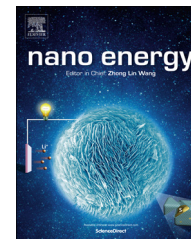


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RAPID COMMUNICATION

Highly efficient organic solar cells based on a robust room-temperature solution-processed Copper iodide hole transporter

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KEYWORDS

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Solution processed

Abstract

Q3 Achieving high performance and reliable organic solar cells hinges on the development of stable and energetically suitable hole transporting buffer layers in tune with the electrode and photoactive materials of the solar cell stack. Here we have identified solution-processed Copper(I) iodide (CuI) thin films with low-temperature processing conditions as an effective hole-transporting layer (HTL) for a wide range of polymer:fullerene bulk heterojunction (BHJ) systems. The solar cells using CuI HTL show higher power conversion efficiency (PCE) in standard device structure for polymer blends, up to PCE of 8.8%, as compared with poly(3,4-ethylenedioxy-thiophene):poly(styrenesulfonate) (PEDOT:PSS) HTL, for a broad range of polymer:fullerene systems. The CuI layer properties and solar cell device behavior are shown to be remarkably robust and insensitive to a wide range of processing conditions of the HTL, including processing solvent, annealing temperature (room temperature up to 200 °C), and film thickness. CuI is also shown to improve the overall lifetime of solar cells in the standard architecture as compared to PEDOT:PSS. We further demonstrate promising solar cell performance when using CuI as top HTL in inverted device architecture. The observation of uncommon properties, such as photoconductivity of CuI and templating effects on the BHJ layer formation, is also discussed. This study points to CuI as being a good candidate to replace PEDOT:PSS in solution-processed solar cells thanks to the facile implementation and demonstrated robustness of CuI thin films.

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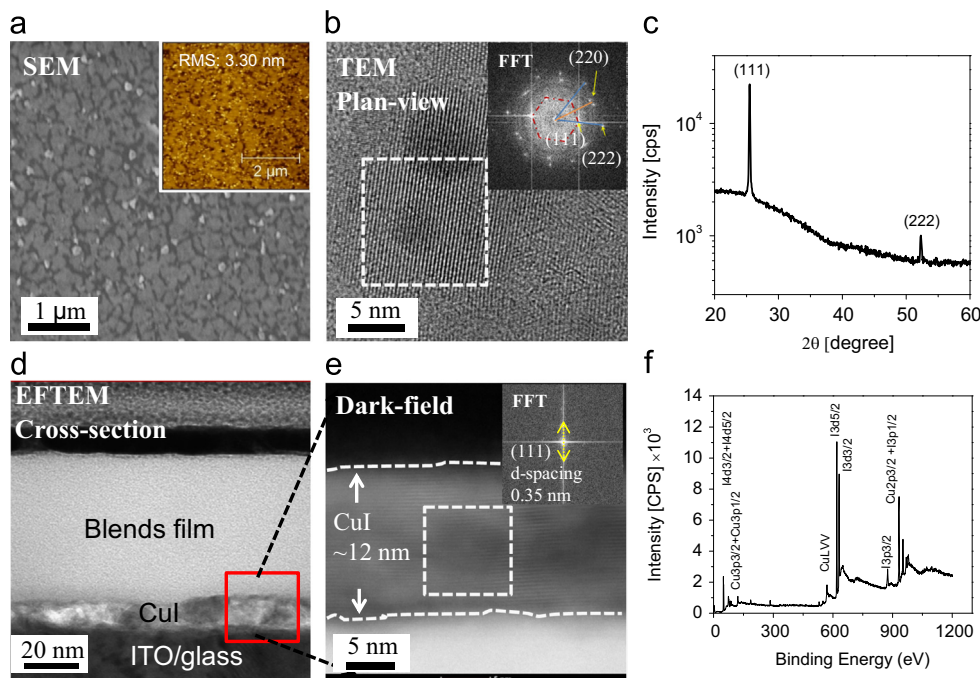
1 Introduction

