The optimal $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ electrode for organic solar cell application with high device-specific Haacke figure of merit

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Abstract

As a promising indium-free electrode, transparent composite electrode (TCE) $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ is an attractive alternative to indium tin oxide (ITO). The multilayer structure of $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ is deposited on glass substrate by room-temperature sputtering. The effects of deposition rate, thickness of Ag and TiO_2 layers on the electrical and optical properties of this TCE are presented. Results show that the Ag films tend to have fewer voids to form, if the Ag deposition rate is sufficiently high. The presence of fewer voids in the Ag layer results in higher transmittance and lower sheet resistance of the corresponding $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ structure. In order to increase the solar input to the $\text{P}_3\text{HT}:\text{PC}_{61}\text{BM}$ organic solar cell, the layer thicknesses of $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ electrodes are theoretically and then experimentally optimized, such that its optical transmittances are maximized across the wavelength range where the light absorption of $\text{P}_3\text{HT}:\text{PC}_{61}\text{BM}$ is highest. In this study, the device-specific Haacke figure of merit (FOM) of the $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ electrode is optimal with the use of 42 nm TiO_2 layers and 10 nm Ag layer. This optimal Haacke FOM that is two times higher than that of ITO electrodes. The conventional bulk heterojunction organic solar cell (OSC) fabricated on optimized $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ electrode shows high power conversion efficiency (PCE), almost 100% higher than PCE of OSC fabricated on ITO. Our finding indicates that $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ electrode can be specifically tailored to particular applications. It can be implemented in organic solar cell and has considerable potential to replace ITO in solar cell applications.

Keywords: device-specific Haacke figure of merit; $\text{TiO}_2/\text{Ag}/\text{TiO}_2$; Organic solar cell; Ag deposition rate.

1. Introduction

The transparent conducting electrodes are widely used in the applications of organic solar cells and light emitting devices [1-4]. Their electrical and optical properties significantly affect the device's efficiency. Indium-tin oxide (ITO) is one of the most widely used electrode materials due to its high optical transmittance and low resistivity [1-5]. However, the rare earth element indium in ITO limits its sustainability and is a major concern when considering the implementation of this material in potential devices [2, 5]. As a means to address this concern, transparent composite electrodes (TCEs) have been reported as a promising alternative to ITO-based electrodes due their indium-free composition, the low processing temperature, and superior figure of merit (FOM) [1-3].

A typical TCE is a three-layer structure: equivalent top and bottom transparent metal oxide (TMO) layers and an intermediate metal layer [1-15]. TiO_2 has a wide band gap of ~ 3.4 eV, and this renders it to be transparent in the visible light spectrum [2]. The high dielectric constant, strong mechanical and chemical stability and low fabrication cost make TiO_2 a suitable material for TMO layer in TCE [2]. Ag is an ideal candidate for the metal layer in TCE [16]. Its real part of the refractive index is the one of the lowest among metals and this will help minimize the light absorption of metal layer. Moreover, the resistivity of Ag is one of the lowest of the noble metals, which will help improve the resistivity of TCE. As a result, $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ is a potentially promising candidate of TCEs. To be a desired electrode, the layer thickness of TCE has to be optimized such that its transmittance and resistance are desirable. The optimization of the transmittance of TCE across the entire visible light region has been studied by several investigators [1-3,5,8,10,14]; however, few report actually tailoring the indium-free electrode's transmittance to the organic solar cell's absorption spectrum.

In our study, multilayer structures of $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ are fabricated on glass substrates by room-temperature sputtering. Initially, the effect of Ag deposition rate on TCE's optical transmittance and sheet resistance is studied. Once the optimal Ag deposition rate is obtained that gives a relatively low sheet resistance and high transmittance in TCEs, then the thicknesses of TiO_2 and Ag layers are optimized, such that the maximum

transmittance of the $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ electrode is within the specific wavelength region (450 nm - 600 nm). Results demonstrate that the device-specific Haacke FOM for optimized $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ is significantly higher than that of ITO-based structures. Furthermore, the conventional bulk heterojunction organic solar cells, Anode/ $\text{MoO}_3/\text{P}_3\text{HT}:\text{PC}_{61}\text{BM}/\text{LiF}-\text{Al}$, are fabricated on optimized $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ electrode and ITO. Their *J-V* curves and device parameters are compared and studied. The PCE of $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ based solar cells is significantly higher than that of ITO based OSC.

