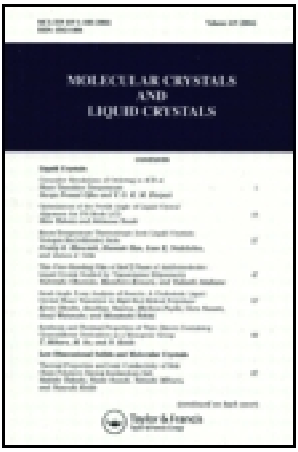

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Important Role of Polymorphs of Organic Semiconductors on the Reduction of Current Leakage in an Organic Capacitor

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Leakage currents in two different capacitors, metal-insulator-metal (MIM) and metal-insulator-semiconductor-metal (MISM), are examined. The leakage current of the MIM capacitors with a Nylon6-TiO_2 nanocomposite film was larger than that for the devices with a poly(4-vinylphenol) (PVP) buffer layer stacked on top of the composite film by a factor of 3.6. When the organic active layer of pentacene is introduced for the MISM capacitor, the reduction ratio of leakage current by stacking the PVP buffer layer onto the composite film is found to get more pronounced as much as about a factor of 25. Such a significant difference of reduction ratio for the leakage current between the MIM and MISM capacitors results from the different polymorphs of pentacene which are critically dictated by the morphological surface of the underlying layer.

Keywords Current leakage; Polymorphs; Organic semiconductor; Organic capacitor

Introduction

Field-effect transistors (FETs) are widely used for applications in analog switching, digital integrated circuits and various amplifiers [1, 2]. Recently, the demand for portable and robust electronic devices has been on the remarkable increase with the development of informationization and the information society, which expedites the development of flexible and plastic electronics [3–5]. However, traditional silicon-based FETs have great challenges for these applications on aspects of processing compatibilities with plastic or thin glass films. It is thus expected that organic semiconductors will be the cornerstone of future semiconductor electronics industry because they offer low-temperature and large-area processes [6–8]. This is also exemplified by pioneering works on the integration of organic FETs into bendable displays and disposable chips. Now, low-power consumption is crucial for their practical applications [9, 10].

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To reduce an operating voltage of organic FETs, intensive efforts have been devoted to develop nanocomposite insulators with prospects of simple solution processes of polymers and high dielectric properties of inorganic particles [11–13]. Meanwhile, nanocomposite layers often cause high gate-leakage currents in organic FETs [14], which are mostly attributed to percolation pathways through aggregated inorganic particles in the nanocomposite insulator itself, so far. However, our results suggest that such leakage currents are intimately associated with the morphological characteristics of an organic semiconductor as well as the nature of a composite insulator, which were obtained from the capacitor which is the most basic component to comprise the FETs. In this study, we exploit polymorphs of pentacene to comprehensively understand the leakage-current mechanism in organic devices by comparing the two different structured capacitors; metal-insulator-metal (MIM) and metal-insulator-semiconductor-metal (MISM).

Experimental

Figure 1 shows two different conformations of the capacitors used for our study; an MIM type (formed by stacking a metal, an insulator and a metal) and an MISM type (formed by stacking a metal, an insulator, an organic semiconductor and a metal). The indium-tin-oxide was used for the bottom electrode. As an insulator, Nylon6-TiO_2 nanocomposite films with and without a polymeric buffer layer of poly(4-vinyl phenol) (PVP) were formed by spin-coating from solutions. For the composite solution, TiO_2 nanoparticles were uniformly dispersed in a polymer matrix Nylon 6. The preparation of Nylon6-TiO2 nanocomposite film was published in greater details in a previous work [13]. For the PVP solution, the concentration of PVP molecules was approximately 1 wt.% in a solution of isopropyl alcohol. The resulting thicknesses of a Nylon6-TiO_2 nanocomposite and a PVP films were about 350 and 30 nm, respectively. To fabricate an MISM device, a 60-nm-thick pentacene layer was thermally deposited at a rate of 0.05 nm/s under a basal pressure of $10–^6$ Torr. Finally, an 80-nm-thick Au top-electrode was thermally deposited for the top electrode. The active area was 1 cm $\times$ 1 cm. The electrical characterization of our devices was undertaken in dark under ambient condition, using a semiconductor parameter analyzer (HP 4155A). The surface morphologies of both the insulating film and the pentacene layer were examined by an atomic force microscope (AFM).

