Planar Perovskite Solar Cells Using Fullerene C70 as Electron Selective Transport Layer

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ABSTRACT - **Owing amongst other to its high electron mobility, fullerene C70, has been widely used as an electron transporting layer in organic solar cells. In this research, we report the use of C70 thin films as electron transport layers of planar perovskite solar cells (PSCs) using a conventional device structure. The thickness of the C70 layer has been optimized to achieve the best efficiency of 12%. It is demonstrated that ultra-thin C70 films can effectively block holes and thus become selective to the transport of electrons in PSC devices.**

KEYWORDS: Perovskite solar cell, CH3NH3PbI3, Electron transport layer (ETL), Fullerene, C70, Interface engineering

I. INTRODUCTION

Organometallic halide perovskite solar cells (PSCS) have recently attracted significant interest owing to their superior optoelectronic properties, such as long electron–hole diffusion lengths (>1μm) [1], high absorption coefficient, tunable band gaps, high charge carrier mobility and long charge carrier life span (>100 ns) [2-5], which mainly arise due to their specific crystalline structure [6]. The general structure of perovskite absorber materials is AMX3, where A is a large cation (typically CH3NH3+), M is a divalent metal cation (typically Pb2+) and X is a halogen (typically Cl, Br or I) [7, 8]. Miyasaka *et al*. presented the first perovskite solar cells in 2009 [9]. They used a liquid electrolyte as a hole transport material, resulting in the power conversion efficiency (PCE) of around 3-4%. Nowadays, within a relatively short time, the PCE of PSCs has reached around 23.3% [10] . Most high-efficiency PSCs have a mesoscopic structure, but these structures require high-temperature fabrication processes (up to 500°C), which increases manufacturing costs and limits printable and flexible fabrication [11, 12]. Recently, planar PSCs based conventional and inverted structures have attracted extensive attention because of their low-temperature processes, simple preparation and flexible manufacturing capacity [13-16]. The electron transport layer (ETL), however, is critical for planar perovskite solar cells. It is usually sandwiched between the perovskite absorber layer and the cathode to aid carrier separation and especially support efficient collection of electrons at the cathode [12]. Fullerenes and their derivatives are good candidates as ETL in planar PSCs. This is due to their relatively high electron mobility and efficient hole-blocking property [7, 17], arising from a low-lying HOMO level. Shao *et al*. showed that fullerene layers, such as PC61BM ETLs, can effectively passivate charge trap states at CH3NH3PbI3 perovskite film surfaces and grain boundaries, and from that reduce the hysteresis in perovskite devices

[18]. C70 is another fullerene derivative that have been utilized heavily in organic solar cell development [19-22] and potentially can be utilized as an efficient ETL in perovskite solar cells. As shown in Fig. 1(b), C70 has a Lowest Unoccupied Molecular Orbital (LUMO) at 4.2 eV [23], which provides an excellent ETL for CH3NH3PbI3 perovskite absorber-based PSCs with a LUMO level at 3.9 eV [24], although it should be noted that the exact alignment across the interface has not been measured. In addition, the hydrophobic surface of C70 compared to PCBM (water contact angle of 99.6° [25] vs. 86.5° [26]) can result in enhanced stability of PSCs in ambient atmosphere. This is potentially of high importance due to the multiple different degradation mechanisms that currently impact PSC technology [27, 28]. In this study, we introduce for the first time a C70 thin film deposited (at substrate room temperature) to serve as an electron transport layer for planar perovskite solar cells with a conventional device structure. The significant quenching intensity of the Photoluminescence (PL) measurement showed that a C70 layer can be an excellent electron extraction and efficient hole blocking layer for PSCs. Due to the thickness optimization of C70 and the high quality of the perovskite layer, the best efficiency of 12% was obtained for these devices.

