaction for efficient semi-transparent perovskite solar cells

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ABSTRACT

Our research group fabricated low temperature based inverted semi-transparent $CH_3NH_3PbI_3$ perovskite photovoltaics. The work function of PEDOT:PSS (PH1000) electrode was modified using polyethylenimine (PEI) and both layers are transferred on the ITO/PEDOT:PSS/$CH_3NH_3PbI_3$/PCBM layer using hydrophobic polyurethane acrylate (PUA) polymer stamp. Even if the PH1000 spin coating on the $CH_3NH_3PbI_3$/PCBM layer shows degradation of perovskite due to the inter-diffusion of PH1000 solvent, drying transfer shows effective layer stacking without degradation. The PEI treatment on the PH1000 contributes to reduce the work function from 5.1eV to 3.97 eV and this modification makes to improve the device performance from 0.07% to 4.02%. The full device average visible transmittance (AVT) exhibits 24.53% and the work function modification shows the decreased charge transporting resistance (R_c), which directly correlate to the enhanced open circuit voltage (V_{oc}) short circuit current (J_{sc}) and fill factor (FF).

1. Introduction

In the recent research, perovskite photovoltaics showed tremendous power conversion efficiencies (PCE) and considered as the emerging photovoltaics due to the high performance, mechanical flexibility, low-cost fabrication and solution process [1–8]. Perovskite is a kind of material structure which composed of ABX_3 crystal structure [9]. As the perovskite structure has long exciton diffusion length [10], wide light absorption range [11], small energy band gap (Eg) [12], perovskite layer has been used as the active layer in photovoltaic device. Perovskite photovoltaics can be composed of two types device structure: conventional and inverted. In conventional perovskite structure, n-type electron transporting layer (ETL) is coated below the perovskite film and p-type hole transporting layer (HTL) is coated on the perovskite film (n-i-p type). On the other hand, in inverted perovskite structure, p-type HTL is coated below the perovskite film and n-type ETL is coated on the perovskite film (p-i-n type). In case of conventional perovskite photovoltaics, the highly stable metal oxide (e.g., TiOx) have been used as the ETL [13–15]. Even though these materials have effective ETL condition in photovoltaic device, the high thermal treatment (> 500 °C) is required to fabricate ETL, which is not suitable to scale up the photovoltaic device. However, in case of inverted perovskite device, the relatively low temperature (< 150 °C) process is possible to fabricate device using the highly transparent and conductive HTL (e.g., PEDOT:PSS), and conductive ETL (e.g., PCBM), which means that this structure is more suitable to apply into photovoltaics like wearable and flexible device which low temperature process is essential to fabricate.

In previous research, many types of transparent electrodes like thin metal electrode [16], metal nanowire electrode [17] and dielectric-metal-dielectric (DMD) electrode [18] have been applied into semi-transparent perovskite photovoltaics with various methods. In case of thin metal and DMD based electrode, researchers normally used thermal evaporation method to fabricate uniform transparent top electrode on semi-transparent photovoltaic device. Even though this process is essential to fabricate metallic based electrode, this process is not suitable to scale up the device. In previous studies, stamping transfer have considered as the alternative process to fabricate transparent electrodes in organic, perovskite photovoltaics [19–22]. These researchers used the polymethyl-methacrylate (PMMA) and polydimethylsiloxane (PDMS) as the polymer stamp. As the PMMA, PDMS stamp has swelling problem for organic solvents, PMMA, PDMS stamp has limitation to use repeatedly. In the recent research, our group

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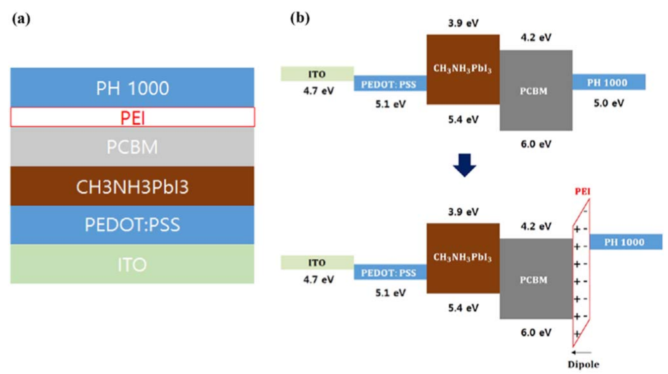

Fig. 1. (a) Schematic and (b) Energy diagram of semi-transparent perovskite photovoltaic device.

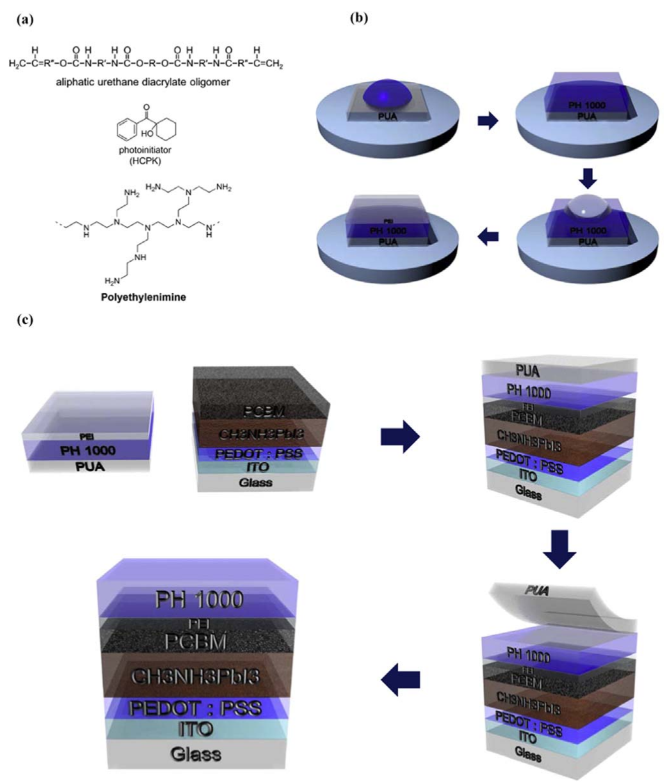

Fig. 2. (a) Chemical structures of PUA raw materials and PEI. (b) Schematic diagrams of work function modification of PH 1000 using PEI. (c) Schematic diagrams of PUA based stamping transfer.

