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Semitransparent, thin metal grid-based hybrid electrodes for polymer solar cells



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ABSTRACT

A conducting polymer/thin Au grid hybrid electrode was investigated to replace an indium tin oxide (ITO) electrode in polymer solar cells (PSCs). Semitransparent, thin Au films were combined with transparent conducting PEDOT:PTS films (70 nm thickness, ~90% of transmission), to form Au grid/conducting polymer hybrid electrodes. The mixed self-assembled monolayers coated on the Au grids and glass substrate provided uniform and adherent coating of conducting polymer on the monolayer, achieving a low contact resistance of 0.6 Ω mm. This resulted in a robust PEDOT:PTS/Au grid hybrid structure.

Theoretical calculation showed the dependence of figure of merits (FM) on the filling ratio (= grid width/(grid spacing + grid width)) and Au thickness. In addition, grid spacing had an effect on the surface morphology of the conducting polymer; decreasing the grid spacing produced more flat surface of the overlayers, leading to enhanced performance of PSCs. The fabricated PSCs based on these hybrid electrodes showed that the best efficiency of 3.54%, comparable to that of devices based on an ITO electrode, was obtained at the filling ratio of 0.5 for 15 nm-thick Au electrodes, which was different from that predicted from the theoretical calculation, probably due to the grid spacing effects on the charge collection efficiency.

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1. Introduction

Polymer solar cells (PSCs) based on conjugated polymers (CPs) have received considerable attention for low-cost solar energy conversion [1–4], thanks to their potential advantages, such as the low cost fabrication of polymers-based devices, their compatibility with flexible substrates, and the facile control of optoelectronic properties of the CPs [5–7]. Indium tin oxide (ITO) has been widely used as a window electrode in PSCs devices [8]. However, ITO is

problematic due to the scarcity of indium element, vulnerability to acid or base, and susceptibility to ion diffusion into polymer layers [9,10]. In addition, the high temperature processing required for high conductive ITO and lack of mechanical flexibility limits its flexible applications [9,10]. In order to address these problems, alternative materials for transparent conducting electrodes are needed to replace ITO. There have been several reports on exploring conductive polymers [11,12], carbon nanotubes [13,14], graphene [15,16], metal nanowires [17–19], and metal grids/meshes [20–24] as potential replacements to ITO.

Among those alternative materials, hybrid electrodes consisting of high conductive, opaque metal grids combined

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with low conductive and transparent films [20,25,26], are of particular interest, because the structures are easily designed to tune the optical transparency and the electrical conductivity by changing the filling ratio and the metal thickness [18,20]. There are several reports on metal grid-based electrode for ITO-free PSCs [27–30] depicting the design of the structure and its effect on the photovoltaic conversion efficiency. The designed structures were usually made of opaque metal grids combined with a transparent, low conductive layer, thus optimizing the hybrid structures at very low filling ratio of metal grids, and providing the non-conformal coating of the overlayers [25,31,32]. It was also reported that metal grid electrodes with smaller grid separation facilitated better charge collection from in between the grids, leading to enhanced performance of PSCs.

In this study, we have used semitransparent thin Au grids of less than 25 nm, instead of thick and opaque metal, to form an Au grid/conducting polymer hybrid electrode. The use of very thin metal with conducting polymer caused a minimum step height of metal grids, facilitating the smooth surface of the conducting polymer. In addition, two different types of SAMs, octadecyltrichlorosilane (OTS, $(\text{CH}_3)_3\text{Si}(\text{CH}_2)_{17}\text{SiCl}_3$) and hexadecanethiol (HDT, $\text{HS}-(\text{CH}_2)_{15}-\text{CH}_3$) [33], selectively coated on Au and glass surface, respectively, were activated by UV illumination, to allow for the uniform coating of oxidants. This led to conformal and adherent PEDOT:PTS films on the Au grid and glass substrate. We have established simple equations for the dependence of transmission and sheet resistance on the filling ratio and Au thickness, and compared them with the measured values. Based on these equations, the hybrid structures were optimized for the highest figure of merits (FMs). The PSCs fabricated based on 15 nm-thick Au grid with different filling ratios ($f=0.5, 0.3, 0.1$) revealing the maximum efficiency of 3.54% were obtained at the filling ratio of 0.5 instead of 0.3 as predicted by the theoretical calculation, indicating that the grid spacing has an influence on the performance.

