APPLIED PHYSICS LETTERS **86**, 042105 (2005)

Humidity effect on electrical performance of organic thin-film transistors

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(Received 23 August 2004; accepted 22 November 2004; published online 18 January 2005)

Humidity dependence of electrical performance of *p*-channel organic thin-film transistors (OTFTs) with various semiconductor compounds has been investigated. All devices showed decreased current output and mobility as the relative humidity (RH) was increased. The moisture sensitivity of the OTFT saturation current depends on the device geometry (bottom or top contact device) and channel length. The OTFT configuration with a short channel length and bottom contact was most affected by humidity compared to the top contact and larger channel length OTFT structures. The degradation of electrical performance under high RH is attributed to charge trapping at grain boundaries by polar water molecules reducing the rate of charge transport. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1852708]

Organic thin-film transistors (OTFTs) have been demonstrated as promising candidates for flexible displays,[1–3] smart cards (badges),[4] and gas sensors.[5,6] Displays with organic electronics offer many advantages, such as light weight, low-cost processing, and mechanical flexibility. On the other hand, organic devices also have some significant shortcomings, such as relative low mobility and performance degradation due to moisture and oxygen. Extensive research effort has been focusing on synthesizing organic semiconductor materials with a high mobility and $I_{\mathrm{ON}}/I_{\mathrm{OFF}}$ ratio,[7–11] and improving OTFT performance by device engineering and optimizing processing conditions.[12,13] However, material stability and operational reliability must be satisfactory in order for OTFTs to be useful in practical applications. Moisture in the atmosphere is one of the causes for the degradation of electrical performance of OTFTs. Packaging of the organic device is one way to enhance device reliability.

The effect of humidity on pentacene transistor devices has been previously reported.[14,15] It was found that increasing humidity resulted in significant degradation of transistor performance. In this study, the effect of humidity on OTFTs performance was investigated with a broader class of *p*-channel organic semiconductor materials used as active layers for OTFTs, including oligofluorene derivatives (C12FTTF and C6TFT) as well as pentacene. Two types of structures, i.e., top and bottom contact, with relatively large (200 microns) or small (25 microns) gaps between the source and drain electrodes were investigated. A standard conductive silicon wafer with a thermally grown silicon dioxide (3000 Å) layer was used as the substrate. The heavily doped silicon serves as the gate electrode, and the thermal oxide layer as the gate insulator. Organic and metal thin films were deposited by vacuum evaporation at a pressure below 5×10^{-6} Torr with substrate temperature of 25 °C, unless stated otherwise. Samples with an identical organic semiconductor were made from the same evaporation. The normal thickness of the organic semiconductor thin film is 400 Å, and the morphology was polycrystalline. In such cases, which are typical for organic semiconductor films, the observed mobility is in fact severely limited by barriers at grain boundaries, and does not necessarily reflect the intrinsic mobility of a bulk crystal.

With the top contact OTFT configuration (source and drain electrodes on the top of semiconductor material), the semiconductor active layer was deposited on the thermal oxide first, then the source and drain electrodes were formed by depositing gold on the organic material through a metal shadow mask. For a bottom contact device, the source and drain electrodes were deposited on the silicon dioxide layer first, followed by the organic semiconductor. It was previously reported that organic semiconductors, such as pentacene, tend to form small grains when deposited on the source and drain electrodes.[12] Therefore, compared to the organic semiconductor deposited in the channel region (on SiO_2) of a bottom contact device, the grain size on and near the source/drain electrodes tends to be smaller. This implies that compared to a top contact structure with the same channel length, more grain boundaries exist in a bottom contact structure near the contacts. Furthermore, a pronounced dewetting of semiconductor film from Au electrodes can be observed for bottom contact structure.[16] This detwetting increases the possibility that polar water resides at the interface between the organic film and electrodes.

The humidity response measurements were carried out by enclosing OTFT samples, micropositioners, and a humidity gauge in a plastic glove bag filled with nitrogen. To prevent the leakage current between gate electrode and source/drain electrodes mediated by water vapor in a high relative humidity (RH) environment, the gate electrode was isolated by painting a fluorinated coating (Fluorad solution purchased from 3M) at the edge of substrate and around the gate electrode. Initially, a wet paper towel was placed in the sealed glove bag to increase the RH. The RH can normally reach 70–80% after a few hours. After the first data point was taken at a high RH level, the glove bag was purged with dry N_2 slowly so that the RH in the glove bag decreased. A minimum of 5 min was allowed between data points so that sample could come to equilibrium with the environment. At different RH levels, OTFTs are characterized by measuring transfer characteristics (I_d versus V_{gs}) and output characteristics (I_d versus V_{ds}). From the obtained characterization curves, electrical properties of OTFTs can be extracted. The