2. Experiment:

2.1 Electrode fabrication and characterization

The multilayer structure of $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ was sputter deposited on glass substrate at room temperature with a base pressure of 8×10^{-7} Torr. The bottom TiO_2 layer was deposited by RF sputtering of a TiO_2 target (99.9% purity) with a power of 66 W and Ar pressure of 2.5 mTorr. Sequentially, the middle Ag layer was deposited by DC sputtering of an Ag target (99.9% purity) at Ar pressure of 4 mTorr. Various DC power were used to achieve different deposition rate of Ag. Finally, the top TiO_2 layer was deposited on Ag layer using the same deposition condition as the bottom TiO_2 . The thickness of each layer was confirmed by the variable angle spectroscopic ellipsometer measurement and WVASE 32 software analysis. The optical transmittance was measured by Cary 5000 UV-Vis spectrometer over 300 nm to 800 nm. The sheet resistance was determined by four-point probe analysis. Secondary electron microscopy (SEM) was conducted to evaluate the surface morphology of Ag layers that were deposited on $\text{TiO}_2/\text{glass}$ using different sputtering rates.

2.2 OSC fabrication

2.2.1 Reagent and materials

The electrode donor material regioregular poly (3-hexylthiophene) (P_3HT) was from Reike Metals, Inc. The electron acceptor material [6,6]-phenyl C_{61} butyric acid methyl ester (PC_{61}BM) and 1,2-dichlorobenzene were from Sigma Aldrich.

2.2.2 Device fabrication and testing

The hole transfer layer, MoO₃ with a thickness of 14 nm, was deposited on the TiO₂/Ag/TiO₂ electrode by thermal evaporation of MoO₃ powder (99.998% purity). The electron donor P₃HT and electron acceptor PC₆₁BM with a weight ratio of 1:0.8 were dissolved in the 1,2-dichlorobenzene to make total 36 mg/ml solution. The solution was stirred at room temperature for overnight in nitrogen-filled glove box. Then the blended solution was spin coated at 600 rpm for 60s, followed by drying at room temperature for overnight, then baked at 130 °C for 30 min in glove box. In the end, the electron transfer layer LiF of 0.1 nm and metal cathode Al of 96 nm were sequentially deposited by thermal evaporation using a shadow mask that defines an active area of 0.2 cm². Similarly, the control sample was fabricated using ITO as the anode.

The current density-voltage (*J-V*) curve were measured under simulated AM 1.5 global solar irradiation (100 mW/cm²) using a xenon-lamp solar simulator (Spectra Physics, Oriel Instruments, USA).

3. Results and Discussion

The deposition rate of Ag affects the morphology of Ag layer [17], which in turn impacts the optical transmittance of TCE [18], due to the light scattering effect at the Ag and TiO₂ interface [19, 20] or light absorption caused by localized surface plasmons in the Ag surface [21]. To study the effect of the Ag deposition rate on TCE's optical transmittance and sheet resistance, three sets of TCE samples with the same layer thickness are prepared using various Ag deposition rates (0.03 nm/s, 0.18 nm/s, and 0.40 nm/s). Their optical transmittance and sheet resistance are compared in Fig. 1.

The TCEs with high Ag deposition rate (0.18/s and 0.40 nm/s) shows almost identical transmittance spectrum and similar sheet resistance values. On the other hand, TiO₂/Ag/TiO₂ TCE prepared with a low Ag deposition rate (0.03 nm/s) has two times higher sheet resistance and relatively lower transmittance in the wavelength range of 400 nm to 600 nm.

Fig. 1 (a) The optical transmittance of (30 nm) TiO₂/ (10 nm) Ag/ (30 nm) TiO₂ fabricated with various Ag deposition rate. The glass substrate is taken as the reference. (b) The sheet resistance as a function of Ag deposition rate from (30 nm) TiO₂/ (10 nm) Ag/ (30 nm) TiO₂

To investigate the reason for the differences and similarities in the transmittance and sheet resistance of the above TCEs, the morphology of Ag layers deposited by different rates is further studied with the use of SEM. The SEM micrographs of Ag films are shown in Fig. 2, and indicate that all three deposition rates result in non-continuous films that contain voids. The voids are relatively large, but in a small amount when the Ag deposition rate is 0.18 nm/s and 0.40 nm/s. This similarity in morphology of Ag film deposited at 0.18 nm/s and 0.4 nm/s explains their similarity in their transmittance spectra and sheet resistances. On the other hand, many tiny voids are evenly distributed in the Ag film when the deposition rate is low (0.03 nm/s). These voids tend to disrupt the continuous path of the flowing current. As a result, the sheet resistance of TCE is the highest when its Ag layer is deposited at 0.03 nm/s. Similarly, due to this relatively high voids density in Ag layer deposited at low rate (0.03 nm/s), its TCE's transmittance is lower in the wavelength between 450 nm and 600 nm. Based on the comparison above, the TCEs with the Ag layers deposited at 0.18 nm/s and 0.40 nm/s both shows the optimal sheet resistance and optimal transmittance between 450 and 600 nm. As a result, 0.4 nm/s is selected as the optimal deposition rate for Ag layer in our study.

