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RSC Advance

PAPER

Efficient and stable polymer solar cells with electrochemical deposition of CuSCN as an anode interlayer

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Electrochemical deposition (ECD) of copper thiocyanate (CuSCN) film as an efficient anode interlayer (AIL) in polymer solar cells (PSCs) is reported. By elaborately optimizing the ECD conditions for the preparation of CuSCN films, a power conversion efficiency (PCE) of 6.16% has been achieved through employing P3HT:IC₆₀BA as active layer. Concerning the long-term stability, the PSCs with CuSCN as AIL maintain nearly 84% of their initial PCEs after storing in ambient conditions for 400 hours without any encapsulation, while the PCEs of the reference PSCs with PEDOT:PSS as AIL largely decrease to 35% of the initial values. Due to the advantages of the high hole mobility and superior chemical stability, ECD-prepared CuSCN is a promising alternative to PEDOT:PSS as AIL for PSCs.

1 Introduction

The Bulk heterojunction (BHJ) polymer solar cells (PSCs) have attracted increasing attention owing to their advantages of low cost, light weight, and feasibility to fabricate flexible devices.¹⁻³ A BHJ PSC is composed of a photoactive layer containing a semiconducting polymer as donor and a fullerene derivative as acceptor sandwiched between two electrodes. However, such a simple device configuration suffers from large energy barrier, severe charge carrier recombination, and remarkable current leakage at the interfaces between electrodes and semiconducting materials, resulting in low device performance. In order to solve this problem, the interlayer inserted between active layer and electrodes is thus widely used.⁴⁻⁵ Desired characteristics of an ideal interlayer include: (1) high transparency across the solar spectrum, (2) appropriate energy levels to form ohmic contact between electrodes and the active layer, (3) suitable wetting properties to facilitate the deposition of active layers, and (4) easy

preparation over large-area substrates.

While many efforts have been made on cathode interlayer using materials such as metal oxides, metal carbonates,⁶⁻⁸ titanium chelate⁹ and polyelectrolytes¹⁰⁻¹⁵ for improved device performance, relatively less materials are developed for AIL. To date, poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS)¹⁶ and metal oxides¹⁷⁻²⁰ are the mostly used AIL materials to increase hole collection in PSCs. However, PEDOT:PSS is not an ideal AIL material due to its chemical instability stemming from intrinsic natures of hygroscopicity and high acidity,²¹ yet the metal oxides require vacuum deposition and thermal treatment, which is incompatible with the large-scale fabrication and practical application of PSCs. In this regard, it is imperative to develop chemically stable and solution processible alternative AIL materials for enhanced PSC performance.

Recently, copper thiocyanate (CuSCN) has emerged as a promising interfacial material owing to its high hole mobility ($\approx 0.1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$), excellent transparency in the visible region, and solution processibility. Although some solvents such as dipropylsulfide and diethylsulfide have been adopted to prepare the CuSCN film as AIL for PSCs, perovskite solar cells, light-emitting diodes and field-effect transistors,²²⁻²⁷ the solubility of CuSCN is really limited resulting in the formation of CuSCN film with insufficient uniformity/continuity.²⁸ Moreover, these solvents are toxic and non-ecofriendly. Thus, an electrochemical deposition (ECD) approach in aqueous electrolyte has been used to prepare CuSCN film, which can avoid the use of undesired solvents for solution processing, being compatible with large-area fabrication for flexible substrates. In particular, Chappaz-Gillot et al. have reported on CuSCN thin film or nanowires prepared through ECD method with different electrolytes as AIL for PSC application, in which a PCE of 5.1% was achieved from a PCDTBT/PC₇₀BM-based device.²⁹ However, it should be noting that the CuSCN film quality and morphology are greatly dependent on the ECD conditions including potential, time, as well as temperature, which has been rarely investigated; especially, stability test of

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devices with CuSCN as AIL which is significant for a PSC application has been never carried out.³⁰⁻³¹

