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Nanoarrayed Cu₂O/CuSCN HTLs for High-Efficiency Organic Solar Cells

Organic solar cells (OSCs) offer a promising low-cost and flexible alternative to conventional photovoltaic technologies; however, their performance remains limited by inefficient charge transport and carrier recombination losses. In this work, we present a novel nanoarrayed hole-transport layer (HTL) based on Cu₂O/CuSCN heterostructures fabricated via a two-step electrodeposition process. Vertically aligned Cu₂O nanocrystals were grown on CuSCN films, forming a nanostructured charge-extraction framework with enhanced interfacial contact and potentially reduced carrier transport distances. The morphology and properties of the films were systematically tuned by controlling the electrochemical parameters, enabling the formation of well-defined nanoarchitectures. Photoelectrochemical measurements revealed enhanced interfacial photocurrent and improved charge-extraction characteristics of the composite structures compared with the corresponding single-layer films. When integrated into PTB7-th:PCBM-based OSCs, the nanoarrayed HTL substantially improved device performance, increasing the open-circuit voltage from 0.62 to 0.84 V, the short-circuit current density from 7.99 to 11.8 mA cm⁻², and the power conversion efficiency from 2.76% to 5.59%. These improvements are attributed to more efficient charge collection and suppressed recombination enabled by the nanoarray architecture. This study demonstrates a low-temperature electrodeposition strategy for engineering advanced HTLs and highlights the potential of nanoarrayed Cu₂O/CuSCN heterostructures for future organic and hybrid photovoltaic devices.

Keywords: Organic solar cells, copper thiocyanate films, PTB7-Th: PCBM, Cu₂O nanocrystals, electrodeposition, bulk heterojunction, hole transport layer, charge transfer

Introduction

Organic (polymer-based) solar cells are widely regarded as one of the most promising renewable-energy technologies and are becoming increasingly competitive with traditional silicon devices. This interest is largely driven by their low-cost fabrication methods [1] and the distinctive properties of organic materials [2]. In particular, organic photoactive and conductive polymers are flexible and semi-transparent [3], allowing them to be deposited onto flexible or transparent substrates and integrated into a variety of surfaces, such as building windows [4]. Additionally, many organic materials can be processed from solution, enabling thin-film fabrication through techniques such as spin-coating, dip-coating, and roll-to-roll printing. These processes can be carried out at or near room temperature with only mild thermal treatment (≤ 110 °C),

significantly lowering production costs compared to conventional inorganic technologies [5, 6]. Organic materials are also often compatible with biomaterials, which open the door to environmentally friendly and bio-integrated applications [7].

Despite these advantages, the efficiency of organic solar cells (OSCs) still trails behind that of silicon and perovskite technologies [8, 9]. In conventional OSC designs, the photoactive layer consists of a blend of a donor polymer and a fullerene-based acceptor, forming a bulk heterojunction (BHJ) positioned between hole- and electron-transport layers. When light is absorbed, excitons are generated in the donor material and must diffuse to donor–acceptor interfaces to dissociate into free charge carriers. However, because exciton diffusion lengths in many polymers are relatively short compared to the thickness required for efficient light absorption, the BHJ architecture is employed to maximize interfacial area [10]. Even so, device performance remains strongly limited by morphology [11, 12] and by imbalances in charge carrier mobility—hole mobility is typically lower than electron mobility [13]—leading to charge accumulation and recombination losses. In addition, regions within the BHJ often lack efficient percolation pathways to the charge transport layers, resulting in incomplete charge collection [14].

To mitigate these limitations, extensive research has focused on improving hole-transport layers (HTLs), including the use of conductive polymers (e.g., PEDOT:PSS), transition metal oxides, and inorganic p-type semiconductors such as CuSCN. While these materials can provide suitable energy level alignment and chemical stability, they are typically implemented as planar thin films that rely on one-dimensional (1D) vertical charge extraction. This configuration does not fully match the three-dimensional nature of charge generation within the BHJ layer, where photogenerated carriers are distributed throughout the film volume. As a consequence, holes must often travel laterally over relatively long distances before reaching the HTL interface, increasing the probability of recombination and limiting device efficiency. Although previous studies have improved HTL performance through doping, interfacial engineering, and conductivity enhancement, comparatively little attention has been given to engineering the spatial architecture of HTLs to fundamentally alter charge extraction pathways. This represents a key gap in the current development of OSCs.