For electrodes used in organic solar cell and light emitting diodes, optimization of the average transmittance over the entire visible light spectrum

Fig. 2 SEM micrographs for 10 nm Ag film on TiO₂/glass deposited at rate of a) 0.03 nm/s, b) 0.2 nm/s, and c) 0.4 nm/s.

might not be able to satisfy the specific needs of the devices. For example, the light absorption peak of the active layer in $P_3HT:PC_{61}BM$ organic solar cell is between 450 nm to 600 nm [22]. Maximizing the average transmittance of TCE across 450 nm to 600 nm can specifically increase the light input to the organic solar cells. Before the experimental optimization of layer thickness to achieve high transmittance over the wavelength of 450 nm to 600 nm, the numerical simulation of TCE's transmittance via transfer-matrix method is used to estimate the optimal thickness of TiO_2 and Ag layers [21]. Through the simulation, the range of desired thickness for experimental fabrication is narrowed, which facilitates the optimization process. Our previous study has shown that the percolation threshold (where the Ag islands begin to connected with each other and form network) of Ag film is approximately 9.5 nm in $TiO_2/Ag/TiO_2$ structures on flexible substrate [2]. Hence, the Ag layer is fixed at 10 nm in the simulation, then the TiO_2 layers are varied from 30 nm to 50 nm to determine the optimal thickness. The simulated dependence of transmittance spectrum on the TiO_2 layer thickness is shown in Fig. 3(a). The red shift occurs at the absorption edge when the top and bottom TiO_2 thickness increases. Results show that TiO_2 thickness between 35 nm to 45 nm gives the optimal transmittance over 450 nm and 600 nm.

Fig. 3 (a) The simulated transmittance of TiO_2/Ag (10 nm)/ TiO_2 with different TiO_2 thicknesses of 30 nm, 35 nm, 40 nm, 45 nm and 50 nm. (b) The simulated transmittance of TiO_2 (40 nm)/Ag/ TiO_2 (40 nm) with different Ag thickness of 8 nm, 10 nm, 12 nm and 14 nm.

To determine the optimal Ag thickness, the TiO_2 layers are fixed at the 40 nm and the silver thicknesses vary from 8 nm to 14 nm. The simulated dependence of transmittance on Ag layer thickness is shown in Fig. 3(b). In this case, the TCE having 10 nm Ag layer shows the optimal transmittance over 450 to 600 nm.

It is noticed that the simulation results can only provide the first-order approximation of the

transmittance's dependence on the layer thicknesses. The actual transmittance of the fabricated TCE might deviate from simulated transmittance due to the discontinuity of the metal film [21]. The optimal layer thickness obtained by simulation is ideal for continuous films; however for the non-continuous film in this study, the thicknesses obtained by simulation are in the range of optimal thickness values. Hence, the layer thicknesses near the optimal thickness are selected for fabrication.

Fig. 4 The optical transmittance spectra for $TiO_2/Ag/TiO_2$ (TAT) on glass as a function of TiO_2 thickness and Ag thickness. Glass substrate as the reference.

Based on the transmittance simulation results, two different thicknesses (35 nm and 42 nm) of TiO_2 are selected for experimental fabrication. In addition to the consideration of optical transmittance, the sheet resistance of TCE has to be taken into account in order to maximize the Haacke FOM [23] across 450 nm to 600 nm. The sheet resistance of TCE is mainly determined by metal layer [16]. A thicker metal film in the TCE results in a lower sheet resistance and unfortunately a lower transmittance as well. As a result, even 10 nm is the optimal Ag thickness for achieving high transmittance, the Ag thicknesses of 8.5 nm and 13 nm are also selected for TCE fabrication in order to achieve a trade-off between sheet resistance and optical transmittance.

Six sets of TCEs are fabricated. As the alternative to ITO, their properties are compared with ITO. The optical transmittance spectra are shown in Fig. 4. The sheet resistances are shown in Table I. From these

Table I The sheet resistance, average transmittance over 450 nm and 600 nm and the corresponding Haacke FOM of ITO and TiO₂/Ag/TiO₂ that having different TiO₂ and Ag thickness.