Herein, we focus on studying the effect of aforementioned ECD conditions on the CuSCN films as well as the device efficiency and stability of their use as AIL for PSCs. Based on elaborately optimized ECD conditions of CuSCN films, a PCE of 6.16% with $V_{oc} = 0.82$ V, FF = 0.70, and $J_{sc} = 10.76$ mA cm⁻² is achieved from the PSC with P3HT:IC₆₀BA as active layer, which is a 17% of improvement in comparison to a reference device using PEDOT:PSS as AIL ($V_{oc} = 0.82$ V, FF = 0.70, and $J_{sc} = 9.81$ mA cm⁻², with a PCE of 5.63%). More importantly, CuSCN-based PSCs can maintain 84% of their initial PCEs after storage in ambient conditions for 400 hours, much more stable than devices that based on PEDOT:PSS as AIL, whose PCEs decrease to only 35% under the same conditions. Our results verify that CuSCN is a promising alternative to PEDOT:PSS as the AIL material in PSCs for remarkably improved device stability.

2. Experimental section:

Preparation of CuSCN films: The ITO-coated glass substrates (AimCore Technology Co., Ltd) were first sonicated in detergent, acetone, ethyl alcohol, and deionized water in sequence. The CuSCN films were then electro-deposited from an aqueous solution containing 12 mM copper sulphate (CuSO₄), 12 mM ethylene diamine tetra acetic acid (EDTA) and 12 mM potassium thiocyanate (KSCN) at a different potential between 0.25 and -0.55 V versus Ag/AgCl (3 M KCl) electrode. After drying with nitrogen flow, the ITO/CuSCN substrates were transferred to N₂ filled glove box for device fabrication.

Fabrication and characterization of the PSCs: The P3HT:IC₆₀BA active layers (ca. 160 nm) were spin-casted on top of ITO/CuSCN in a N₂ filled glove box from the solution of P3HT:IC₆₀BA (1:1 weight ratio in o-chlorobenzene) with the overall concentration of 25 mg mL⁻¹. Finally 10 nm of Ca and 100 nm of Al were deposited sequentially under 6 × 10⁻⁶ Torr by thermal evaporation through a shadow mask to form an active area of ~0.064 cm². In order to ensure the credibility of our results, a batch of device containing 20 cells were fabricated.

The hole mobility test: The structure of hole-only devices is ITO/AILs/MoO₃/Au. In the trap-free region over the trap-filled limit, SCLC can be characterized by the Mott-Gurney square law $J = (9/8)\epsilon_r\epsilon_0\mu_h(V^2/L^3)$, where ϵ is the vacuum permittivity, ϵ_r is the dielectric permittivity of the active layer, L is the thickness of the AILs, and μ_h is the hole mobility.

Characterization: The electro-deposition of the CuSCN film was performed on a CHI660 electrochemical workstation (CH Instrument, Inc.). A computer-controlled Keithley 2400 source measure unit was used to characterize the J-V performance of devices with an AM 1.5G solar simulator at illumination intensity of 100 mW cm⁻². The corresponding incident photon to current conversion efficiency (IPCE) was characterized on the QTest Station 2000ADI system (Crowntech, Inc., USA). The optical transmittance spectra were measured on a Varian Cary 5500 spectrometer. Atomic force microscopy (AFM) height images were obtained using a Bruker Metrology Nanoscope III-D atomic force microscope in tapping mode under atmospheric conditions. The work functions were measured in

air by scanning Kelvin probe microscopy (SKPM) with a Bruker Metrology Nanoscope III-D atomic force microscope. Conducting AFM tips (SCM-PIT/PtIr, Bruker, USA) were used for this study with a typical spring constant of 2.8 N m⁻¹ and a resonance frequency of 75 kHz. Typical scan line frequency was 0.3 Hz and each image contained 512 × 512 pixels. The surface potential images were unprocessed original data. Advancing contact angle measurements were performed using a DSA100 system (KRÜSS).

3. Results and discussion:

The ECD system used in this work is illustrated in Fig 1(a) (see details in SI). As an initial study, we examine the crystalline structures of CuSCN films by XRD technique as shown in Figure 1(b). The XRD patterns of distinct peaks at $2\theta = 16.1^\circ$, 27.2° , and 30.3° correspond to typical diffractions of (003), (101), and (012) planes, respectively, which indicates the formation of rhombohedral phase (β -phase) in CuSCN films.³² Figures 1(c) and 1(d) show device architecture and chemical structures of donor/acceptor materials as well as their energy levels relative to vacuum level. The work function of CuSCN films is slightly higher than PEDOT:PSS, (Fig.S1), which is beneficial for hole extraction and blocking electrons reaching ITO electrode to some extent.