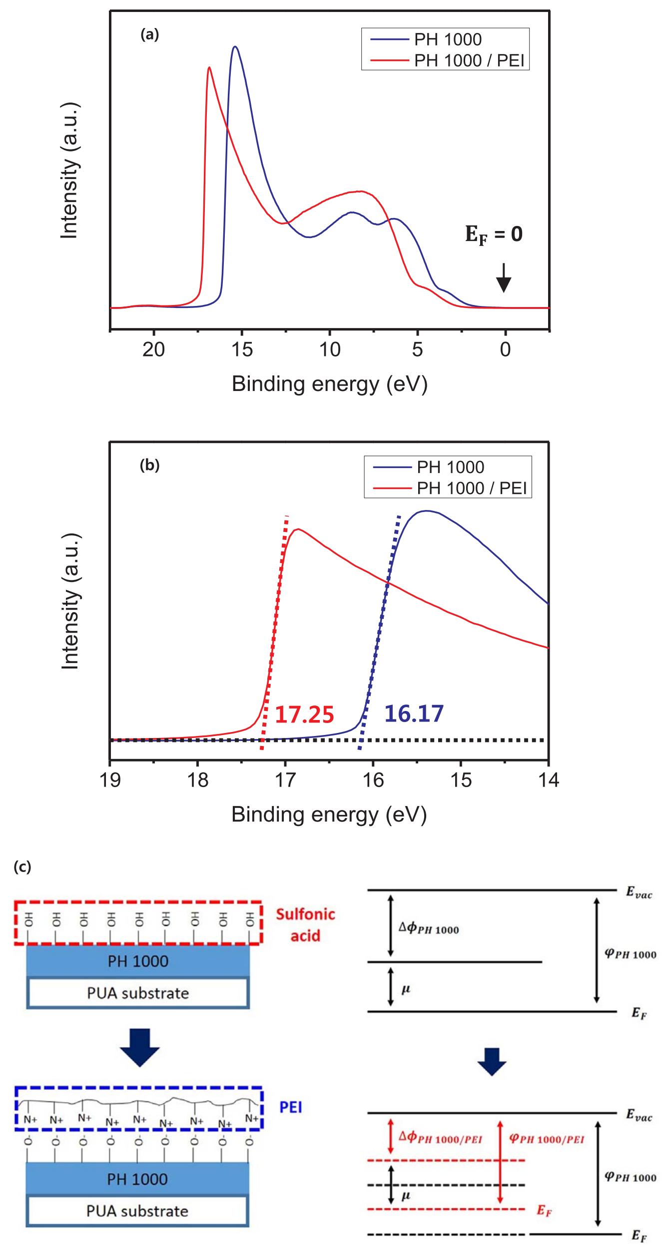

Fig. 3. UPS spectra of PH 1000 and PH 1000/PEI electrode with (a) binding energy under He I (21.22 eV) irradiation, (b) secondary cutoff region. (c) Work function modification mechanism of PH1000 using PEI.

suggested polyurethane acrylate (PUA) as the alternative polymer stamp [23]. In contrast to PMMA, PDMS polymer, PUA polymer shows more effective mechanical stability for organic solvents and have surface energy tuneable by modifying raw materials composition [24].

Here in, we fabricated low temperature based inverted semi-transparent perovskite photovoltaics via stamping transfer. Highly conductive and transparent PEDOT:PSS (PH1000) electrode used and the work function was modified through PEI layer coating on the PH1000 electrode. To transfer the hydrophilic PH1000 or PH1000/PEI layer, we fabricated hydrophobic PUA polymer to reduce the interface affinity between PH1000 and PUA stamp. From the stamping transfer, this layers are clearly transferred on the ITO/PEDOT:PSS/$CH_3NH_3PbI_3$/PCBM without degradation of perovskite layer, and this semi-transparent photovoltaics exhibits 24.53% average visible transmittance (AVT) and 0.07 ± 0.01% (w/o PEI treatment), 4.02 ± 0.09% (w/ PEI treatment) PCE respectively. The physical effects of PEI in photovoltaic device can be explained as the result of improved ohmic contact condition between PCBM ETL and PH1000 electrode, by analysing UPS, dark current, IPCE and nyquist plot.

2. Experimental

2.1. Fabrication of hydrophobic PUA

The PUA films were fabricated using a method described in a previous report, with some modifications [23]. To fabricate the hydrophobic PUA, 19 g oligomer and 2 g of HCPK were mixed with vigorous stirring at room temperature and were sonicated for 10 min. Then, the mixture was poured into a silicon wafer and was rubbed using a roller slightly after being exposed to ultraviolet (UV) light for 10 min.

2.2. Fabrication of perovskite photovoltaics

The devices were prepared on an indium tin oxide (ITO) patterned