In this work, we address this limitation by introducing a nanoarrayed Cu₂O/CuSCN heterostructured HTL, which transforms the conventional planar extraction interface into a quasi-two-dimensional (2D) charge collection network. The structure is formed by electrodepositing vertically aligned Cu₂O nanocrystals onto CuSCN films, creating an extended interfacial framework that penetrates toward the active layer. Unlike planar HTLs, this nanoarrayed architecture increases the effective contact area and provides multiple localized pathways for hole extraction. As a result, the average transport distance for photogenerated holes is reduced, leading to suppressed recombination and more efficient charge collection. Importantly, this approach combines the favorable electronic properties of CuSCN with the structural advantages of Cu₂O nanocrystals, while remaining compatible with low-temperature and scalable fabrication processes.

A two-step electrodeposition method was developed to precisely control the nucleation and growth of Cu₂O nanocrystals on CuSCN films, enabling the formation of well-defined nanoarrayed heterostructures. The resulting architecture promotes strong interfacial coupling and efficient charge transfer. Photoelectrochemical measurements demonstrate enhanced interfacial photocurrent and improved hole extraction in the composite structures compared to single-layer counterparts. When integrated into PTB7-Th:PCBM-based OSCs, the nanoarrayed HTL leads to significant improvements in open-circuit voltage, short-circuit current density, and overall power conversion efficiency.

Overall, this work demonstrates that engineering the geometry of the HTL—from planar to nanoarrayed—provides a fundamentally different strategy for improving charge extraction in OSCs. The proposed Cu₂O/CuSCN nanoarray architecture offers a simple, low-temperature, and scalable route to enhance charge management and device performance, thereby addressing a critical limitation of conventional HTL designs and contributing to the development of next-generation organic photovoltaic technologies.

Experimental

Materials

To ensure the fabrication of high-purity thin films with minimal contamination, all reagents of analytical grade or higher purity were purchased from Sigma-Aldrich. The chemicals used included copper(II) sulfate (anhydrous, ≥99.99 %), ethylenediaminetetraacetic acid (EDTA, OmniPur® Grade, ≥99.5 %), potassium thiocyanate (ReagentPlus®, ≥99.0 %), diethanolamine (DEA, BioUltra, ≥99.5 %), lactic acid (ACS reagent, ≥85 %), and sodium hydroxide (ACS reagent, ≥97.0 %).

All aqueous solutions were prepared using deionized water with a resistivity of $18.2 \text{ M}\Omega\cdot\text{cm}$, obtained from a Smart-Q30UT purification system (Drawell Scientific). The use of high-purity reagents and ultrapure water minimized the incorporation of impurities during thin-film formation.

Preparation of CuSCN Thin Films

CuSCN layers were deposited onto fluorine-doped tin oxide (FTO) glass substrates from Ossila with active area 90 mm^2 (sheet resistance $\sim 15 \text{ }\Omega/\text{sq}$). Prior to deposition, the substrates were sequentially cleaned in an ultrasonic bath using a 2 % Hellmanex solution (10 min), deionized water (10 min), and 2-propanol (10 min), followed by drying under a nitrogen stream. The substrates were subsequently treated under UV irradiation for 15 min to remove residual organic contaminants and improve surface wettability.

Two types of CuSCN films were fabricated. The first, denoted CuSCN-E, was electrodeposited from an aqueous solution containing 12 mM CuSO_4 with equimolar concentrations of EDTA and KSCN. The second type, CuSCN-D, was obtained from a precursor solution composed of 15 mM CuSO_4 , 67.5 mM diethanolamine (DEA), and 45 mM KSCN. In both cases, EDTA or DEA was added prior to KSCN to suppress the formation and precipitation of $\text{Cu}(\text{SCN})_2$ complexes. The pH values of the EDTA- and DEA-based electrolytes were adjusted to 1.6 and 8.2, respectively.