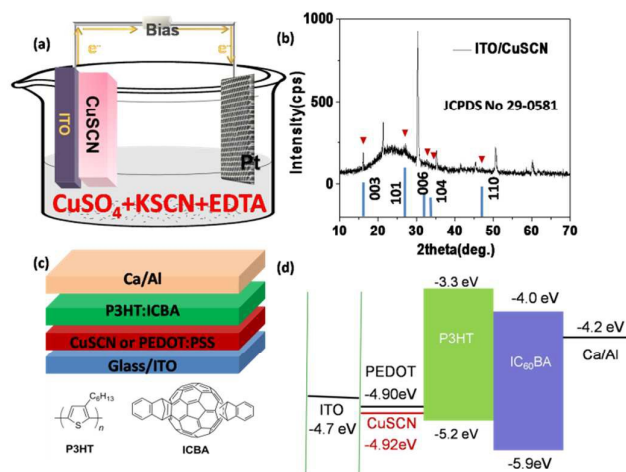


Fig1. (a) Schematic illustration of the electrochemical deposition method. (b) XRD patterns of the CuSCN film at R.T. (c) Device architecture and chemical structures of P3HT and IC₆₀BA. (d) Energy levels of the materials used in PSCs.

The CuSCN films were prepared under variable deposition potential, time, and deposition temperatures to investigate the effect of ECD conditions on their film morphology. Firstly, the surface morphology of CuSCN films with different deposition potentials (-0.255, -0.355, -0.455, -0.555V) was examined by atomic force microscopy (AFM). The more negative deposition potential is beneficial for grain refinement leading to homogeneous CuSCN films as shown in Figure 2, but the deposition potential of -0.555V may form metal Cu that induce charge carriers recombination based on the device performance described as follows.³³ Secondly, the coverage of CuSCN film is obviously time-dependent, hence the deposition

time varying from 10 to 40s was applied at a constant potential of -0.455V (Fig.S2). When the deposition time is in the range of 20 to 30 s, the ITO substrate can be fully covered with the CuSCN film with a thickness of ca. 40 nm and roughness of 7nm. Finally, it was found that the morphology of CuSCN films was also ECD temperature dependent (0°C and R.T.). When depositing at 0°C , a denser and smoother film can be obtained due to the slow nucleation and growth rate of the CuSCN molecules as shown in Fig.2(e). In addition, it is worthy of noting that as depicted in Fig.2(e)-(f) the CuSCN film is more hydrophobic than the PEDOT:PSS film such that the suitable surface energy of CuSCN film would facilitate the deposition of the active layer. (Table.S1-S2)

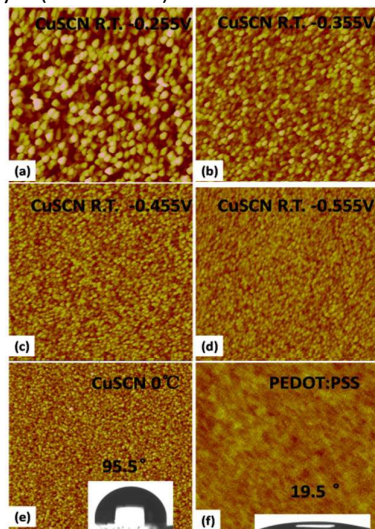


Fig.2.(a-d) AFM topography images of CuSCN films with different deposition potentials (deposition temperature at R.T.). (e-f) AFM topography images and the surface contact angles of CuSCN (deposition temperature at 0°C) and PEDOT:PSS films. Scale bar: $5\mu\text{m} \times 5\mu\text{m}$.