3 Organic photovoltaics (OPV) have attracted tremendous
 4 attention in recent years because of the potential for
 5 manufacturing solution-processable, large area devices
 6 using potentially low cost, non-toxic materials and manu-
 7 facturing methods which have a reduced carbon footprint
 8 [1]. Efficient OPV devices require hole and electron trans-
 9 porting and buffer layers which prevent recombination of
 10 charges at the anode and cathode [2,3]. In the standard OPV
 11 device architecture, a transparent anode is traditionally
 12 coated with poly(3,4-ethylene dioxythiophene):(polystyrene
 13 sulfonic acid) (PEDOT:PSS), so far the most widely used
 14 hole-transporting layer (HTL) in OPVs [4]. However, the
 15 acidic and hygroscopic characteristics of PEDOT:PSS can
 16 cause indium loss from the indium tin oxide (ITO) transpar-
 17 ent anode, adversely affecting the device stability [5,6].
 18 Recently a few transition metal oxides (TMOs) have been
 19 reported as candidate replacements to PEDOT:PSS owing to
 20 the energy level compatibility with ITO and to the semi-
 21 conducting nature of the oxides. This includes materials
 22 such as Nickel oxide (NiO_x), [7] Molybdenum oxide (MoO_3),
 23 [8] tungsten oxide (WO_3), [9] and Vanadium oxide (V_2O_5)
 24 [10].

25 An important requirement for such TMO materials is their
 26 compatibility with solution-processing to enable large-area
 27 and high-throughput production of OPV devices. While most
 28 hole transporting oxides are typically vacuum deposited,
 29 some of them have been successfully shown to be solution
 30 processable, including NiO_x , [11] MoO_3 [12] and V_2O_5 [13].
 31 However, research is still directed toward identifying low

32 temperature means of producing such oxides, as many of
 33 them require process temperatures incompatible with roll-
 34 to-roll fabrication of OPV devices on plastic substrates. The
 35 oxide thin film synthesis methods can also be complex when
 36 compared with PEDOT:PSS thin film deposition and can yield
 37 highly variable properties depending upon the processing
 38 conditions and environments [14-17].

39 In this manuscript, we report a room-temperature solu-
 40 tion-processed inorganic pseudohalide HTL that is simple-
 41 to-process and produces high performance OPV devices over
 42 a broad range of preparation conditions. Copper(I) iodide
 43 (CuI) is inexpensive, abundant, and non-toxic. It is stable
 44 enough to be successfully vacuum-deposited as HTL, [18,19]
 45 yet can be easily solution-processed at room temperature
 46 [20,21]. CuI was successfully introduced in the past into
 47 dye-sensitized solar cells (DSSCs) by Tennakone and cow-
 48 orkers, [22] in which p-type CuI was used as hole conducting
 49 layer [21-27]. Vacuum-deposited CuI has also been used to
 50 template the stacking orientation of vacuum-deposited
 51 small-molecule thin films such as Zinc phthalocyanine
 52 (ZnPc) [28,29] and Copper phthalocyanine (CuPc), [30]
 53 as well as polymers such as poly(3-hexylthiophene) (P3HT)
 54 [31,32] and poly(4-(2-thiophenylthiophene)) (P42TA) in OPV
 55 applications [33]. Here, we characterize in detail solution-
 56 processed CuI thin films in terms of their microstructure,
 57 optical and electronic properties. We then successfully
 58 apply CuI as HTL to get power conversion efficiency (PCE)
 59 of up to 4.0% with P3HT:PC₆₀BM, 6.8% with PBDTTPD:
 60 PC₇₀BM, 8.0% with PTB7:PC₇₀BM, and 8.8% with PTB7-Th:
 61 PC₇₀BM in the standard device architecture, exceeding the
 62 performance achieved by PEDOT:PSS with the same material



63 **Figure 1** (a) SEM image of a solution-processed CuI film on ITO, inset showing atomic force micrographs of CuI film. (b) Plan-view
 64 bright-field high resolution TEM (HRTEM) on CuI film deposited on a TEM Copper grid. (c) X-ray diffraction (XRD) spectrum of the
 65 solution-processed CuI film (~ 12 nm) showing (111) and (222) crystalline planes in γ -phase. (d,e) cross-sectional TEM of a CuI film
 66 sandwiched between ITO and the organic photoactive layer in an actual device shown on the left. (f) X-ray photoelectron
 67 spectroscopy (XPS) of the solution-processed CuI film (~ 12 nm). The spectrum shows that the film mainly contains Copper and
 68 iodine.

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