Electrodeposition was performed in a three-electrode configuration using a platinum counter electrode and an Ag/AgCl (saturated KCl) reference electrode (Fig. 1). Chronoamperometric deposition was carried out at fixed potentials of -0.3 V (CuSCN-E) and -0.45 V (CuSCN-D) versus Ag/AgCl. The deposition time was varied in the range of 60–300 s to control film thickness. The resulting CuSCN films had a typical thickness of 40–60 nm, as estimated from deposition charge and confirmed by profilometry measurements.

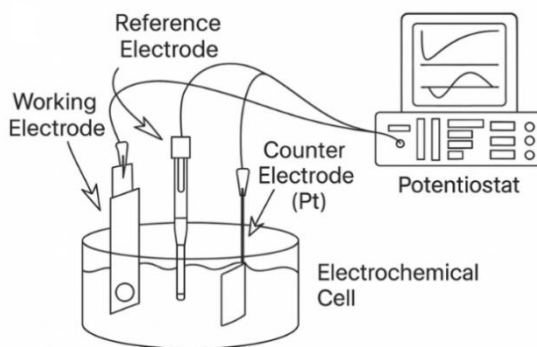


Figure 1. Three-electrode setup for electrodeposition

Preparation of Cu_2O Thin Films and $\text{Cu}_2\text{O}/\text{CuSCN}$ Nanoarrays

Cu_2O films were deposited electrochemically onto CuSCN/FTO substrates using an alkaline electrolyte containing lactate-complexed copper ions. The electrolyte was prepared by dissolving 0.2 M CuSO_4 and 3 M lactic acid in deionized water, followed by the addition of 0.5 M K_2HPO_4 as a buffering agent. The solution pH was adjusted to 12 by gradual addition of 2 M KOH.

To form nanoarrayed $\text{Cu}_2\text{O}/\text{CuSCN}$ heterostructures, a two-step electrodeposition process was employed. First, a galvanostatic nucleation step was carried out at a current density of $-0.1 \text{ mA}\cdot\text{cm}^{-2}$ for 10 s to generate spatially separated Cu_2O nuclei. This was followed by a potentiostatic growth step at -0.3 V versus Ag/AgCl for 60 s, enabling controlled growth of vertically aligned Cu_2O nanocrystals. Deposition was conducted at 30°C . The resulting Cu_2O nanocrystals had an average lateral size of $\sim 80 \text{ nm}$ and an effective height of approximately 50–80 nm, forming a nanoarrayed morphology on the CuSCN layer.

Device Fabrication

Organic solar cells were fabricated using the following architecture:

FTO / CuSCN (or $\text{Cu}_2\text{O}/\text{CuSCN}$) / PTB7-Th: PC_{61}BM / Al

The active layer (photoactive layer, PL) consisted of a PTB7-Th: PC_{61}BM bulk heterojunction. The solution was prepared by dissolving PTB7-Th and PC_{61}BM in chlorobenzene at a weight ratio of 1:1.5 with a total concentration of 25 mg/mL. The solution was stirred at 60°C for 12 h prior to deposition.

The active layer was spin-coated onto the HTL-coated substrates at 3000 rpm for 60 s, resulting in a film thickness of approximately 90–110 nm. After deposition, the films were dried under inert conditions.

A 100 nm thick Al electrode was thermally evaporated under high vacuum ($\sim 10^{-6}$ Torr) through a shadow mask, defining an active device area of 0.1 cm². The devices were subsequently encapsulated using epoxy in a nitrogen atmosphere to minimize degradation.

Characterization

The surface morphology of the films was analyzed using scanning electron microscopy (SEM, Carl Zeiss Crossbeam 540 with GEMINI II). Atomic force microscopy (AFM, NTEGRA ACADEMIA) was used to evaluate surface roughness and morphology of the active layer. The all thickness of the layers was measured by AFM making gentle scratch.

Photoelectrochemical (PEC) measurements were carried out using a CS300 potentiostat (Corrtest Instruments) under chopped illumination from an Ossila solar simulator (AM 1.5G, 100 mW·cm⁻²). Current–voltage (I–V) characteristics of the fabricated devices were measured under the same illumination conditions by Ossila source meter.