II. EXPERIMENTAL

A. *Materials and device fabrication*

Pre-patterned ITO-coated glass substrates (Kintec Company, Hong Kong) were used for a PSC area of 10 mm2. The sheet resistance of the ITO was approximately 15 Ω/sq. The substrates were cleaned sequentially in an ultrasonic water bath with a detergent, deionized water, acetone and IPA (10 min for each). Then, they were blow dried with a nitrogen gun. C70 (Sigma-Aldrich) layers with the thicknesses of X= 7.5, 10, 20, 30 and 40 nm were deposited at a growth rate of 0.2 Å/s through Organic Molecular Beam Deposition (OMBD) at a base pressure of 3×10^{-8} mbar. A PCBM precursor solution was prepared through dissolving 20 mg PCBM in 1 mL of Chlorobenzene (Sigma-Aldrich) and then spin-coated at 1500 rpm for 60 seconds. CH3NH3PbI3 perovskite films were deposited by a one-step method and with antisolvent treatment; 461 mg of PbI2 (Alfa Aesar, 99.99%), 159 mg of CH3NH3I (Sigma-Aldrich) and 70.9 µl of DMSO (Sigma-Aldrich) were mixed in 635 µl of N,N-dimethylformamide (DMF) (Sigma-Aldrich) at 70 oC. The mixture was stirred for an hour until a completely dissolved solution was obtained. This solution was then spin-coated on the ETL at 4000 rpm for 35 seconds, and 70 µl of chlorobenzene was slowly dripped on the rotating substrate in 10 seconds before the surface became turbid due to the rapid vaporization of DMF. The transparent film was heated at 100oC for three minutes in order to obtain a dense CH3NH3PbI3 film. For the HTL, a Spiro-OMeTAD precursor solution was prepared through dissolving 72.5 mg of Spiro-OMeTAD (Sigma-Aldrich), 28.5 µL of 4-tert-butylpyridine (Sigma-Aldrich) and 17.5 µL of a lithium bis(trifluoromethanesulfonyl) imide (Sigma-Aldrich) solution (520mg/mL in acetonitrile) in 1 mL of chlorobenzene. This hole transport solution was spin-coated at 4000 rpm for 30 seconds. Finally, 80 nm of an Au electrode layer was deposited by the E-beam evaporation system through a shadow mask.

B. *Film and device characterization*

All the characterizations were performed in an ambient environment. The current density-voltage (J-V) characteristics of the PSCs were measured using a 2400 source measure unit (Keithley Instruments Inc., USA) and a class AAA solar simulator (Sun 3000, Abet Technologies Inc., USA). The J-V characteristics were measured through applying a voltage sweep from +1.5 to −0.5 V under a calibrated lamp intensity of 100 mW/cm2. The PL intensity measurements of the structure ITO/C70 (0 and 10 nm)/Perovskite were performed through a microscope objective (Nikon E Plan 50X 0.75 EPL) with a fluorescence microscope (Nikon Eclipse ME600) connected to a Maya2000Pro spectrometer (from Ocean optics). The spectrometer had a mercury short-arc lamp with a filtered excitation wavelength of 330–

380 nm serving as a source of light. Finally, the SEM image of the perovskite layer was taken with a scanning cold field emission electron microscope (Hitachi S-4800 SEM).

III. RESULTS AND DISCUSSION

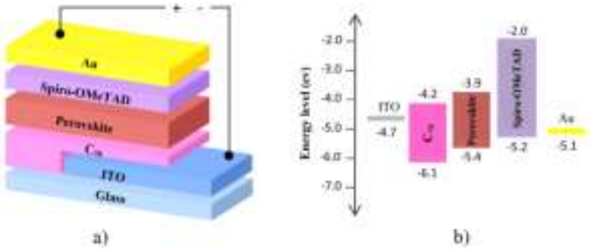

Fig. **1** a) Planar PSC device architecture with C70 as ETL, b) Energy level diagram of PSC with a conventional device configuration.

Figure 1(a) illustrates the device configuration used in this study. The structure of the planar PSC device consists of a compact C70 layer as ETL, a CH3NH3PbI3 perovskite absorber film and a spiro-OMeTAD film as hole transport layer (HTL). Figure 1(b) shows the energy levels of each layer in the device.

To have high-efficiency PSCs, the quality of perovskite films should be given utmost importance. As demonstrated in numerous studies[29], it is challenging to control the formation process of high-quality perovskite films that possess a high surface coverage with no pinholes on planar substrates. Different crystallization strategies, such as antisolvent engineering and fast deposition–crystallization, can affect the crystalline domains, and thus also the performance of the resulting solar cells [29-31]. In this study, a high-quality perovskite film was formed by spin-coating of a solution containing equimolar MAI, PbI2, DMF and DMSO. Also, chlorobenzene was used to remove solvent DMF [32]. As the scanning electron microscopy (SEM) image in Fig. 2 confirms, a high-quality CH3NH3PbI3 perovskite film was prepared on C70-coated ITO substrates by the one-step solution method using an antisolvent (chlorobenzene) treatment. The figure also shows the dense and smooth surface of the perovskite film with large crystalline grains.

