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# Indium-free multilayer semi-transparent electrodes for polymer solar cells



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#### ABSTRACT

We have explored the fabrication of indium-free electrodes for use in a PCDTBT:PC<sub>70</sub>BM organic photovoltaic (OPV) device, and compare different multilayer electrodes as the device anode. Two oxide/ metal/oxide structures were investigated that consisted of MoO<sub>3</sub>/Ag/MoO<sub>3</sub> (MAM) and TeO<sub>2</sub>/Ag/MoO<sub>3</sub> (TAM) multilayers. Using scanning electron microscopy measurements, we find that the electrode utilising a TeO<sub>2</sub> seed layer encouraged the growth of a more continuous silver layer at low film thickness relative to an MoO<sub>3</sub> seed layer, and thus combines enhanced optical transmission (by around 7%) with low sheet resistance (14  $\Omega/\Box$ ). This enhanced optical transmittance results in an increased short-circuit current in photovoltaic cells, with TAM-based devices having a power conversion efficiency around 6% higher than those fabricated using a comparable MAM electrode.

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#### 1. Introduction

In the decade to the end of 2014, world primary-energy consumption grew on average by 2.1% per year [1]. Against this background there is increasing concern about climate change, resource constraints and the security of energy supply, leading to great interest in renewable energy generation. Photovoltaics are expected to play a major role in this sector, and whilst silicon currently dominates the market [2], organic photovolatics (OPVs) are a promising route to lowering the cost of solar power. The solution processed nature of the photoactive layer allows for the possibility of high throughput production via roll-to-roll techniques such as spray coating or printing [3,4], and in addition OPVs can be fabricated on flexible, lightweight substrates such as PET [5–7] or even metal foils [8,9].

Due to its combination of high transmittance and good conductivity, indium tin oxide (ITO) is currently the ubiquitous material used as the transparent front electrode in OPVs. There are, however, concerns about the cost and scarcity of indium [7,10] and the embodied energy of the ITO layer [11,12]. Furthermore, high quality ITO is not easily compatible with the cheap, flexible

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polymer substrates that facilitate roll-to-roll production processes, as the brittle-nature of ITO results in a significant increase in sheet resistance after repeated bending [13,14]. Furthermore, such polymer substrates cannot tolerate the high temperatures that are commonly used in the deposition of highly conductive ITO. This second limitation leads to the sheet resistance of ITO on PET being around five times higher than that of similar ITO layers on glass [5,15–17].

There has thus been significant interest in the development of alternative materials for use as transparent conductive electrodes in OPVs. Here, possible replacements include the highly conductive polymer PEDOT:PSS in combination with metal grids [18,19], carbon nanosheets [20], silver nanowires [21–23] and oxide/metal/oxide stacks. In such oxide/metal/oxide multilayer electrodes, the initial oxide layer acts as a 'seed layer' for the subsequent growth of an ultrathin metal film. When used in an OPV, the metal film provides high lateral conductivity ensuring a suitably low series resistance, with the second oxide layer facilitating charge extraction or charge-blocking from the device [24,25]. In addition to their electronic functionality, such oxide layers also suppress reflection from the metal film and maximise optical transmittance [26–29].

In this paper, we explore two different oxide/metal/oxide stacks as the hole extracting electrode in an OPV device based on the polymer:fullerene blend PCDTBT:PC<sub>70</sub>BM. Whilst MoO<sub>3</sub>/Ag/MoO<sub>3</sub> (MAM) electrodes have previously been investigated for use

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**Fig. 1.** Transmittance spectra for multilayer MAM ((a) and (b)) and TAM ((c) and (d)) electrodes on glass with varying thickness of the silver film. All spectra are referenced to air, meaning that the spectra presented herein include reflection and absorption by the glass substrate. Average transmittance in the 350–700 nm range (e) and sheet resistance (f) for varying silver thicknesses are also shown. In all cases the oxide layers are all kept at a thickness of 10 nm.

in OPVs [30–33], TeO<sub>2</sub>/Ag/MoO<sub>3</sub> (TAM) electrodes have not. One study which investigated a TeO<sub>2</sub>/Ag/PEDOT:PSS electrode [34] suggested that a tellurium dioxide seed layer should lead to an enhanced short circuit current ( $J_{SC}$ ) in comparison with an equivalent MoO<sub>3</sub>/Ag/PEDOT:PSS structure, resulting from the larger (real-part) refractive index of TeO<sub>2</sub> [27]. Whilst the devices fabricated using such electrodes achieved a promising performance, the expected enhancement in  $J_{SC}$  resulting from the use of TeO<sub>2</sub> was not verified experimentally. Here we find that replacement of the MoO<sub>3</sub> seed layer by TeO<sub>2</sub> leads to significantly higher transmittance at low silver thickness, attributable to the formation

of a more continuous silver layer, as observed by scanning electron microscopy (SEM). When applied as the window electrode in PCDTBT:PC<sub>70</sub>BM polymer solar cells the improved transmittance of the TAM electrode results in an increased  $J_{SC}$  in comparison to devices based on a MAM electrode.

#### 2. Experimental

The TAM and MAM devices explored were deposited upon 1.1 mm thick quartz-glass substrates. As a control, identical devices

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