Results and Discussion

Next, we developed a method for the electrodeposition of a Cu₂O nanocrystal array on the surface of CuSCN. To form the array, electrodeposition was first performed in a galvanostatic regime at a low current density (-0.1 mA·cm⁻²) at 30 °C and pH 12 for 10 s. This stage enabled a slow nucleation rate and the formation of Cu₂O nuclei sufficiently spaced from each other. Subsequently, the process was switched to a potentiostatic mode, and electrodeposition was continued at -0.3 V versus Ag/AgCl reference electrode for 1 min. This combination of modes allowed the deposition of well-separated Cu₂O nanocrystals on the CuSCN surface, which shown in Figure 2A with average size of the Cu₂O nanocrystals of 80 nm.

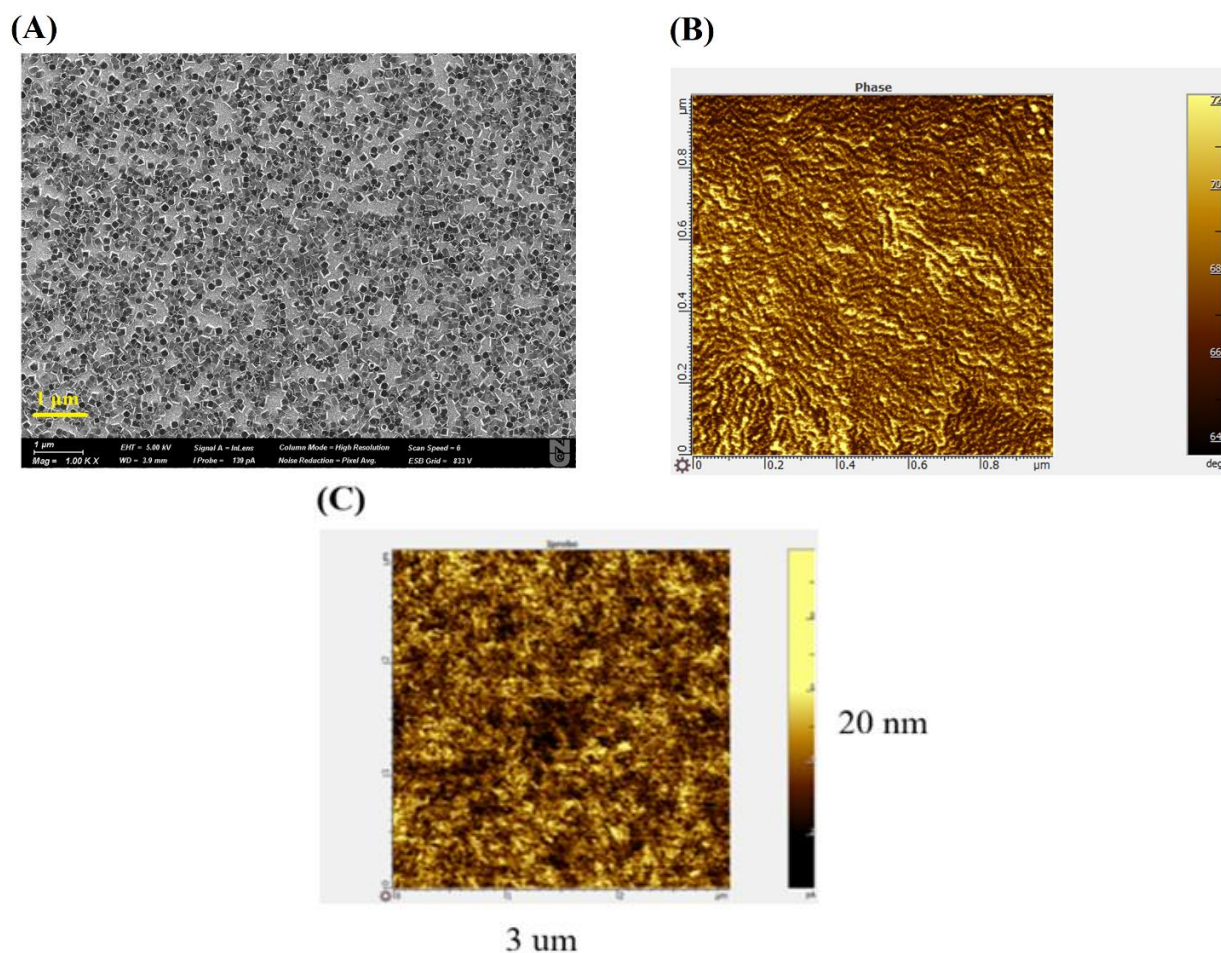


Figure 2. (A) SEM image of the nanoarrayed Cu₂O/CuSCN composite film. AFM phase (B) and topography (C) images of the PTB7-th:PCBM blend layer deposited on the surface of the Cu₂O/CuSCN composite film