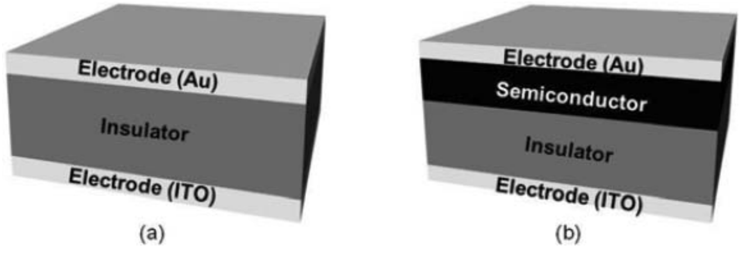

Figure 1. Two different structured organic capacitors: (a) MIM capacitor and (b) MISM capacitor. Each insulator in both two categorized capacitors is constituted as two classes, one of which is only Nylon6-TiO_2 nanocomposite layer and the other is a hybrid layer, PVP buffer layer stacked onto a Nylon6-TiO_2 nanocomposite layer.

Results and Discussion

Figure 2(a) shows the current density versus the electric field plots of the MIM capacitors with different insulating layers. At an electric field of 0.5 MV/cm across the Nylon6-TiO_2 nanocomposite film, the current density is 9.8×10^{-4} A/cm^2, which is larger than that 2.7×10^{-4} A/cm^2 for the other MIM capacitor with an extra PVP buffer layer onto the nanocomposite film by a factor of 3.6. Previously, it was reported that aggregated nanoparticles provide a current path through a nanocomposite insulator, so that can result in a significant amount of leakage currents in organic FETs [14, 15]. This explanation is primarily based on the percolation theory and viable to understand the intrinsic breakdown distribution in a nanocomposite insulator. On the other hand, lower leakage-currents for the device with a hybrid layer which has an extra PVP buffer layer are obtained because the PVP buffer layer effectively blocked a charge injection from the top Au electrode into the Nylon6-TiO_2 nanocomposite film. In contrast to the Nylon6-TiO_2 nanocomposite only case, physical processes that might account for the current leakage through the PVP buffer layer are Schottky emission and the Poole-Frenkel effect. The former process occurs at the interface between the electrode and the dielectric layer, which has a current versus voltage dependence of the form [16]

$$I \propto T^2 \exp\left(\frac{B_{Sc}V^{1/2} - \Phi_{Sc}}{k_B T}\right), \tag{1}$$

where Φ_{Sc} is the Schottky barrier height, β_{Sc} is the Schottky coefficient, T is the temperature, and k_B is the Boltzmann constant. Poole-Frenkel conduction is a similar process to Schottky emission, but it results from the lowering of the potential barriers around impurity centers in the bulk of the insulator. The current-voltage relationship is given by [17]

$$I \propto V \exp\left(\frac{B_{PF}V^{1/2} - \Phi_{PF}}{k_B T}\right), \tag{2}$$