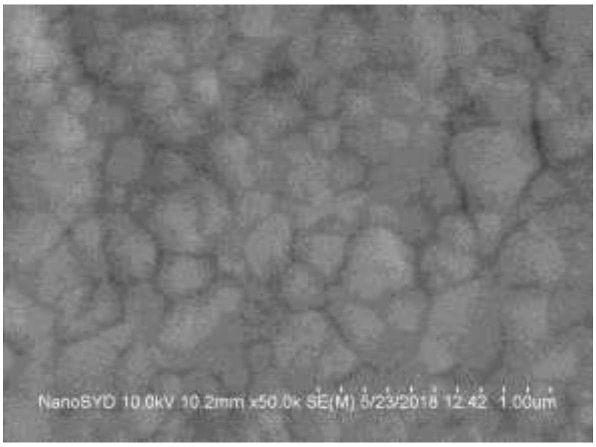

Fig. **2.** Top view SEM image of a perovskite film prepared on ITO substrate covered with a 10 nm C70 film.

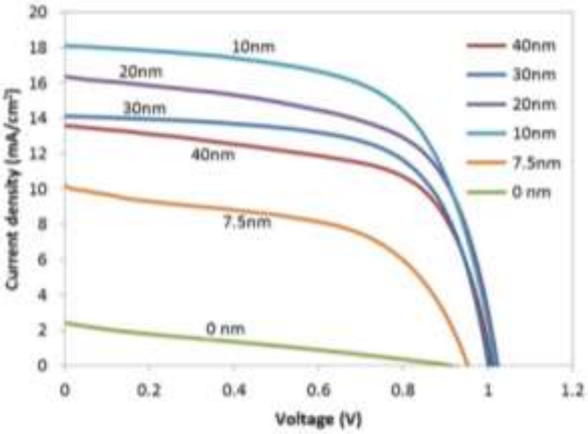

Fig. **3.** The JV characteristics of the representative J-V curves for the PSC devices with various thicknesses of the C70 layer as ETL.

Due to the limited electron mobility of organic ETLs and the absorption of C70, the thickness of C70 films in PSC devices should be optimized. The JV characteristics and the performance parameters of PSC devices with different thicknesses of C70 as an ETL are shown in Fig. 3 and Table 1 respectively. A summary of the performance parameters reported in Table 1 is plotted in Fig. 4. As it can be seen in the table, a device without a C70 interface layer can only achieve a PCE of 0.56% with a VOC of 0.89 eV, JSC of 2.42 mA cm-1 and very low FF about 25.97%. It is well-known that a PSC device without an electron transport and hole blocking layer exhibits severe charge recombination effects and shunts at the ITO/perovskite interface [3]. After an increase in the C70 thickness, no major change was observed in the open-circuit voltage (VOC) (Fig. 4), whereas the JSC first increased and then decreased. The optimized thickness of the C70 interface layer is about 10 nm (with the average PCE of about 11.52%). If the thickness of the C70 layer is increased above 10 nm, both the FF and JSC decreases.

Table 1. Performance parameters of the PSC devices with various thicknesses of the C70 layer as ETL. All the performance data were obtained from the average values of in total ten PSC devices for each thickness, where the error bars represent the standard deviation.

Thickness of C70	VOC (V)	JSC (mA/cm^2)	FF (%)	PCE (%)
0 nm	0.89±0.04	2.42±0.40	25.97±3.15	0.56±0.23
7.5 nm	0.95±0.03	10.21±0.36	53.82±3.19	5.22±0.44
10 nm	1.03±0.01	18.10±0.45	61.82±1.63	11.52±0.50
20 nm	1.02±0.01	16.16±0.33	62.60±1.44	10.31±0.31
30 nm	1.04±0.04	14.12±1.13	63.27±4.07	9.29±0.22
40 nm	1.04±0.05	14.02±0.22	58.71±3.34	8.54±0.16
PC61BM	0.95±0.04	19.69±0.45	57.43±2.20	10.74±0.30