Further, the nanoarrayed $\text{Cu}_2\text{O}/\text{CuSCN}$ composite films were employed as the hole transport layer in organic solar cells. In the next step, a bulk heterojunction (BHJ) layer based on a PTB7-th: PCBM blend was spin-coated onto the surface of the as-grown nanoarrayed $\text{Cu}_2\text{O}/\text{CuSCN}$ composite films. The AFM image of the 100 nm-thick PTB7-th: PCBM layer is shown in Figure 2B. As shown in Figure 2B, the AFM phase image of the PTB7-Th: PC₆₁BM film reveals a continuous and laterally homogeneous surface with nanoscale contrast. The observed phase variations suggest the presence of donor–acceptor phase separation within the blend, which is typical for bulk heterojunction systems. The uniform distribution of phase domains indicates good film formation and consistent coverage over the underlying $\text{Cu}_2\text{O}/\text{CuSCN}$ layer. Such morphology is expected to support efficient interfacial contact and facilitate charge transfer processes at the HTL/active layer interface.

Finally, device fabrication was completed by thermally evaporating a 100 nm thick Al electrode onto the active layer, followed by encapsulation with epoxy. A schematic of the full fabrication process is shown in Figure 3. For comparison, reference organic solar cells were also prepared using only a CuSCN layer as the hole transport layer (HTL), with the CuSCN thickness fixed at 50 nm in both device configurations. The current–voltage (I–V) characteristics and the corresponding photovoltaic parameters of the fabricated devices are presented in Figure 4 and summarized in Table 1.

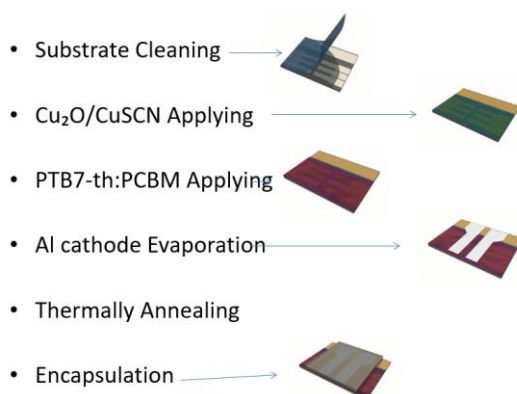


Figure 3. Schematic illustration of the fabrication process of organic solar cells