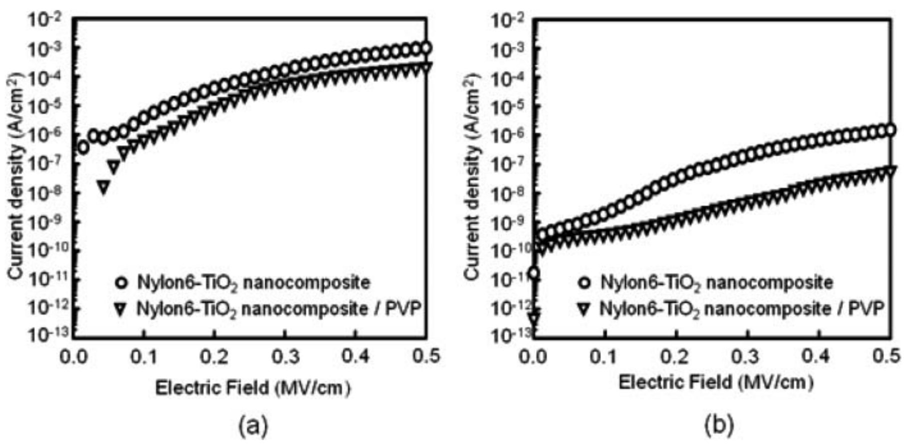

Figure 2. Current density per unit area as a function of applied electric field in (a) two types of the MIM capacitors and (b) two types of the MISM capacitors. Open circles and open triangles represent the Nylon6-TiO_2 nanocomposite layer only and the hybrid layer, respectively.

Table 1. The current density at the 0.5 MV/cm in four different organic capacitors is summarized.

MISM capacitors		MIS capacitors	
Nylon6-TiO_2 nanocomposite	Hybrid layer	Nylon6-TiO_2 nanocomposite	Hybrid layer
9.34×10^{-4} (A/cm^2)	2.74×10^{-4} (A/cm^2)	1.53×10^{-6} (A/cm^2)	6.14×10^{-8} (A/cm^2)

where Φ_{PF} is the potential barrier height and β_{PF} is the Pool-Frenkel coefficient. Considering the decent insulating properties of a PVP film, the Pool-Frenkel effect might be responsible for the currents in the PVP layer. Therefore, a defect-free buffer layer is clearly needed to minimize Pool-Frenkel conduction in the film, which is expected to effectively block a charge injection into a leaky nanocomposite insulator.

Another important observation is the difference in the magnitude of the leakage currents for both MISM-type capacitors, as shown in Fig. 2(b). For the MISM capacitor with the Nylon6-TiO_2 nanocomposite film, the current density at an electric field of 0.5 MV/cm is measured to be approximately 1.5×10^{-6} A/cm^2. Meanwhile, the device with a hybrid layer which is incorporating a PVP buffer layer onto the nanocomposite film exhibited significantly lower currents; it is as low as 6.1×10^{-8} A/cm^2 at the same electric field of 0.5 MV/cm. Of great interest is that the current difference at both MISM-type capacitors exceeds an order of magnitude (approximately 25), which is more prominent than that observed in the two MIM-type capacitors. This implies that a pentacene film may correlate with the magnitude of the leakage currents in the MISM-type structure. Each the magnitude of the leakage current density for four different organic capacitors at 0.5 MV/cm is summarized at Table 1.

In order to explain such a pronounced difference in leakage currents for the two MISM-type capacitors compared to the two MIM capacitors, we have analyzed the surface morphologies of pentacene films grown on the Nylon6-TiO_2 nanocomposite and hybrid layers, using an AFM. Figures 3(a) and 3(b) reveal that the Nylon6-TiO_2 nanocomposite layer has a relatively rough surface with a root-mean-square (RMS) roughness value of 9.1 nm, compared to the hybrid film (a RMS roughness $\sim$ 4.5 nm). The AFM images in Figs. 3(c) and 3(d) clearly show different polymorphs in the pentacene films; smaller grains of pentacene molecules are evident for the pentacene film grown on a rough nanocomposite films. Note that the surface properties of an underlying layer essentially affect the growth of organic semiconductor molecules [18–20]. Based on the growth dynamics of pentacene molecules, it can be stated that the rough Nylon6-TiO_2 nanocomposite layer limit diffusions of pentacene molecules on its surface during an initial growth stage, thereby producing small grains of pentacene molecules. Note that the only difference between two MISM capacitors is the different grain size of pentacene film which is directly governed by the insulator surface. From the experimental fact that the leakage current is much decreased through the enlarging the grain size of the pentacene film, it is found that the current leakage could be much reduced by controlling the interfacial interaction between an organic semiconductor and the dielectric beyond the amount of the possible reduction of the current leakage by inserting an extra buffer layer as described in the MIM capacitor.

