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Citation: *Appl. Phys. Lett.* **90**, 253511 (2007); doi: 10.1063/1.2751108

View online: <http://dx.doi.org/10.1063/1.2751108>

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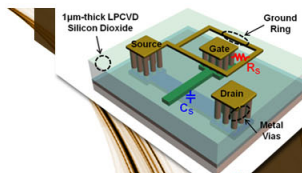
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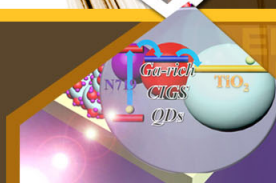
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Encapsulation of pentacene/C₆₀ organic solar cells with Al₂O₃ deposited by atomic layer deposition

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(Received 27 March 2007; accepted 29 May 2007; published online 21 June 2007)

Organic solar cells based on pentacene/C₆₀ heterojunctions were encapsulated using a 200-nm-thick film of Al₂O₃ 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 Al₂O₃ 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 Al₂O₃ films by ALD have been used alone⁵ 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/m² day have been measured through Al₂O₃ films grown on polyethylene naphthalate substrates.⁸ The research presented here describes the encapsulation of organic solar cells based on pentacene/C₆₀ heterojunctions with a layer of Al₂O₃ deposited by ALD and its effect on the OPV performance.

Organic solar cells were fabricated as previously reported⁹ with a device geometry comprised of indium tin oxide/pentacene (50 nm)/C₆₀ (40 nm)/bathocuproine (BCP) (7 nm)/Al and an active device area of ~0.1 cm². 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 Al₂O₃ was deposited on top of the completed devices. ALD was performed at a temperature of 100 °C

with pulses of H₂O for 100 ms and trimethylaluminum (TMA) for 100 ms. The chamber was pumped for 20 s after H₂O 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/cm².

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

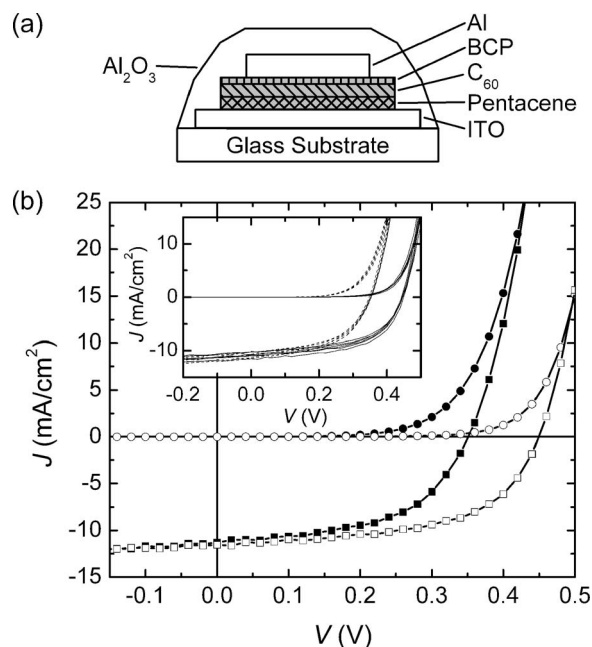


FIG. 1. (a) Schematic of device structure. (b) Electrical characteristics measured in the dark (circles) and under illumination (squares) for the same pentacene/C₆₀ solar cell before (filled shapes) and after (empty shapes) deposition of a 200-nm-thick layer of Al₂O₃ by ALD. Inset: overlap of electrical characteristics for six devices produced in the same batch before (dashed line) and after (solid line) Al₂O₃ deposition in the dark and under illumination.

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TABLE I. Initial performance characteristics of solar cells tested after encapsulation.

Encapsulant	η (%)	J_{SC} (mA/cm ²)	FF	V_{OC} (mV)
None	2.0	10.5	0.53	361
UV Epoxy	2.1	10.6	0.54	363
Al ₂ O ₃	2.8	11.1	0.57	449
Al ₂ O ₃ and UV Epoxy	2.7	10.4	0.57	451

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

$$J_{SC} = J(V=0 \text{ V}) \frac{100 \text{ mW/cm}^2}{I_L}. \quad (1)$$

Overall, power conversion efficiency η under the broadband light source increased from $2.0 \pm 0.1\%$ before encapsulation to $2.7 \pm 0.2\%$ after. By overlapping the external quantum efficiency with the solar spectrum and integrating, η after the ALD process is estimated to be $1.4 \pm 0.1\%$ 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 Al₂O₃ film as an encapsulation layer was tested over time. In addition to Al₂O₃, 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 Al₂O₃ layer, and the epoxy was then cured with UV light. Therefore, three types of encapsulation were tested and compared in this experiment: Al₂O₃ deposited by ALD, UV epoxy, and UV epoxy on top of Al₂O₃ 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 removed from the glovebox and exposed to ambient air. The cells were all fabricated in the same batch, and η is higher for the cells with Al₂O₃ because of the thermal annealing effects associated with the ALD process discussed previously. Figures 2(a)–2(c) show how the key performance parameters of η , FF, and J_{SC} for 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, η 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 re-