2. Experimental

Glass substrates were cleaned in piranha solution ($\text{H}_2\text{SO}_4:\text{H}_2\text{O}_2=4:1$) for 10 min, and exposed to UV illumination for 30 min. For the fabrication of Au grids, a lift-off technique was employed. Here, Au films were thermally evaporated on a micro-patterned photoresist, fabricated by standard photolithography, and the dissolution of the PR patterns with acetone produced the Au grids on the glass substrate. Fig. 1(a) shows three types of Au grid patterns fabricated with width and separation of (1) 5 μm and 5 μm (5 $\mu\text{m}/5 \mu\text{m}$), (2) 5 μm and 20 μm (5 $\mu\text{m}/20 \mu\text{m}$), (3) 5 μm and 45 μm (5 $\mu\text{m}/45 \mu\text{m}$), respectively. The Au grids sample was dipped into OTS solution (0.1 mM OTS in hexane) for 1 h, and then rinsed with hexane to remove the loosely bound monolayers [34], to self-assemble OTS monolayer on the glass surface. After that, the OTS-coated sample was dipped into HDT solution (1 mM HDT in ethanol) [35,36] for 12 h, and then rinsed with ethanol, to coat the HDT monolayer on the Au grids, and then, the sample was exposed to UV illumination to activate the surface of the

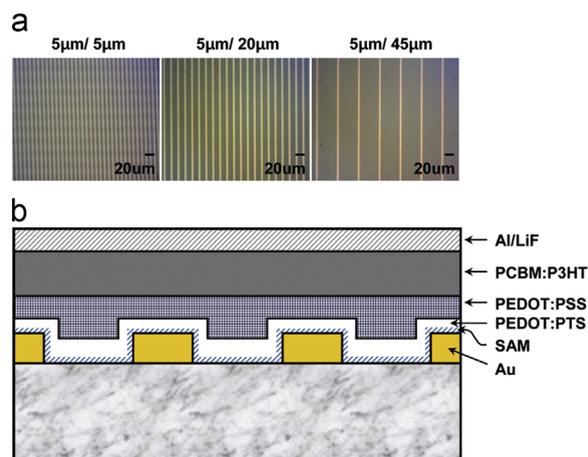


Fig. 1. (a) Three types of Au grid patterns fabricated as width and separation, with (1) 5 μm and 5 μm (5 $\mu\text{m}/5 \mu\text{m}$), (2) 5 μm and 20 μm (5 $\mu\text{m}/20 \mu\text{m}$), (3) 5 μm and 45 μm (5 $\mu\text{m}/45 \mu\text{m}$), respectively, and (b) a schematic of the fabricated PSCs.

monolayer [37,38]. After that, an iron(III) *p*-toluene sulfonate hexahydrate ($\text{Fe}(\text{PTS})_3$) solution (1.5 wt% pyridine ($\text{C}_5\text{H}_5\text{N}$), added to 20 wt% $\text{Fe}(\text{PTS})_3$ in ethyl alcohol) [39,40], was spread onto the activated surface, to form a uniformly coated oxidant layer. The $\text{Fe}(\text{PTS})_3$ coated monolayer sample was placed in an oven, where the 3,4-ethylenedioxythiophene (EDOT) was vaporized for 3 min at 80 $^\circ\text{C}$ to form a uniform thickness of PEDOT:PTS on the Au grids. This led to uniformly grown PEDOT:PTS (70 nm) on the Au grid lines.

The fabrication of PSCs started with the patterning of PEDOT:PTS conducting polymer on Au grids formed on the glass substrate: the OTS-coated glass and HDT-coated Au grid surface were UV-exposed, using a shadow mask to activate the SAMs surface, by creating surface functional groups, which allowed for the selective coating of $\text{Fe}(\text{PTS})_3$ solution on the UV-exposed monolayer surfaces and thus leading to the patterning of PEDOT:PTS conducting polymers. Subsequently, spin coating was conducted at 2000 rpm for 60 s, to produce ca. 70 nm-thick layers of poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate), PEDOT:PSS (Clevios, P Lot NR: GNU 1061, provided by H.C. Starck) on PEDOT:PTS, patterned on Au grids. Samples were thermally annealed at 110 $^\circ\text{C}$ for 10 min, while being covered with a shallow Petri dish. The active layers were then deposited by spin-coating (at 1500 rpm for 60 s) equal weights of P3HT (Rieke Metals) and PCBM (Nano-C) in *o*-dichlorobenzene (ODCB) to produce ca. 140 nm-thick bulk-heterojunction (BHJ) film; 15 nm LiF, and subsequently 100 nm aluminum cathodes were deposited using a thermal evaporator in which the base pressure was, through a shadow mask, producing devices with active areas of 0.06 cm^2 (0.3 cm (width) \times 0.2 cm (length)). The devices were thermally annealed at 150 $^\circ\text{C}$ for 10 min, while covered with the shallow glass Petri dish. The fabricated PSCs are shown in Fig. 1(b).

The optical transmittance and absorption spectra of the Au grids, and of the hybrids, (the triple layers) were measured using a UV-visible spectrometer (UV-3150, Shimadzu, Japan). The sheet resistance was measured

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