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Encapsulation of pentacene/C$_{60}$ organic solar cells with Al$_2$O$_3$ deposited by atomic layer deposition

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Organic solar cells based on pentacene/C$_{60}$ heterojunctions were encapsulated using a 200-nm-thick film of Al$_2$O$_3$ deposited by atomic layer deposition (ALD). Encapsulated devices maintained power conversion efficiency after exposure to ambient atmosphere for over 6000 h, while devices with no encapsulation degraded rapidly after only 10 h of air exposure. In addition, thermal annealing associated with the ALD deposition is shown to improve the open-circuit voltage and power conversion efficiency of the solar cells. © 2007 American Institute of Physics. [DOI: 10.1063/1.2751108]

Photovoltaic cells made from organic materials are promising alternative energy sources that have been largely fabricated and tested in controlled laboratory environments, but such cells will be subjected to a variety of uncontrollable conditions when deployed for real world applications. In fact, exposure to simply oxygen and water is well known to cause degradation in many organic electronics devices including organic photovoltaic (OPV) cells.$^{1–3}$ Therefore, adequate encapsulation is typically required for many of the material systems that are currently under study for OPV cells. The desired encapsulation method must not only block oxygen and water effectively, but it should also be thin and lightweight to take full advantage of the potentials of OPV cells in practical applications. One candidate is a film of Al$_2$O$_3$ deposited directly on top of the devices by atomic layer deposition (ALD). ALD is a chemical vapor deposition technique that involves the cycling of alternate precursor gases into a chamber to react on the surface of samples and grow high quality, conformal films.$^4$ Al$_2$O$_3$ films by ALD have been used alone$^5$ and in multilayer stacks$^6,7$ to encapsulate organic light-emitting diodes (OLEDs) and have been demonstrated as potential barrier films for the devices. In addition, water vapor transmission rates of 1.7 × 10$^{-5}$ g/m$^2$·day have been measured through Al$_2$O$_3$ films grown on polyethylene naphthalate substrates.$^8$ The research presented here describes the encapsulation of organic solar cells based on pentacene/C$_{60}$ heterojunctions with a layer of Al$_2$O$_3$ deposited by ALD and its effect on the OPV performance.

Organic solar cells were fabricated as previously reported$^9$ with a device geometry comprised of indium tin oxide/pentacene (50 nm)/C$_{60}$ (40 nm)/bathocuproine (BCP) (7 nm)/Al and an active device area of ~0.1 cm$^2$. After fabrication, devices were briefly exposed to ambient air when being transferred to the ALD deposition chamber (Savannah100, Cambridge Nanotech, Inc.), where approximately 200 nm of Al$_2$O$_3$ was deposited on top of the completed devices. ALD was performed at a temperature of 100 °C with pulses of H$_2$O for 100 ms and trimethylaluminum (TMA) for 100 ms. The chamber was pumped for 20 s after H$_2$O pulses and for 15 s after TMA pulses for an overall growth rate of ~10 nm/h. Figure 1(a) shows a schematic of the device structure. After the ALD process, samples were stored in ambient air in a case loosely wrapped with aluminum foil to minimize eventual light-induced degradation. For testing, the samples were loaded in a nitrogen-filled glovebox where a 175 W Xenon lamp (ASB-XE-175EX, CVI) was used as a broadband light source (350–900 nm) with an irradiance of approximately 100 mW/cm$^2$.

The electrical characteristics of six devices on two substrates were measured in the dark and under illumination before and after undergoing ALD. Figure 1(b) shows the...
characteristics for the device with the highest efficiency. When averaged over six devices, the open-circuit voltage \( V_{OC} \) and the fill factor FF both increased from 350±3 mV and 0.53±0.01 to 445±3 mV and 0.56±0.02 after ALD. Short-circuit current density \( J_{SC} \) remained essentially constant. Because of slight variations in irradiance \( I_{L} \) during measurements carried out over a long period of time, all \( J_{SC} \) values presented here are normalized to 100 mW/cm\(^2\) by

\[
J_{SC} = \frac{100 \text{ mW/cm}^2}{I_{L}}. \tag{1}
\]

Overall, power conversion efficiency \( \eta \) under the broadband light source increased from 2.0±0.1% before encapsulation to 2.7±0.2% after. By overlapping the external quantum efficiency with the solar spectrum and integrating, \( \eta \) after the ALD process is estimated to be 1.4±0.1% under AM1.5 G illumination. The electrical characteristics for all six devices tested are shown in the inset of Fig. 1(b) and show a reproducible trend for all of the devices that underwent ALD. These changes are very similar to those observed when annealing similar devices on a hot plate for 1 min at 200 °C.\(^{10,11}\)

Next, the effectiveness of the Al\(_2\)O\(_3\) film as an encapsulation layer was tested over time. In addition to Al\(_2\)O\(_3\), UV curable epoxy (NOA 65, Norland Products) was also tested as an encapsulant. The epoxy was drop cast on top of devices without any other encapsulation and on top of a device that already had an Al\(_2\)O\(_3\) layer, and the epoxy was then cured with UV light. Therefore, three types of encapsulation were tested and compared in this experiment: Al\(_2\)O\(_3\) deposited by ALD, UV epoxy, and UV epoxy on top of Al\(_2\)O\(_3\) deposited by ALD. A reference sample with no encapsulation was also tested during this experiment.

Table I lists the initial performance characteristics of the cells presented here after encapsulation but before they were exposed to ambient air. As shown in Fig. 3 and Table I, there is a decrease in performance parameters of \( \eta \), FF, and \( J_{SC} \) of the solar cells with different encapsulation processes changed relative to their initial values after exposure to ambient atmosphere.