Sample	R_s (Ω)	T_{ave}	Device specific Haacke FOM (sq/ Ω)
ITO	18.9	0.96	35.2×10^{-3}
TiO ₂ (35 nm)/Ag(8.5 nm)/ TiO ₂ (35 nm)	5.62	0.85	35.0×10^{-3}
TiO ₂ (35 nm)/Ag(10 nm)/ TiO ₂ (35 nm)	4.33	0.84	40.4×10^{-3}
TiO ₂ (35 nm)/Ag(13 nm)/ TiO ₂ (35 nm)	3.74	0.74	13.2×10^{-3}
TiO ₂ (42 nm)/Ag(8.5 nm)/ TiO ₂ (42 nm)	5.95	0.85	33.1×10^{-3}
TiO ₂ (42 nm)/Ag(10 nm)/ TiO ₂ (42 nm)	4.48	0.89	69.6×10^{-3}
TiO ₂ (42 nm)/Ag(13 nm)/ TiO ₂ (42 nm)	3.50	0.82	39.3×10^{-3}

results, the TCEs show an overwhelming advantage over ITO in terms of sheet resistance; where, the sheet resistance values of the TCEs are typically at least three times lower than that of the ITO.

The Haacke figure of merit (ϕ_{TC}) takes into account of both sheet resistance and transmittance, and it is calculated as following [23]:

$$\phi_{TC} = \frac{T_{av}^{10}}{R_s} \quad (1)$$

where the T_{av} is the average transmittance, R_s is the sheet resistance.

Since the TCEs are used in the application for P₃HT:PC₆₁MB organic solar cell (OSC), we are interested in the wavelength range between 450 nm and 600 nm (where the OSC is most efficient [22]). The Haack FOM in this wavelength range is defined as device-specific Haacke FOM here. The average transmittance over 450 nm and 600, and device-specific Haacke figure of merit are compared in Table I. Among all the TCEs and ITO, TiO₂ (42 nm)/Ag (10 nm)/TiO₂ (42 nm) proposes the highest device-specific Haacke FOM. More importantly, its device-specific Haacke FOM is two times higher than that of ITO. In conclusion, we have demonstrated that the optimized TiO₂/Ag/TiO₂ shows much better performance in regards to the Haacke FOM.

To study the performance of TiO₂/Ag/TiO₂ when it is employed in OSC, bulk heterojunction OSC were fabricated on the optimized TiO₂/Ag/TiO₂ anode. For comparison, control sample was fabricated using ITO as the anode.

It was noticed that the conventional hole transfer layer (HTL), poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), attacked oxide anode due to its acidic nature [24, 25]. To avoid this, a thermal evaporated MoO₃ layer was used as the alternative to PEDOT:PSS in our fabrication [25-28]. After the devices fabrication, the J - V curves of ITO based and TiO₂/Ag/TiO₂ based OSCs were measured and shown in Fig. 5. The device parameters of the OSCs were compared in Table II.

Results show that the PCE of TiO₂/Ag/TiO₂ based OSC is 100% higher than that of the ITO based OSC. Moreover, the J_{sc} in TCE based OSC is 45% higher than that of ITO based OSCs. In fact, J_{sc} is a measurement of generation and collection of photo-generated carriers [24]. The Ohmic contact between electrode and active layer affects J_{sc} [24]. Hence, the small series resistance (R_{series}) can result in increased J_{sc} , which can also lead to improvement in PCE [24, 29]. According to Table II, the R_{series} of TCE based OSCs is smaller than that of ITO based OSCs, which is attributed to the lower sheet resistance of TiO₂/Ag/TiO₂ anode. The significantly high shunt resistance (R_{sh}) in TCE based solar cell leads

to low leakage current and contribute to its high J_{sc} . As a result, J_{sc} of TCE based OSC is higher.

Fig. 5 Current density- (J - V) characteristics of bulk-heterojunction solar cells on ITO and $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ electrode.

Table II. Device parameters of Anode/ $\text{MoO}_3/\text{P}_3\text{HT}:\text{PC}_{61}\text{BM}/\text{LiF-Al}$ solar cells with ITO and $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ as the anode, respectively.