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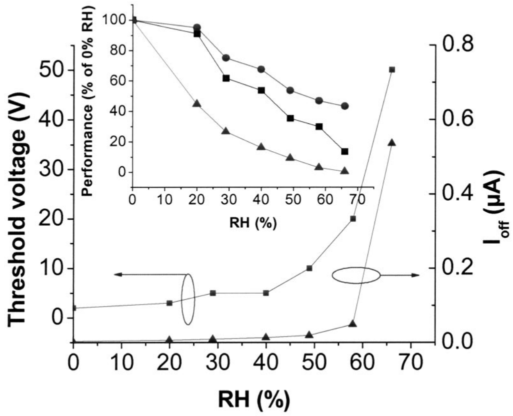

FIG. 1. Changes of OTFT electrical properties with respect to (RH): (1) (inset) Field-effect mobility [■], saturation current [●], and on/off current ratio [▲]; and (2) threshold voltage [■] and off current [▲]. Parameters are extracted from pentacene TFT with bottom contact, small gap ($L=25$ μm) structure.

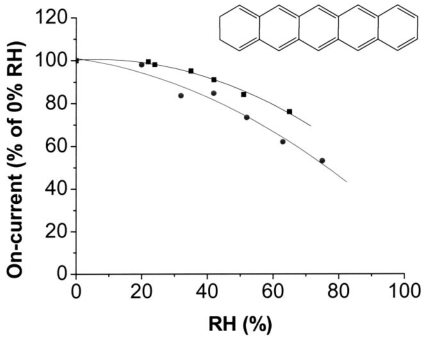

FIG. 2. Humidity dependence of OTFT with *p*-type organic material pentacene as a result of different OTFT device structures: (1) Large gap ($L=200$ μm), top contact [■]; and (2) large gap ($L=200$ μm), bottom contact [●].

conventional field-effect transistor (FET) model in the saturation regime is used to extract field-effect mobility and threshold voltage.

Figure 1 shows the effect of RH on saturation current, field-effect mobility, and I_{ON}/I_{OFF} ratio of a pentacene thin-film transistor (TFT). The change of electrical parameters is expressed by the percentage of change of electrical properties at certain RH level to the value in dry N_2. From Fig. 1, it can be clearly seen that electrical parameters, such as saturation current, field-effect mobility, and I_{ON}/I_{OFF} ratio, decrease with increasing RH. A similar trend was also observed with the two other *p*-channel organic semiconductors investigated.

The reduction of charge carrier mobility due to polar water molecules may be explained by the following possible microscopic mechanisms. Polar water molecules residing at the grain boundaries interact with hole charge carriers, reducing charge carrier mobility;[17] Diffusion of water molecules into grain boundaries changes the intermolecular interactions in those regions, increasing energy barrier for charge carrier intergrain transport;[18] or ions associated with water screen the electric field at the channel and lower the concentration of gate-induced mobile carriers.[19] All of the above mechanisms would lead to reduced on-current and field-effect mobility of OTFTs.

Another common trend that we observed was that both the threshold voltage (positive from zero in the depletion regime) and off-current increase with RH (Fig. 1), which indicates that mobile charges can be induced in the organic semiconducting layer by water vapor. A large positive gate bias is needed to compensate for the holes in the channel to turn off the transistor. Since threshold voltage moves to more positive values with RH, and the accumulation gate voltage is negative, there should be an increase in the saturation current for high negative gate voltages. However, it was observed that the saturation current always decreased with increasing RH. The reduction of saturation current is mainly attributed to a reduction of field-effect mobility—which, as mentioned above, is probably an indication of grain-boundary-limited charge transport. The combined effects just discussed lead to the on/off current ratio decreasing with increasing RH. For many OTFT-related applications, such as displays and circuits, the OTFTs should have a high saturation current, field-effect mobility, and I_{ON}/I_{OFF} ratio, and a low threshold voltage and off current. Our results here indicate that it is important to encapsulate OTFTs for these applications to prevent their performance from varying or degrading with humidity fluctuations.

The electrical properties of two other *p*-type organic semiconductors C12FTTF and C6TFT show a similar performance dependence on RH as pentacene. Here, only the on/saturation current (at $V_{ds}=-100$ V, $V_{gs}=-100$ V) is plotted against humidity, with mobility and off-current tracking with humidity, as before.