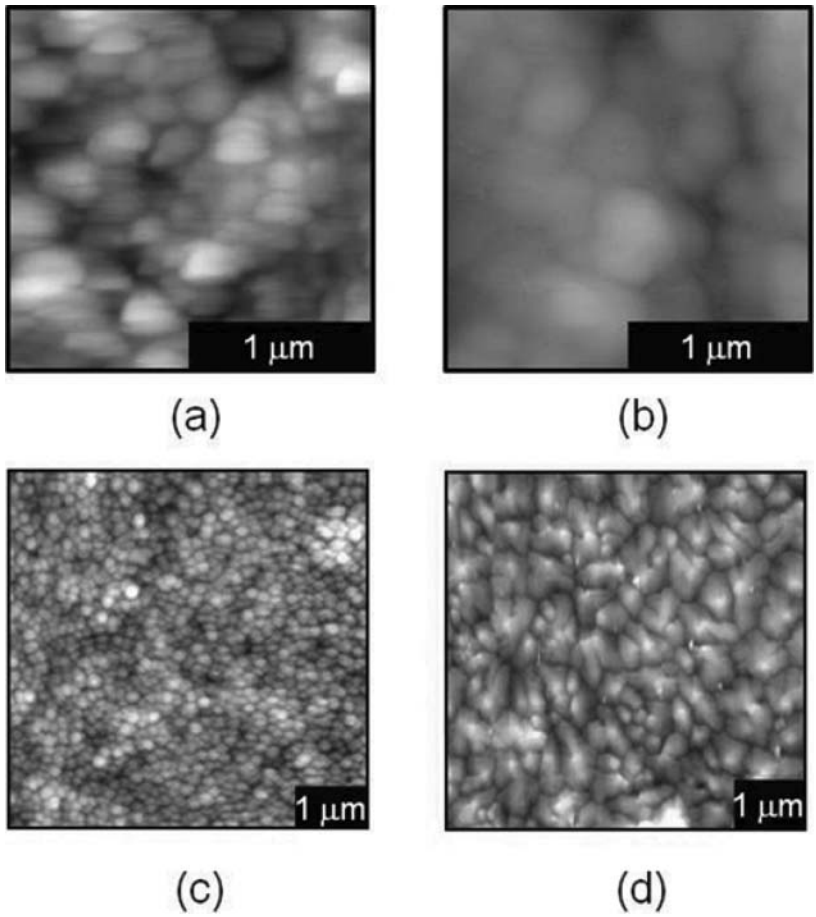

Figure 3. The AFM images of the two different insulators, (a) Nylon6-TiO_2 nanocomposite layer and (b) the hybrid layer. The corresponding AFM images of (c) the pentacene deposited on the Nylon6-TiO_2 nanocomposite layer and (d) the pentacene on the hybrid layer, respectively.

Conclusion

We present how the different polymorphs in a pentacene film as an organic semiconductor affects the magnitude of the current leakage in the MISM capacitor in relation to the MIM capacitor. While the current leakage is reduced by a factor of 3.6 in the MIM capacitors by introducing the PVP buffer layer onto a Nylon6-TiO_2 nanocomposite film, the reduced ratio of the current leakage reaches as high as a factor of 25 in the MISM capacitors due to the large grains of the pentacene film on the hybrid layer. When the size of the grain in an organic semiconductor becomes larger, the magnitude of current leakage is found to be more reduced for the same device structure without any additional processes. This work is expected to provide a scientific platform to fabricate a variety of organic semiconductor used devices for low current leakage property.

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