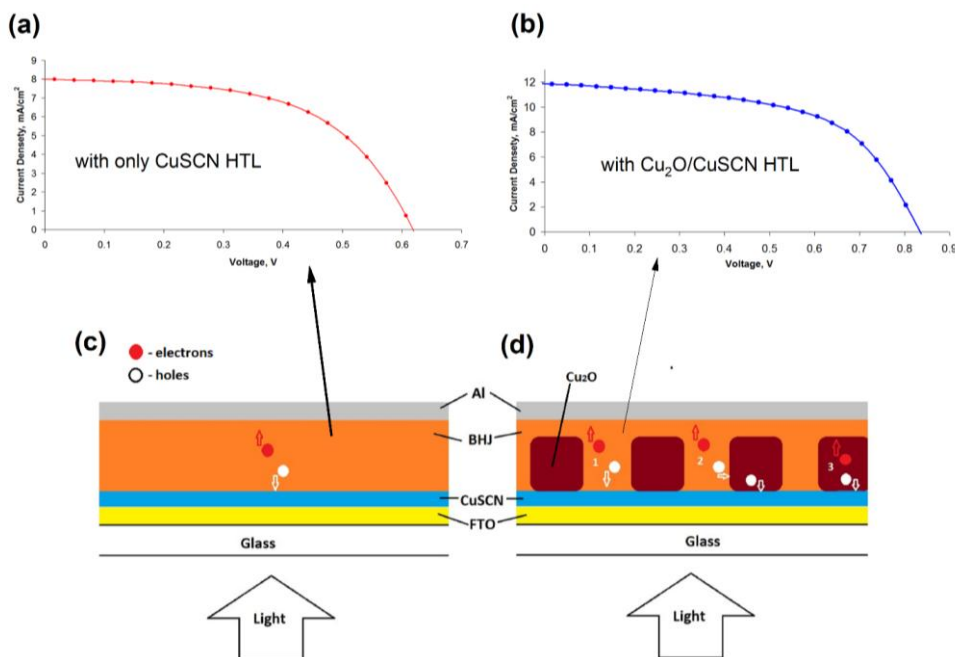


Figure 4. Schematic current–voltage characteristics of organic solar cells (OSCs) incorporating (a) CuSCN and (b) nanoarrayed $\text{Cu}_2\text{O}/\text{CuSCN}$ composite hole transport layers; and mechanism of charge carrier separation in OSCs: (c) one-dimensional (1D) injection in conventional OSCs based on CuSCN, and (d) two-dimensional (2D) injection in the developed OSC with nanoarrayed $\text{Cu}_2\text{O}/\text{CuSCN}$ HTL

Table 1

Photovoltaic parameters of the fabricated devices

Device	Voc, V	Jsc, mA/cm ²	Fill Factor	PCE, %
With CuSCN HTL	0.62	7.99	56.1	2.76±0.05
With nanoarrayed Cu ₂ O/CuSCN HTL	0.84	11.8	56.4	5.59±0.05

As shown in the I–V characteristics, replacing the planar CuSCN HTL with the nanoarrayed Cu₂O/CuSCN composite markedly improves device performance. The open-circuit voltage (Voc) increases from 0.62 V to 0.84 V ($\approx +35.5\%$), the short-circuit current density (Jsc) increases from 7.99 mA·cm⁻² to 11.8 mA·cm⁻² ($\approx +47.7\%$), and the power conversion efficiency (PCE) rises from 2.76 % to 5.59 % ($\approx +102.5\%$, i.e., roughly doubled). The fill factor remains essentially unchanged (56.1 $\rightarrow$ 56.4), indicating that the series resistances is no substantially increased by the nanoarrayed layer.

These performance enhancements can be attributed to the nanoarrayed Cu₂O/CuSCN morphology, which establishes efficient two-dimensional (2D) pathways for hole transport. The vertically oriented Cu₂O nanocrystals increase the interfacial contact area with the PTB7-th: PCBM active layer and introduce additional channels for hole extraction into the HTL. As a result, the average lateral distance that holes must travel within the active layer is reduced, decreasing recombination losses and improving charge collection efficiency, as reflected in the significant increase in Jsc. The higher Voc is likely due to improved energy level alignment and suppressed interfacial recombination at the donor/HTL interface provided by the composite structure. Collectively, enhanced charge collection (higher Jsc) and reduced recombination (higher Voc) account for the approximately twofold increase in PCE.

In our previous article the electrodeposition and detailed characterization of Cu₂O, CuSCN, and Cu₂O/CuSCN composite thin films were systematically explored with the aim of developing efficient hole-transport layers for organic solar cells. Careful optimization of electrochemical parameters—such as applied potential, current density, pH, and temperature—enabled precise tuning of the films' morphology, crystallinity, and interfacial properties [15].

Potentiostatic and galvanostatic investigations of Cu₂O electrodeposition revealed that the structural and morphological properties of the films are strongly dependent on the deposition regime. Well-defined cubic grains with a preferred orientation (111) were obtained under conditions of high pH (12) and moderate potentials (Fig. 5), whereas deviations from these parameters resulted in rougher surfaces and the partial formation of secondary phases [15]. An increase in deposition temperature led to smoother and denser films, reflecting enhanced ion mobility and thermally driven grain reorganization.

