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Organic solar cells with carbon nanotube network electrodes

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We fabricated flexible transparent conducting electrodes by printing films of single-walled carbon nanotube (SWNT) networks on plastic and have demonstrated their use as transparent electrodes for efficient, flexible polymer-fullerene bulk-heterojunction solar cells. The printing method produces relatively smooth, homogeneous films with a transmittance of 85% at 550 nm and a sheet resistance ($R_s$) of 200 $Ω/□$. Cells were fabricated on the SWNT/plastic anodes identically to a process optimized for ITO/glass. Efficiencies, 2.5% (AM1.5G), are close to ITO/glass and are affected primarily by $R_s$. Bending test comparisons with ITO/plastic show the SWNT/plastic electrodes to be far more flexible. © 2006 American Institute of Physics. [DOI: 10.1063/1.2209887]
ITO, where $T$ is mainly determined by the reflectance of the film, $T<100\%$ for SWNT films is due mainly to absorbance in the film. The network can be tuned for an appropriate thickness, or density, and thus to an optimal optical transparency and sheet resistance. Consequently such networks, or films, can be used as transparent electrodes for a variety of applications. For solar cells, the optimal trade-off between $T$ and $R_s$ will vary depending on the intrinsic current-voltage characteristics of the device.

We have used 125-μm-thick PET flexible substrates coated with 30-nm-thick SWNT network films ($T=85\%$, $R_s=200\,\Omega/\square$) to fabricate solar cells with both poly[2-methoxy-5-(3′,7′-dimethoxystyryl)-1,4-phenylene vinylene] (MDMO-PPV)/1-(3-methoxycarbonyl)-propyl-1-phenyl[6,6]C$_{61}$ (PCBM) and poly(3-hexylthiophene) (P3HT)/PCBM. Prior to depositing the active layer, a layer of poly(3,4-ethylenedioxythiophene)/poly(styrene sulfonate) (PEDOT:PSS) was spin cast on the SWNTs at 1000 rpm and then placed directly on a 110 °C hot plate and annealed for 20 min. Consistent results were obtained when either the PEDOT:PSS solution was applied on the surface and let free to diffuse several minutes before the spin-coating operation or in order to fill in the open porosity of the SWNT film or when a PEDOT:PSS/1:1 mix was used to improve the wetting. The PEDOT:PSS reduced $R_s$ by approximately 20% to 160 $\Omega/\square$. The same PEDOT:PSS film spun on glass with no nanotubes (95 nm thick) had a sheet resistance of 15 k$\Omega/\square$, which is too high of a resistance to account for the drop in $R_s$ simply due to parallel conduction. Therefore, two possible explanations are a reduction in the resistance between conducting nanotubes and/or doping of any semiconducting nanotubes. Figure 1(b) shows the SWNT films after being coated with PEDOT:PSS and the resulting low roughness. Samples were then transferred to an inert glovebox where a solution of MDMO-PPV/PCBM in a 1:4 weight ratio or P3HT/PCBM (Ref. 11) in a 1:0.8 weight ratio (10 mg P3HT/ml) in chlorobenzene was spin cast at 700 rpm. Finally, a 100 nm Al top electrode was evaporated under high vacuum at a rate of 0.1 nm/s. P3HT devices were annealed on a 120 °C hot plate for 10 min and had a postannealed optical density of 0.5. The overlap of the top and bottom electrodes defined a device of $1 \times 4$ mm$^2$. The results are summarized in Table I for the flexible devices as well as a control device on ITO coated glass (15 $\Omega/\square$). The measured open circuit voltages ($V_{oc}$) suggest that, as proposed by Frohne et al., the $V_{oc}$ is driven by the work function of the PEDOT:PSS layer rather than by the carbon nanotubes. Figure 3 shows the current-voltage curves for P3HT/PCBM cells under simulated AM1.5G conditions.