Table 1. Summary of device parameters of the P3HT:IC₆₀BA-based PSCs with CuSCN and PEDOT:PSS as AILs under AM1.5 G illumination (thickness error: $\pm 5\text{nm}$)

AIL	Varied ECD conditions (thickness)	$J_{sc}(\text{mAcm}^{-2})$	$V_{oc}(\text{V})$	FF(%)	PCE(%)	
CuSCN	Potential	-0.355V(45nm)	9.70	0.82	68	5.44
		-0.455V(45nm)	9.87	0.82	70	5.67
		-0.555V(45nm)	9.54	0.80	66	5.04
	Time	10s (25nm)	10.81	0.76	57	4.71
		20s (35nm)	10.37	0.80	69	5.76
		30s (40nm)	10.19	0.82	70	5.86
		40s (50nm)	10.14	0.80	54	4.38
	Temperature	R.T. (40nm)	10.19	0.82	70	5.86
0°C (40nm)		10.76	0.82	70	6.16	
PEDOT:PSS	(40nm)	9.81	0.82	70	5.63	
CuSCN ^a	-0.4V, R.T. (90nm)	9.60	0.90	59	5.1	

^aData extracted from the literature work.²⁹

The PSCs were fabricated using the CuSCN film prepared under aforementioned optimized conditions as AIL and P3HT:IC₆₀BA as the active layer. The J-V curves of these devices under AM 1.5G illumination at 100mW cm^{-2} are displayed in Fig.3(a-d). Detailed characteristic parameters of PSCs are summarized in Table 1. A PCE of 6.16% ($V_{oc} = 0.82\text{V}$, FF = 0.70, $J_{sc} = 10.76\text{mA cm}^{-2}$) employing CuSCN film prepared from the optimized ECD condition (deposition potential of -0.455V for 30 s at 0°C) as AIL was achieved, while the PCE of reference device using PEDOT:PSS as AIL can only reach 5.63% ($V_{oc} = 0.82\text{V}$, FF = 0.70, and $J_{sc} = 9.81\text{mA cm}^{-2}$). Obviously, the CuSCN-based device shows better PCE than PEDOT:PSS one due to the increased J_{sc} . What's more, the better PCE of CuSCN-based device results from the higher J_{sc} and FF as compared to the best results of the CuSCN nanowire as AIL,²⁹ which might attribute to the denser and smoother CuSCN film with electro-deposited at 0°C .

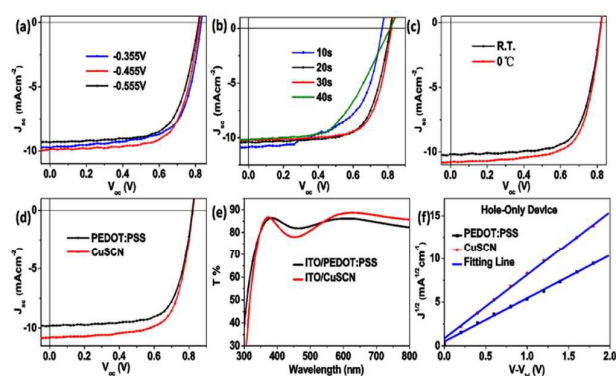


Fig.3. J-V curves of the PSC with CuSCN as AIL by (a) different deposition potentials; (b) different deposition time; (c) different deposition temperature and (d) J-V curves of the PSCs with different AILs. (e) Optical transmission spectra of different AILs. (f) Hole mobility of CuSCN and PEDOT:PSS films measured by SCLC method.

The enhanced J_{sc} can be attributed to the following reasons. Firstly, the transparency of CuSCN is even slightly higher than PEDOT:PSS in the range of 550 nm to 800 nm (Fig. 3e), which allows more light to be absorbed by the active layer for generating photocurrents. (Fig. S3) Another reason can be the higher hole mobility of CuSCN film than PEDOT:PSS film, which is estimated by the space-charge-limited current (SCLC) model from the hole-only devices (ITO/AILs/MoO₃/Au)³⁴. Fig. 3f shows the $J^{1/2}$ -V curves of the hole-only devices in dark. The hole mobility of CuSCN and PEDOT:PSS are $1.84 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $8.16 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively. The enhanced hole mobility of CuSCN film could further contribute to the enhancement of J_{sc} . Finally, the reduced series resistance (R_s) and increased parallel resistance (R_{sh}) with CuSCN as AIL (R_s : $8.2 \Omega / \text{cm}^2$, R_{sh} : $1252.56 \Omega / \text{cm}^2$) has been achieved compared to PEDOT:PSS as AIL (R_s : $8.68 \Omega / \text{cm}^2$, R_{sh} : $729.96 \Omega / \text{cm}^2$), which might be the formation of better ohmic contact between ITO/CuSCN and active layer.

