Investigation of long-term stability of pentacene thin-film transistors encapsulated with transparent SnO₂

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Abstract

The long-term stability of pentacene thin-film transistors (TFTs) encapsulated with a transparent SnO₂ thin-film prepared by ion beam assisted deposition (IBAD) was investigated. With a buffer layer of thermally evaporated 100 nm SnO₂ film deposited prior to IBAD process, our encapsulated OTFTs sustained its initial field-effect mobility up to one month and then gradually degraded showing only 37% reduction compared to 90% reduction of non-encapsulated OTFTs after 100 days in air ambient. The encapsulated OTFTs also exhibited superior on/off current ratio of over 10^5 to that of the unprotected devices (10^4) which was reduced from ~ 10^6 before aging. Therefore, the enhanced long-term stability of our encapsulated OTFTs should be attributed to well protection of permeation of H_2O and O_2 into the devices by the IBAD SnO₂ thin-film which could be used as an effective inorganic gas barrier for transparent organic electronic devices.

1. Objectives and Background

Organic thin-film transistors have drawn much attention due to their great compatibility with plastic substrates available for a flexible display, smart cards, and rf tags application over the last decades.[1-2] Among organic semiconductors considered as active materials in organic thin-film transistors (OTFTs), pentacene was widely used for its mobility as high as that of a-Si or in some cases even exceeding that value.[3] However, when the OTFTs are operated in air ambient, they tend to easily degrade in the diodes (OLEDs) is limited.[4] In order to enhance the stability of devices, some encapsulation layers capable of protecting H₂O or O₂ in the air from penetrating through the organic layers have been employed. SiOx, AlOx, AlOxNv, and, SiO_xN_y are common amorphous oxides used as a inorganic gas barrier.[5-6] But, during the deposition process of the above mentioned oxides, some serious damages might be induced on the organic layers, which would be attributed to energetic ions, x-rays and electron beam.[7-8] Among the transparent amorphous oxides which can perform function of gas barrier deposited by simple thermal evaporation, SiO_x and SnO₂ would be taken into account. It was previously reported by our group that SnO₂ was more desirable one due to its high polarizability resulting in effectively suppressing permeation of water vapor by strong chemical interaction, particularly when the film could be made to have much denser microstructures.[9] Therefore, the SnQ thin-film prepared by ion beam assisted deposition (IBAD) is a promising candidate for transparent thin-film encapsulation adaptable for top-emitting organic light-emitting diodes(TE-OLEDs) since it has relatively high transmittance (85% over 450 nm) as well as quite low water vapor transmission rate (WVTR) of below 10⁻³ $g/m^2/day$. In order to prevent possible damages induced from energetic ions during IBAD process, it is also desirable that a proper buffer layer should be employed.[10] In this paper, we report on the investigation of the long-term stability of our OTFTs in terms of electrical properties associated with materials characterization by X-ray photo-

similar way that the life time of organic light-emitting

emission spectroscopy (XPS) when the TFT was encapsulated with the IBAD SnO₂ thin-film having a buffer layer of same kind of material on top of the device.

2. Results and discussion

The I-V measurements of our OTFTs were performed with an HP 4145B parameter analyzer in the dark at RT.



Fig.1 (a) The output characteristics of the reference OTFT without any encapsulation (b) Degradation of the drain current in the saturation regime when V_G is -40V after 100 days in air.

Figure 1(a) displays the output characteristics of our OTFT without encapsulation and the drain current in the saturation regime at a gate bias of -40V was around -120uA and corresponding I_G measured between the gate and source electrode was below 2 nA. The curves in figure 1(b) are representing the drain current of the OTFT without encapsulation (open circle) and with encapsulation (close circle) when the gate bias of V_G is – 40V. According to Fig. 1 (b), the saturation current of the unprotected OTFT is decreased to 35 uA from 120 uA after 100 days in air ambient, whereas the encapsulated device showing a relatively low initial current of 100 uA maintained considerable current level of 60 uA. The initial degradation of the OTFT with encapsulation might be attributed to some possible damages induced on the pentacene active layer during the encapsulation process in which small portion of the energetic ions could penetrate through the thermally evaporated buffer layer.[10]

The plots of $\sqrt{-I_D}$ vs. V_G and Log_{10} - I_D vs. V_G with and without encapsulation are presented in Figure 2. The fieldeffect mobility of holes in the pentacene layer was obtained from the slope in V_G versus plot in the saturation regime \sqrt{where} the source-drain voltage of $V_D = -40V$ was applied. The initial field-effect mobility of the encapsulated and unprotected OTFT was calculated to be $0.5 \text{ cm}^2/\text{V}$ s and $0.62 \text{ cm}^2/\text{V}$ s respectively. As expected from serious degradation of saturation current of the OTFT without encapsulation in Figure 1(b), the mobility of the device decreased sharply from 0.62cm²/V s to 0.10 cm²/V s after 100 days. On/off current ratio and sub-threshold slope also showed a similar degradation trend according to the Log_{10} - I_D vs. V_G plots. The On/off current ratio decreased down to $\sim 10^4$ from initial value of $\sim 10^6$ with a very inferior subthreshold slope of 7.2 V/dec. In the mean while, the encapsulated device maintained an on/off current ratio of $\sim 10^5$ even after 100 days with similar sub-threshold slope to the initial value. It is also interesting to note in the