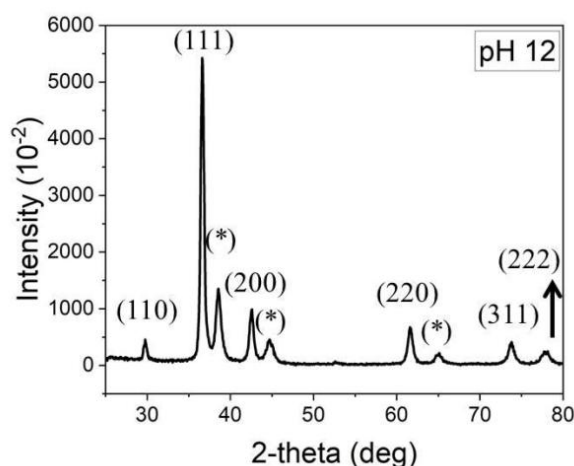


Figure 5. XRD spectra of Cu₂O thin films grown by potentiostatic electrochemical deposition [15]

For electrodeposited CuSCN films, the choice of complexing agent (EDTA or DEA) was found to significantly influence film uniformity and grain size [15]. Films prepared from EDTA-based electrolytes (CuSCN-E) exhibited smoother and more homogeneous morphologies, making them more suitable as hole-transport layers. These films also served as effective templates for subsequent Cu₂O deposition, enabling the formation of high-quality Cu₂O/CuSCN bilayer composites with well-defined interfaces.

Photoelectrochemical measurements further confirmed the enhanced interfacial performance of the Cu₂O/CuSCN heterostructures, demonstrating approximately a twofold increase in photocurrent density compared to single-layer Cu₂O films. This enhancement is attributed to favorable band alignment and more efficient hole extraction across the heterojunction [15].

Finally, nanoarrayed Cu₂O/CuSCN composite films were successfully implemented as hole-transport layers in PTB7-th: PCBM-based organic solar cells. The introduction of Cu₂O nanocrystal arrays led to a significant improvement in device performance, doubling the power conversion efficiency (from 2.76 % to 5.59 %) as a result of enhanced charge extraction and reduced recombination losses. The nanoarray architecture provides multiple lateral pathways for hole transport, promoting more efficient coupling between the active layer and the HTL.

Overall, the results presented in this chapter highlight electrodeposition as a versatile and controllable technique for the fabrication of high-quality Cu₂O/CuSCN heterostructures. These nanoarrayed composites demonstrate strong potential as efficient, stable, and scalable hole-transport layers for next-generation organic and hybrid photovoltaic devices.

Conclusion

In this work, Cu₂O, CuSCN, and Cu₂O/CuSCN composite thin films were fabricated by electrodeposition, and a two-step deposition approach combining galvanostatic nucleation and potentiostatic growth was applied to form nanoarrayed Cu₂O structures on CuSCN layers. This method enabled the preparation of heterostructured films with controlled morphology and good interfacial contact between the layers.

The incorporation of nanoarrayed Cu₂O/CuSCN structures as hole-transport layers in PTB7-Th: PC₆₁BM-based organic solar cells resulted in improved device performance compared to reference devices based on planar CuSCN. In particular, increases in open-circuit voltage and short-circuit current density were observed, leading to an enhancement of power conversion efficiency from 2.76 % to 5.59 %. These results indicate that the modified HTL structure can contribute to more efficient charge extraction.

The observed improvement in device performance is attributed to the increased interfacial contact area and the presence of additional pathways for hole transport provided by the nanoarrayed structure. This may reduce the average transport distance of charge carriers within the active layer and contribute to more effective charge collection.

Overall, the results demonstrate that electrodeposited Cu₂O/CuSCN nanoarrayed structures are a promising approach for modifying hole-transport layers in organic solar cells. Further work is required to systematically optimize the deposition parameters and to perform more detailed structural and electrical characterization in order to fully understand the mechanisms governing the observed improvements.

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Жоғары тиімді органикалық күн батареяларына арналған Cu₂O/CuSCN наноөлшемді массивті кемтіктерді тасымалдау қабаттары