As is generally expected for organic devices, the performance of the OPV cell with no encapsulation degraded the fastest. After only 10 h, \( \eta \) and \( J_{SC} \) dropped to less than 20% of their initial values. \( V_{OC} \) did not consistently decrease with air exposure and generally fluctuated around the initial value for all of the devices that were tested regardless of the encapsulation. This kind of rapid degradation in overall performance has been seen with other materials\(^{1-3}\) and is a reminder of how important it is to adequately encapsulate organic devices. Annealing alone without encapsulation did not have a significant effect on the sample stability to air. Devices encapsulated with only UV epoxy degraded at a slower rate with \( \eta \) dropping to ~50% of the initial after 100 h and to ~25% after almost 250 h of exposure to ambient atmosphere. Though not an ideal encapsulation, UV epoxy may be applied to slow degradation and provide short-term protection.

Devices with Al\(_2\)O\(_3\) deposited by ALD, both with and without an additional UV epoxy layer, have \( \eta \) and \( J_{SC} \) within 6% of the initial values after 6145 h of exposure to ambient atmosphere. The reduced rate of deterioration is thought to be caused by the Al\(_2\)O\(_3\) layer effectively blocking oxygen and water from reaching and reacting with the active materials of the devices. Studies on oxygen and water permeation through Al\(_2\)O\(_3\) films on plastic substrates\(^{8}\) have shown that Al\(_2\)O\(_3\) may be a better barrier to water than to oxygen; however, the present experiments do not provide enough information on the degradation mechanism of the organic layers to weight the relative contributions of oxygen and water to the overall lifetime. Intense light is also expected to be a source of degradation, but that effect was kept minimal in this study to estimate only the effectiveness of the barrier films as encapsulation. FF and \( \eta \) fluctuate between 100 and 6145 h, but these fluctuations are suspected to be related to contact issues at the electrodes when making repeated contact to the devices for testing.

In addition to \( J-V \) characteristics, the external quantum efficiency (EQE) of the devices was also measured. EQE of a device without encapsulation is compared to that of a device with Al\(_2\)O\(_3\) encapsulation in Fig. 3 before and after exposure to ambient air. As shown in Fig. 3(a), the EQE of the

### Table I. Initial performance characteristics of solar cells tested after encapsulation.

<table>
<thead>
<tr>
<th>Encapsulant</th>
<th>( \eta ) (%)</th>
<th>( J_{SC} ) (mA/cm(^2))</th>
<th>FF</th>
<th>( V_{OC} ) (mV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>None</td>
<td>2.0</td>
<td>10.5</td>
<td>0.53</td>
<td>361</td>
</tr>
<tr>
<td>UV Epoxy</td>
<td>2.1</td>
<td>10.6</td>
<td>0.54</td>
<td>363</td>
</tr>
<tr>
<td>Al(_2)O(_3)</td>
<td>2.8</td>
<td>11.1</td>
<td>0.57</td>
<td>449</td>
</tr>
<tr>
<td>Al(_2)O(_3) and UV Epoxy</td>
<td>2.7</td>
<td>10.4</td>
<td>0.57</td>
<td>451</td>
</tr>
</tbody>
</table>

![FIG. 2. Relative change in (a) \( \eta \), (b) FF, and (c) \( J_{SC} \) of solar cells with various types of encapsulation from the initial values after exposure to ambient atmosphere. Four devices are shown with the following types of encapsulation: no encapsulation (dash-dotted line, triangle); UV epoxy (dashed line, diamond); Al\(_2\)O\(_3\) (dotted line, square); and Al\(_2\)O\(_3\) and UV epoxy (solid line, circle). The values for each device are normalized to the initial value for that device. Data are symbols, and lines merely provide a guide for the eye.](image-url)
A device without encapsulation significantly decreases after only 1 h over the entire spectrum, and the maximum EQE drops from 41% at 365 nm to less than 20% after 10 h of exposure to ambient air. In contrast, the device in Fig. 3(b) with Al2O3 encapsulation has almost no change in EQE above 500 nm and at most a 10% change in EQE in the range of 325–500 nm after 6145 h of exposure to ambient. The decrease in EQE between 325 and 500 nm in the encapsulated device can be assigned to the degradation of the C60 layer, which has an absorption peak near 450 nm. Note that EQE shows virtually no change in the region where pentacene absorbs light exclusively. If the charge transport or the charge collection at the C60/BCP interface were significantly affected by the degradation, a change in EQE under short-circuit conditions in the spectral region where pentacene absorbs would also have been anticipated. Excitons created in the pentacene layer that dissociate at the pentacene/C60 interface lead to an electron that is transported by the C60 layer and collected by the corresponding electrode. Therefore, the major source of degradation can be tentatively assigned to the formation of exciton quenching centers that result from the degradation of C60 under illumination in the presence of oxygen. While there are measurable reductions in the EQE and JSC of the encapsulated device after 6145 h, JSC is still within 5% of the initial value and the time until η falls to half of the initial remains to be seen.

This study shows that encapsulation with a 200-nm-thick film of Al2O3 deposited by ALD can provide OPV cells with effective protection from ambient air and moisture, which is regarded as an important prerequisite for a long cell lifetime. The ALD process has been shown to also improve the performance of pentacene/C60 based solar cells by increasing VOC and η similarly to what is observed after annealing this type of organic solar cell. More studies are under way to optimize the ALD process, to reduce the length of the deposition, to determine the water vapor transmission rate, and to investigate the mechanical properties and stability under continuous solar illumination of the Al2O3 films.

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