 $\sqrt{-T_p}$ vs. V_G plot that the threshold voltage (V_T) changes toward positive gate bias showing early turn-on upon degree of degradation. Although the initial V_T values of both devices were almost same as ~ -2.5V, the unprotected device dis played about 7V as shown in figure 2 (a), while the same-period-aged but encapsulated one showed only -0.1V of V_T after 100 days in air. Since the pentacene channel/SiO₂ interfaces should be in the same condition for all OTFTs, we suspected if water molecules exist in the channel layer. The water molecules in air are known to be adsorbed on pentacene surface or even penetrate into the channel layer during aging period.[11]



FIG 2. The plots $q \overline{I_{b}}$ vs. V_G and Log₁₀-I_D vs. V_G. for the OTFTs without encapsulation (open circle) and with encapsulation (close circle)



FIG 3. The property-endurance limit or aging effect with time of our OTFTs in terms of field-effect mobility and on/off current ratio (inset).

Then they could be one of the main leakage sources or active deep level traps when the OTFT is particularly under a high electric field of depletion state.[12]

The property-endurance limit or aging effect of our OTFTs in terms of field-effect mobility and on/off current ratio upon time are shown in figure 3. The encapsulated TFT shows the best performance even after a long time exposure to air. The initial degradation of mobility from $0.62 \text{ cm}^2/\text{V}$ s to $0.5 \text{ cm}^2/\text{V}$ s is the expected behavior in line with reduction of the drain current of the encapsulated OTFT in figure 1(b). However, the SnQ film was deposited by using

IBAD on top of the device without any buffer layer, the device was found to seriously degrade due to direct penetration of energetic ions into the organic active layer.[10] In order to avoid deterioration of the device during IBAD process, it is strongly required to employ a proper buffer layer of which is thermally evaporated SnO_2 in this study. Unfortunately, thermally evaporated SnO_2 film had quite open structure resulting in a loose microstructure and this would also be known from the fact that, when being used alone, it could not effectively suppress HO permeation into the device as indicated in figure 3. Therefore the buffer layer itself could not play an important role in blocking permeation of water vapor and it is the IBAD SnO_2 layer that performs this function.

3. Conclusion

We have fabricated pentacene TFTs with the transparent SnO₂ encapsulation layer prepared by IBAD to enhance long-term stability of the devices. The IBAD SnO₂ was found to effectively prevent H₂O or O₂ from permeating into the devices by means of densification of microstructures through IBAD process and chemical interaction between water vapor and SnO₂ which resulted in quite low WVTR value accounting for enhancement of the long-term stability of the OTFTs. It is thus concluded that a transparent SnO₂ thin-film prepared by IBAD is a promising material that could be used as an inorganic gas barrier for transparent organic electronic devices with an appropriate buffer layer including some smoothing layers.

4. Reference

1) A. R. Brown, A. Pomp, C. M. Hart, D. M. de Leeuw, *Science*. **270**, 972 (1995)

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

3) P. V. Necliudov, M. S. Shur, D. J. Gundlach, and T. N.

- Jackson, J. Appl. Phys. 88, 6594 (2000)
- 4) H. Aziz, Z. Popovic, C. Tripp, N. -X. Hu, A. -M. Hor,
- and G. Xu, Appl. Phys. Lett. 72, 2642 (1998)
- 5) M. Vogt, and R. Hauptmann, *Surf. Coat. Technol.* **676**, 74 (1995)
- 6) A. G. Erlat, B. M. Henry, J. J. Ingram, D. B. Mountain, A.
- McGuigan, R. P. Howson, C. R. M. Grovenor, G. A. D.
- Briggs, and Y. Tsukahara, Thin Solid Films, 388, 78 (2001)
- 7) V. Bulovic, P. Tian, P. Burrows, M. R. Gokhale, S. R.
 Forres, M. E. Thompson, *Appl. Phys. Lett.* 70, 2954 (1997)
- 8) L. S. Hung, C. W. Tang, M. G. Manson, P. Raychudhuri, J.
- Madathil, Appl. Phys. Lett., 74, 544 (2001)
- 9) W. H. Koo, S. M. Jeong, S. H. Choi, S. M. Lee, S. J. Lee,
- and H. K. Baik, J. Phys. Chem. B, 108, 18884 (2004).
- 10) S. M. Jeong, W. H. Koo, S. H. Choi, S. J. Jo, S.J. Lee, K.
- M. Song, H. K. Baik, Appl. Phys. Lett., 85, 1051 (2004)
- 11) Y. Qiu, Y. Hu, G. Dong, L. Wang, J. Xie, Y. Ma, *Appl. Phys. Lett.* 83, 1644 (2003)
- 12) J. Lee, K. B. Kim, J. H. Kim, S. I. Im, D. Y. Jung, Appl.
- *Phys. Lett*, **82**, 4169 (2003)