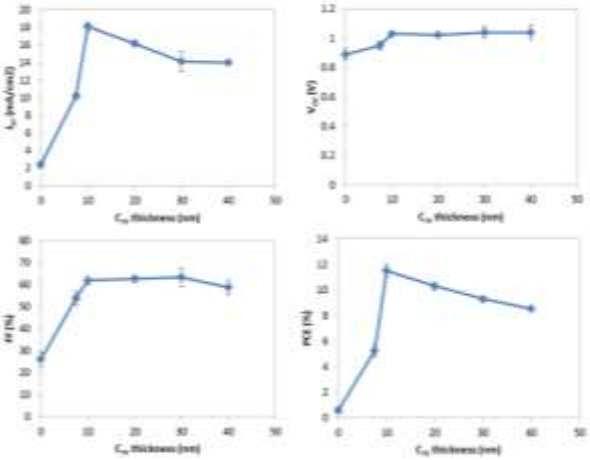

Fig. **4.** The performance parameters of the PSC devices with various thicknesses of the C70 layer as ETL.

C70 has a relatively high absorption coefficient in the wavelength range of 350–550 nm, and the total absorption of course increases with film thickness in the processed layers [33]. This absorption potentially lowers the Jsc and thus PCE of perovskite devices with a conventional structure, as investigated in this work, due to the absorption loss in the active layer in this region. Thus the drop in Jsc beyond 10nm thick C70 films can be explained partly by the drop in absorption of the perovskite layer at this wavelength range, while the increased series resistance in these devices, due to the thicker C70 layer, can explain the drop in FF and partly Jsc at increased thicknesses.

Steady state PL spectroscopy was measured to elucidate the photo-induced charge transfer and the charge recombination loss. Figure 5 shows the PL spectra of the ITO/perovskite and ITO/C70(10 nm)/perovskite films. The PL intensity of the ITO/C70(10 nm)/perovskite film is significantly weaker than that of the ITO/perovskite film. This suggests that the charge transfer at the C70 interface effectively occurred before carrier recombination takes place in the active layer, leading to a PL signal. Thus, the C70 layer significantly improved the electron extraction from the absorber layer, as also expected. According to Fig. 5, a C70 interface layer with a thickness of even a few nanometers can effectively block the holes and transport the electrons out of the active layer, allowing perovskite solar cells to use ultra-thin C70 films to achieve high performance.

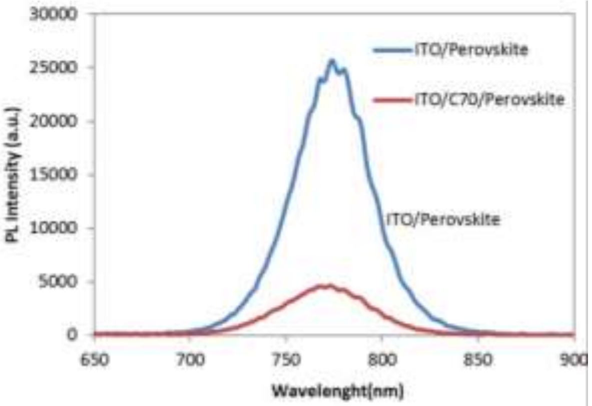

Fig. **5.** The steady state photoluminescence (PL) measurements of the ITO/perovskite and ITO/C70(10nm)/perovskite films

IV. CONCLUSION

In this study, planar perovskite solar cells with a conventional device structure that utilize C70 thin films as electron transport layers were developed. A high-quality CH3NH3PbI3 perovskite film was formed on C70-coated ITO substrates by the one-step solution method and through antisolvent treatment. Photoluminescence studies revealed the high electron-transporting and efficient hole-blocking capabilities of the C70 layer. To achieve the best PSC performance, the C70 layer thickness was optimized to reduce the effects from ETL absorption and series resistance. According to the absorption spectrum of the C70 films, a C70 ETL thickness above 10 nm show relatively strong absorption in the wavelength range 350–550 nm, and thus a reduction of the PSC performance due to drops in Jsc. At thicker C70 films, the FF also drops due to the increased series resistance while the Voc remains constant. The best PCE was 11.52% ± 0.50 reported for a 10-nm-thick C70 layer used as an ETL in the device. As the results demonstrate, ultra-thin C70 films can effectively block holes and transport electrons in PSC devices, making them efficient electron selective contacts in such devices.

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