The OTFT device structure was found to impact the extent of performance degradation in an environment with high RH. The percentage of on-current change for pentacene TFTs, with either top or bottom contact geometry as a function of RH, is shown in Fig. 2. The experimental data are also fitted with polynomial curve-fitting lines for trend analysis. As the RH increases, the saturation current of OTFTs decreases. For OTFT samples with relatively large channel lengths of 200 μm, the change of current versus RH follows similar trends for the top and bottom contact geometries. The bottom contact devices were found to have a slightly faster decay of on-current versus RH.

The degradation of OTFT saturation current in a humid environment is expected to be strongly dependent on the density of grain boundaries between source and drain electrodes.[20] Increasing the density of grain boundaries in the OTFT channel results in more hole carriers trapped in the grain boundaries by water molecules residing at the grain boundaries, thus reducing the charge carrier mobility in the channel. Furthermore, an increased number of grain boundaries increases the possibility of water vapor diffusing into organic semiconductor thin film and reaching the interface between the organic semiconductor and dielectric layer, where device current flows. As a result, the higher density of grain boundaries contributes to the higher sensitivity to moisture. The organic semiconductor deposited on the Au electrodes (bottom contact) tends to have smaller grain sizes and more grain boundaries than those deposited on SiO_2, and a pronounced dewetting of the semiconductor film from electrodes can be formed. Higher moisture sensitivity of the bottom contact can also be expected if there is a higher contact resistance due to moisture residing at the electrode and organic semiconductor interface, either separating the semiconductor from the contact or creating a surface dipolar barrier. For these reasons, with the same channel length, the bottom

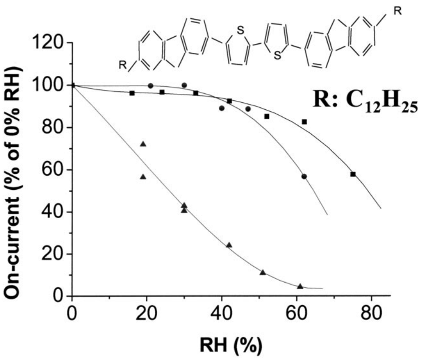

FIG. 3. Humidity dependence of OTFT with *p*-type organic material C12FTTF. Current responses with three OTFT configurations are compared, including: (1) Large gap ($L=200$ μm), top contact [■]; (2) small gap ($L=25$ μm), top contact [●]; and (3) small gap ($L=25$ μm), bottom contact [▲].

contact device should have a larger response to moisture than the top contact device. The difference in current response in Fig. 2, while measurable, however, is not dramatic when the channel length of the transistor is relatively large (200 μm), because the overall number of grain boundaries between the source and drain is not particularly different in the two cases.

A similar trend is also seen for another *p*-channel semiconductor C12FTTF. Also, when the channel length is relatively small, 25 microns in this case, a much faster decay of the on current for bottom contact devices is observed compared to top contact devices with the same channel length or top contact device with larger channel length (Figs. 3 and 4). This is understandable since contact resistance becomes important in the overall transistor resistance for small channel length devices, consistent with the reasoning above. This indicates that for small channel length devices, the moisture response is dominated by the response at the contacts.

There is a slight difference in the moisture sensitivity among these three organic materials. For instance, with the top contact/large gap structure, the on-current reduction of pentacene TFT at a RH level of 60% is about 30% compared to the on current of TFT in a fully dry environment (RH $=0$), while the on current of C12FTTF or C6TFT transistors was reduced by 20%. With the bottom contact, the small channel gap structure of the on current of the C12FTTF thin-film transistor at a RH level of 40% was reduced by 80%, while the C6TFT transistor was reduced by 60%. These differences may be related to the nature of the organic semiconductor and the detailed morphology of the thin film at interfaces between organic material and gold electrodes, as well as between organic material and dielectric surface. More systematic studies are underway.

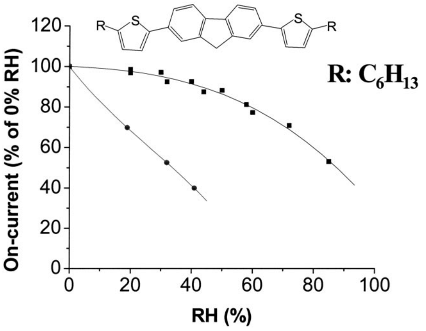

FIG. 4. Humidity dependence of OTFT with *p*-type organic material C6TFT. Current responses with different OTFT structures are compared, including: (1) Large gap ($L=200$ μm), top contact [■]; and (2) small gap ($L=25$ μm), bottom contact [●].