Органикалық күн батареялары дәстүрлі фотовольтаикалық технологияларға перспективалы, арзан әрі оңтайлы балама, алайда олардың тиімділігі әлі де заряд тасымалының тиімсіздігімен және рекомбинация шығындарымен шектеледі. Жұмыста екі сатылы электрлік тұндыру процесі арқылы алынған Cu₂O/CuSCN гетероқұрылымдарына негізделген жаңа наноөлшемді массивті кемтіктерді тасымалдау қабаты (HTL) ұсынылған. Тік бағытталған Cu₂O нанокристалдары CuSCN қабықшаларының үстінде өсіріліп, жақсартылған фазалар аралық байланысы бар және тасымалдаушылардың жүріп өтетін қашықтығы қысқартылған екі өлшемді зарядтарды бөліп алу құрылымын қалыптастырды. Қабықшалардың морфологиясы мен қасиеттері электрхимиялық параметрлерді бақылау арқылы жүйелі түрде реттелді, бұл айқын анықталған нано архитектураларды қалыптастыруға мүмкіндік берді. Фотоэлектрхимиялық өлшеулер композиттік құрылымдарда бірқабатты үлгілермен салыстырғанда фазааралық фототоктың жақсарғанын және кемтіктердің неғұрлым тиімді бөлініп алынатынын көрсетті. PTB7-Th: PCBM негізіндегі органикалық күн батареяларына енгізілген кезде наноөлшемді массивті HTL құрылғылардың сипаттамаларын айтарлықтай жақсартты: ашық тізбектегі кернеу (0,62-ден 0,84 В-қа дейін), қысқа тұйықталу тогының тығыздығы (7,99-дан 11,8 мА·см⁻²-қа дейін) және энергияны түрлендіру тиімділігі (2,76 %-дан 5,59 %-ға дейін) артты. Бұл жақсартулар рекомбинацияның азаюы мен және массивтің наноархитектурасының арқасында зарядтарды неғұрлым тиімді жинаумен түсіндіріледі. Бұл зерттеу жетілдірілген кемтіктерді тасымалдау қабаттарын жасауға арналған ауқымдалатын, төмен температуралы стратегияны көрсетеді және жаңа буын органикалық әрі гибриді күн батареялары үшін Cu₂O/CuSCN наноөлшемді гетероқұрылымдарының әлеуетін айқындайды.

Кілт сөздер: органикалық күн батареялары, мыс тиоцианатының қабықшалары, PTB7-Th: PCBM, Cu₂O нанокристалдары, электрлік тұндыру, көлемдік гетероқосылыс, кемтіктерді тасымалдау қабаты, заряд тасымалы

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Наноразмерные массивные слои переноса дырок $\text{Cu}_2\text{O}/\text{CuSCN}$ для высокоэффективных органических солнечных элементов

Органические солнечные элементы представляют собой многообещающую недорогую и гибкую альтернативу традиционным фотовольтаическим технологиям, однако их эффективность по-прежнему ограничена неэффективным транспортом зарядов и потерями на рекомбинацию. В данной работе мы представляем новый наноразмерный массивный слой переноса дырок (HTL), основанный на гетероструктурах $\text{Cu}_2\text{O}/\text{CuSCN}$, полученных с помощью двухэтапного процесса электроосаждения. Вертикально ориентированные нанокристаллы Cu_2O были выращены на пленках CuSCN , формируя двумерную структуру извлечения зарядов с улучшенным межфазным контактом и сокращёнными расстояниями переноса носителей. Морфология и свойства плёнок систематически настраивались путём контроля электрохимических параметров, что позволило сформировать хорошо определённые наноструктуры. Фотоэлектрохимические измерения показали улучшенный межфазный фототок и более эффективное извлечение дырок в композитных структурах по сравнению с однослойными аналогами. При интеграции в органические солнечные элементы на основе PTB7-th: PCBM наноразмерный массивный HTL значительно улучшил характеристики устройств, увеличив напряжение холостого хода (с 0,62 до 0,84 В), плотность тока короткого замыкания (с 7,99 до 11,8 $\text{mA}\cdot\text{cm}^{-2}$) и коэффициент преобразования энергии (с 2,76 % до 5,59 %). Эти улучшения обусловлены снижением рекомбинации и более эффективным сбором зарядов благодаря наноструктуре массива. Данное исследование демонстрирует масштабируемую низкотемпературную стратегию создания усовершенствованных слоёв переноса дырок и подчёркивает потенциал наноразмерных гетероструктур $\text{Cu}_2\text{O}/\text{CuSCN}$ для солнечных элементов нового поколения — как органических, так и гибридных.

Ключевые слова: органические солнечные элементы, плёнки тиоцианата меди, PTB7-Th: PCBM, нанокристаллы Cu_2O , электроосаждение, объемный гетеропереход, слой переноса дырок, перенос заряда

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