Anode	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PEC (%)	R_{series} (Ωcm^2)	R_{sh} (Ωcm^2)
ITO	0.44	6.04	0.44	1.08	14.18	241.65
$\text{TiO}_2/\text{Ag}/\text{TiO}_2$	0.51	8.78	0.49	2.18	11.06	718.65

The J_{sc} of ITO based OSC is only $6.04 \text{ mA}/\text{cm}^2$. Apart from the relatively high R_{series} of ITO based OSC, MoO_3 might be responsible for such a low J_{sc} . It has been reported that MoO_3 is an ideal hole transfer layer to replace PEDOT:PSS [24-26, 28]. However, when MoO_3 is used in our ITO based OSC, its J_{sc} is much lower than that of ITO based OSC using PEDOT:PSS as reported by our group [30]. The HTL adjusts the band alignment between donor material and anode [24, 25]. A good HTL should have a work function that aligns well the HOMO energy level of the donor such that it forms a desired Ohmic contact between donor and anode, and therefore improves the hole extraction and transportation [24]. The work function and energy level of our $\text{ITO}/\text{MoO}_3/\text{P}_3\text{HT}:\text{PC}_{61}\text{BM}/\text{LiF-Al}$ will be future studied regarding energy level alignment between ITO, MoO_3 and active layer.

4. Conclusion

As the alternative electrodes for organic solar cell (OSC) devices, $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ TCE's thicknesses were optimized for that specific wavelength range of 450 nm to 600 nm. This range corresponds to the light absorption peak of the active layer in $\text{P}_3\text{HT}:\text{PC}_{61}\text{BM}$ OSC. The morphology of the Ag layer in TCE impacts the transmittance and sheet resistance of TCE. Hence, the Ag deposition rate that affects the morphology of the Ag layer was studied in the beginning. The high deposition rates, 0.18 nm/s and 0.40 nm/s, both result in less void density in Ag layer, leading to lower sheet resistance and higher transmittance in the TCEs, as compared to low deposition rate (0.03 nm/s). Therefore, the optimal Ag deposition rate is determined as 0.40 nm/s. To increase the light input to the $\text{P}_3\text{HT}:\text{PC}_{61}\text{BM}$ layer, the Haacke FOM of $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ electrode is specifically optimized across the wavelength from 450 nm to 600 nm. Initially, theoretical simulations are used to estimate the optimal layer thicknesses for achieving highly transparent TCEs. It narrows the possible layer thicknesses for experimental fabrication and facilitates experimental optimization. In experimental fabrication which varies the layer thickness of Ag and TiO_2 near the optimal values, it shows that TiO_2 (42 nm)/Ag(10 nm)/ TiO_2 (42 nm) has the device-specific Haacke FOM as high as $69.6 \times 10^{-3} \text{ sq}/\Omega$, which is two times better than that of the ITO-based structure.

The performance of TCE on conventional bulk heterojunction solar cell is further studied by fabricating Anode/ $\text{MoO}_3/\text{P}_3\text{HT}:\text{PC}_{61}\text{BM}/\text{LiF-Al}$ on the optimized $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ electrode and ITO. It turns out that MoO_3 is a desirable HTL for $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ electrode. $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ electrode based OSCs show higher PCE than ITO based OSCs.

In conclusion, $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ based TCEs can be successfully integrated into the solar cell application. Its high device-specific Haacke FOM make it a promising alternative to ITO in terms of organic solar cell application. More importantly, due to the adjustable nature of TCE's properties, TCE parameters can even be tailored for other specific photovoltaic applications.

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Table 1

Table 1 The sheet resistance, average transmittance over 450 nm and 600 nm and the corresponding Haack FOM of ITO and TiO₂/Ag/TiO₂ that having different TiO₂ and Ag thickness

Sample	R_s	T_{ave}	Device specific Haack FOM (sq/Ω)
ITO	18.9	0.96	35.2×10^{-3}
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TiO ₂ (35 nm)/Ag(10 nm)/ TiO ₂ (35 nm)	4.33	0.84	40.4×10^{-3}
TiO ₂ (35 nm)/Ag(13 nm)/ TiO ₂ (35 nm)	3.74	0.74	13.2×10^{-3}
TiO ₂ (42 nm)/Ag(8.5 nm)/ TiO ₂ (42 nm)	5.95	0.85	33.1×10^{-3}
TiO ₂ (42 nm)/Ag(10 nm)/ TiO ₂ (42 nm)	4.48	0.89	69.6×10^{-3}
TiO ₂ (42 nm)/Ag(13 nm)/ TiO ₂ (42 nm)	3.50	0.82	39.3×10^{-3}

Table 2

Table II. Device parameters of Anode/MoO₃/P₃HT:PC₆₁BM/LiF-Al solar cells with ITO and TiO₂/Ag/TiO₂ as the anode, respectively.

Anode	V _{oc} (V)	J _{sc} (mA/cm ²)	FF (%)	PEC (%)	R _{series} (Ωcm ²)	R _{sh} (Ωcm ²)
ITO	0.44	6.04	0.44	1.08	14.18	241.65
TiO ₂ /Ag/TiO ₂	0.51	8.78	0.49	2.18	11.06	718.65