The devices fabricated using SWNT networks operate almost identically to those for ITO coated glass with the exception of the fill factor (FF), which is likely attributable to the relatively high series resistance due to $R_s$. For solar cell applications, the TC must be as transparent and conductive as possible. The effect of the transparency is straightforward and the effect of the sheet resistance can be estimated to first order by Eq. (1), if the assumption is made that the voltage drop in the electrode is small enough that the current production is approximately homogeneous across the cell. For a cell of width $w$ and length $l$, where current is collected at one of the edges with length $l$, the power loss ($P_{loss}$) due to $R_s$ is given by (see, e.g., Goetzberger et al.):

$$P_{loss} = (jl)^2R_{eff},$$

where $R_{eff} = R_s w / 3l$. For example, to limit the fraction of power lost, $P_{loss} / (jlwV)$, to 10% for a cell with current density $j = 8$ mA/cm$^2$ and voltage $V = 500$ mV at the maximum power point and $R_s = 10 \Omega/\square$, $w$ must be less than 1.4 cm, and if $R_s = 160 \Omega/\square$, $w$ is limited to 0.34 cm. For comparison, commercial Si solar cells typically have an emitter layer...
TABLE I. Current density-voltage characteristics of devices under AM1.5G simulated illumination. (Oriel solar simulator, 100 mW/cm². A spectral mismatch factor of 1.25 was calculated using the measured photocurrent action spectra for our device and for the silicon photodiode, combined with the ideal AM1.5G spectrum and the actual spectrum produced by our simulator. Note that not including this factor would result in an approximately 20% overestimation of the efficiency of our devices.)

<table>
<thead>
<tr>
<th>Device structure</th>
<th>$j_{oc}$ (mA/cm²)</th>
<th>$V_{oc}$ (mV)</th>
<th>FF</th>
<th>Efficiency (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PET/SWNTs/PEDOT:PSS/MDMO-PPV:PCBM/Al</td>
<td>3.6</td>
<td>800</td>
<td>0.42</td>
<td>1.2</td>
</tr>
<tr>
<td>PET/SWNTs/PEDOT:PSS/P3HT:PCBM/Al</td>
<td>7.8</td>
<td>605</td>
<td>0.52</td>
<td>2.5</td>
</tr>
<tr>
<td>Glass/ITO/PEDOT:PSS/P3HT:PCBM/Al</td>
<td>8</td>
<td>610</td>
<td>0.61</td>
<td>3</td>
</tr>
</tbody>
</table>

with a sheet resistance of $40–100 \Omega/\square$ (Ref. 14) with a current density of three to four times as much as today’s organic solar cells. However, by screen printing thin contact fingers with $w=0.2$ cm, losses from shadowing and $R_s$ effects are kept to less than 10%.13 For our P3HT:PCBM devices using SWNT/plastic anodes ($w=0.4$ cm, $j=5.8$ mA/cm², $V=430$ mV, and $R_s=160 \Omega/\square$), Eq. (1) predicts the fraction of power lost to be 12%. This agrees well with the observed loss of 17% using devices fabricated on ITO/glass anodes as the reference.

The stability of devices fabricated on the SWNT/PET films was much greater than devices on ITO/PET during simple bending tests. SWNT devices could be folded over (inducing compressive or tensile strain) down to radii of curvature of $\sim5$ mm with no degradation in power efficiency and radii of $\sim1$ mm with a 20%–25% loss in efficiency.

Interestingly, annealing for 5 min at 130 °C completely restored the efficiency in two out of three devices. The mechanisms involved in the failure or the restoration have not been investigated. In comparison, devices using ITO ($R_s=40 \Omega/\square$) on the same 125-μm-thick PET substrate began to fail at a radius of 1 cm and completely failed at 5 mm with fractures in the ITO visible to the eye.

These initial results suggest that with further optimization such anodes may offer a direct alternative to ITO and other transparent conducting oxides, in particular, under circumstances where mechanical flexibility is desirable. We have demonstrated before that the performance of transparent SWNT transistors remains virtually unchanged upon mechanical distortions15 and here we have demonstrated that the efficiency of a working solar cell also is virtually unchanged. Furthermore, we believe that this represents only the beginning for the performance of SWNT TCs. Little has been done to investigate and control such effects as contact resistance between nanotubes, order of the nanotubes, doping, or use of only metallic nanotubes to name a few. The high mobility of individual nanotubes16,17 indicates that significant improvement of the performance characteristics is possible through network optimization.

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