The incident photon to current conversion efficiency (IPCE) and the integrated current curves of the PSCs with CuSCN and PEDOT:PSS as AILs were measured (Fig. S4). The calculated J_{ph} value by integrating the IPCE spectra is 9.35 mA cm^{-2} and 8.96 mA cm^{-2} , which are very close to the measured values of the PSCs with CuSCN and PEDOT:PSS as AILs, respectively. The dark J-V curves of the PSCs with CuSCN and PEDOT:PSS as AILs exhibit similar reverse saturation and output current, which facilitates the selectivity of the anodes (Fig. S5).³⁵⁻³⁶ These results demonstrate the potential of CuSCN to replace PEDOT:PSS as AIL for achieving efficient PSCs.

The non-encapsulated devices based on CuSCN and PEDOT:PSS as AILs were tested under AM1.5 G illumination at 1 sun measured for 400 hours in order to study the operational stability of the PSCs. Overall, an initial fast decay in the PCE can be observed within the first 50 h followed by a stable platform. The deterioration of the PCE is in general caused by a decrease in FF and J_{sc} , whereas the V_{oc} is relatively stable within the measured period. The CuSCN-based PSC exhibits improved stability measured after 400h, reaching 84% of its initial PCE value, while only 35% of its initial PCE value of the device based on PEDOT:PSS as AIL can maintain. This clearly reveals that the CuSCN-based PSCs provide better stability than the PEDOT:PSS-based counterparts. The poor long-term stability of PEDOT:PSS as AIL can be assigned to its several problems like high acidity and hygroscopicity. In contrast, the CuSCN is more hydrophobic than PEDOT:PSS, which could be manifested by the contact angle test as shown above, favoring the long-term operational stability for the devices. These results provide strong evidences of the general utility of CuSCN as an AIL to be a qualified alternative to PEDOT:PSS for more stable PSCs.

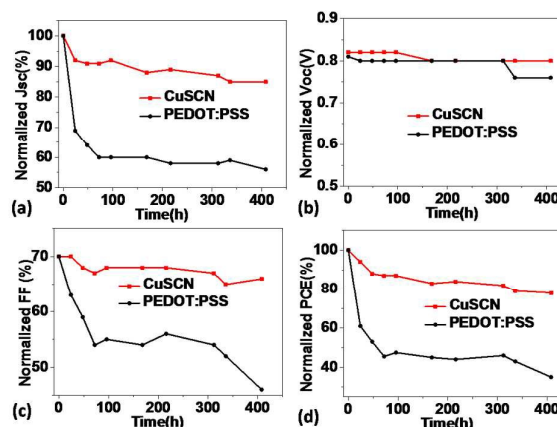


Fig. 4. Lifetime measurements of the non-encapsulated P3HT:IC₆₀BA devices with CuSCN and PEDOT:PSS as AILs under AM1.5 G illumination at 1 sun measured for 400 hours. The curves show the variation in (a) short-circuit current density (J_{sc}), (b) open-circuit voltage (V_{oc}), (c) fill factor (FF) and (d) PCE and each is normalized to the initial value.

4. CONCLUSION:

In conclusion, we have demonstrated the application of ECD method for the preparation of CuSCN film as AIL for efficient and stable polymer solar cells. PSCs based on CuSCN as AIL was found to increase the J_{sc} , while maintaining high V_{oc} and FF in comparison with the currently used PEDOT:PSS, hence improving the PSC's maximum PCE to 6.16% by employing P3HT:IC₆₀BA as active layer. More importantly, the device fabricated with hydrophobic CuSCN film as AIL retained 84% PCE after a storage under ambient condition for 400h without any encapsulation, much more stable than the PEDOT:PSS based PSCs. This work demonstrates that CuSCN is qualified as an effective AIL for efficient and stable PSCs, and meanwhile showing potential application as a stable hole-carrying material for other optoelectronic devices.

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