In summary, we investigated the current response of an OTFT to humidity with different *p*-channel organic semiconductors and various device structures. All of the *p*-channel organic semiconductors investigated in this study showed degraded transistor performance with increased humidity. It was found that OTFTs with small channel lengths and bottom contact structure have the highest sensitivity to RH. OTFTs with the same device structure, but different organic semiconductors, have a slightly different sensitivity to RH. These results indicate that encapsulation of OTFT devices is necessary for many practical applications.

The author are grateful to the National Institute of Standards and Technology (NIST) Advanced Technology Program (00-00-4968) and Lucent Technologies for financial support.

[1] P. Andry and C. R. Kagan, *Thin-Film Transistors* (Marcel Dekker, New York, 2003).

[2] B. Crone, A. Dodabalapur, Y.-Y. Lin, R. W. Filas, Z. Bao, A. LaDuca, R. Sarpeshkar, H. E. Katz, and W. Lin, Nature (London) **403**, 521 (2000).

[3] G. H. Gelinck, H. Edzer, A. Huitema, E. V. Veenendaal, E. van Cantatore, L. Schrijnemakers, J. B. P. H. van der Putten, T. C. T. Geuns, M. Beenhakkers, J. B. Giesbers, B.-H. Hiusman, E. J. Meijer, E. M. Benito, F. J. Touwslager, A. W. Marsman, B. J. E. van Rens, and D. M. De Leeuw, Nat. Mater. **3**, 106 (2004).

[4] P. F. Baude, D. A. Ender, M. A. Haase, T. W. Kelley, D. V. Muyres, and S. D. Theiss, Appl. Phys. Lett. **82**, 3964 (2003).

[5] B. Crone, A. Dodabalapur, A. Gelperin, L. Torsi, H. E. Katz, A. J. Lovinger, and Z. Bao, Appl. Phys. Lett. **78**, 2229 (2001).

[6] L. Torsi, A. Dodabalapur, N. Cioffi, L. Sabbatini, and P. G. Zambonin, Sens. Actuators B **77**, 7 (2001).

[7] H. E. Katz, A. J. Lovinger, J. Johnson, C. Kloc, T. Siegrist, W. Li, Y.-Y. Lin, A. Dodabalapur, Katr, and J. Lovinger, Nature (London) **404**, 478 (2000).

[8] Z. Bao, A. J. Lovinger, and A. Dodabalapur, Appl. Phys. Lett. **69**, 3066 (1996).

[9] P. R. L. Malenfant, C. D. Dimitrakopoulos, J. D. Gelorme, L. L. Kosbar, T. O. Graham, A. Curioni, and W. Andreoni, Appl. Phys. Lett. **80**, 2517 (2002).

[10] M. Halik, H. Klauk, U. Zschieschang, G. Schmid, W. Radlik, S. Ponomarenko, S. Kirchmeyer, and W. Weber, J. Appl. Phys. **93**, 2977 (2003).

[11] A. Afzali, T. L. Breen, and C. R. Kagan, Chem. Mater. **14**, 1742 (2002).

[12] I. Kymissis, C. D. Dimitrakopoulos, and S. Purushothaman, IEEE Trans. Electron Devices **48**, 1060 (2001).

[13] D. J. Gundlach, C. S. Kuo, C. D. Sheraw, J. A. Nichols, and T. N. Jackson, Proc. SPIE **4466**, 54 (2001).

[14] Z.-T. Zhu J. T. Mason, R. Dieckmann, and G. G. Malliaras, Appl. Phys. Lett. **81**, 4643 (2002).

[15] Y. Qiu, Y. Hu, G. Dong, L. Wang, J. Xie, and Y. Ma, Appl. Phys. Lett. **83**, 1644 (2003).

[16] G. Beernink, T. Strunskus, G. Witte, and C. Woll, Appl. Phys. Lett. **85**, 398 (2004).

[17] D. H. Dunlap, P. E. Parris, and V. M. Kenkre, Phys. Rev. Lett. **77**, 542 (1996).

[18] D. J. Gundlach, T. N. Jackson, D. G. Schlom, and S. F. Nelson, Appl. Phys. Lett. **74**, 3302 (1999).

[19] D. Zou and T. Tsutsui, J. Appl. Phys. **87**, 1951 (2000).

[20] T. Someya, H. E. Katz, A. Gelperin, A. J. Lovinger, and A. Dodabalapur, Appl. Phys. Lett. **81**, 3079 (2002).