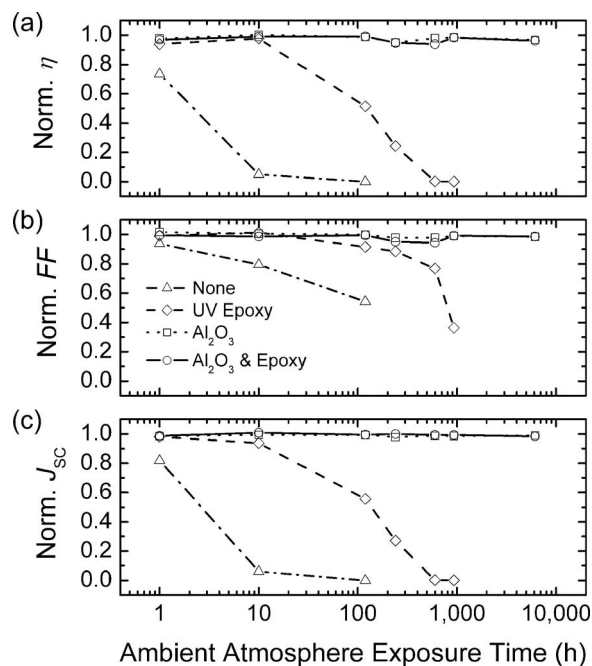


FIG. 2. Relative change in (a) η , (b) FF, and (c) J_{SC} of solar cells with various types of encapsulation from the initial values after exposure to ambient atmosphere. Four devices are shown with the following types of encapsulation: no encapsulation (dash-dotted line, triangle); UV epoxy (dashed line, diamond); Al₂O₃ (dotted line, square); and Al₂O₃ 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.

minder 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 η dropping to $\sim 50\%$ of the initial after 100 h and to $\sim 25\%$ 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 Al₂O₃ deposited by ALD, both with and without an additional UV epoxy layer, have η 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 Al₂O₃ layer effectively blocking oxygen and water from reaching and reacting with the active materials of the devices. Studies on oxygen and water permeation through Al₂O₃ films on plastic substrates⁸ have shown that Al₂O₃ 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 η 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 Al₂O₃ encapsulation in Fig. 3 before and after exposure to ambient air. As shown in Fig. 3(a), the EQE of the

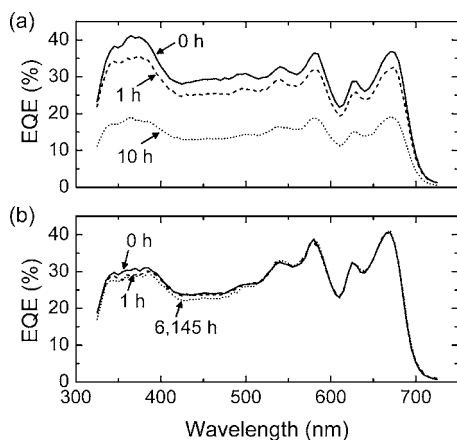


FIG. 3. External quantum efficiency of devices (a) without and (b) with Al_2O_3 encapsulation. EQE for a device without encapsulation is shown before (0 h) and after 1 and 10 h of exposure to ambient air, and EQE for a device encapsulated with Al_2O_3 is shown before (0 h) and after 1 and 6145 h of exposure.

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 Al_2O_3 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 C_{60} 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 C_{60}/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/ C_{60} interface lead to an electron that is transported by the C_{60} 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 C_{60} under illumination in the presence of oxygen.¹² While there are measurable reductions in the EQE and J_{SC} of the encapsulated device after 6145 h, J_{SC} 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 Al_2O_3 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/ C_{60} based solar cells by increasing V_{OC} 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 Al_2O_3 films.

The authors would like to thank Samuel Graham for fruitful discussions, Z. L. Wang for providing access to his ALD system, and X. D. Wang and X. H. Zhang for assistance in making the ALD films. This material is based on work supported in part by the STC Program of the National Science Foundation under Agreement No. DMR-0120967, by the Office of Naval Research (ONR), and by the Georgia Research Alliance.

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