P-19: Flexible Pentacene Organic Thin Film Transistors using Sol-Gel Hybrid Polymer Gate Dielectrics

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Abstract

Flexible organic thin film transistors (OTFTs) have been realized with sol-gel hybrid polymers (hybrimer) gate dielectric and poly(ether sulfone) substrate. The hybrimer thin films have good electrical and surface properties. OTFTs on polymer substrate exhibit good OTFT performance and small hysteresis.

1. Introduction

Organic thin film transistors (OTFTs) have attracted much attention because OTFTs have lots of advantages such as low cost, easy processability and flexibility. By using them, we can fabricate easily the cheap electronic devices, like integrated circuits and driving circuits for active matrix display, sensors, and RFIDs[1]. Their performance has been improved impressively during the last two decades[2]. The organic dielectric is favored as the gate dielectric in OTFTs due to their solution processability and flexibility which can lower process temperature and make it easy to fabricate large-area OTFTs. However, organic gate dielectric has high leakage current densities and poor chemical stability. In addition, the organic gate dielectric causes the large hysteresis of the OTFTs.

The sol-gel derived siloxane based organic-inorganic hybrid polymers (hybrimers) are nanocomposite materials in which inorganic and organic components are intimately linked at the molecular scale by a covalent bond[3,4]. Since the hybrimer combines the characteristics of both glass and polymers, the hybrimer has the good electrical and surface properties such as low leakage current density, smooth and chemically stable surface, and good photo-patternability[3,5]. In this study, we synthesized methacryl-grafted hybrimer and fabricated the hybrimer gate dielectric layer for flexible OTFTs by solution process.

2. Experimental

2.1 Synthesis of Hybrimer Resins and Films

The hybrimer containing oligosiloxanes grafted by methacryl radicals was synthesized using a simple non-hydrolytic sol-gel reaction. 3-(trimethoxysilyl)propyl methacrylate (MPTMS), diphenylsilanediol (DPSD) were used as precursors without further purification. The total proportion of MPTMS and DPSD together was 1 : 1 molar ratio. Barium hydroxide monohydrate ($Ba(OH)_2 \cdot H_2O$, Aldrich) was used as a catalyst to promote a condensation reaction between the two precursors. Synthesis of the hybrimer was briefly illustrated in Figure 1.

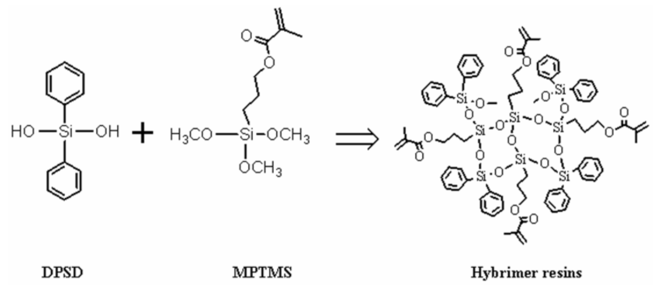

Figure 1 Schematic synthesis diagram of the hybrimer by a non-hydrolytic sol-gel reaction of DPSD and MPTMS

The hybrimer thin films were deposited on highly doped silicon wafer with resistivity of 0.01 Ω cm. The solutions of the hybrimer diluted in propylene glycol monomethyl ether acetate (PGMEA) were spin coated at 5000 rpm for 30s on the substrate. Thickness of the dielectric was controlled by varying the concentration of the hybrimer solution. The deposited films were exposed to UV light (λ=365nm, Hg lamp) for 90 seconds and thermally cured at 150°C for 2 hours in air condition. Aluminum dots were then thermally evaporated on the gate dielectric to prepare the metal-insulator-semiconductor (MIS) capacitors.

2.2 Fabrication of Flexible OTFTs

Flexible OTFTs were fabricated by using a top contact geometry on poly(ether sulfone) (PES) substrate. Aluminum was thermally evaporated on the PES as gate electrodes and the hybrimer gate dielectric is formed on aluminum layer same as the method described above. A 50 nm thick pentacene layer was thermally evaporated on the hybrimer dielectric at a rate of 0.6 ~ 0.8 Å/ s at substrate temperature of 80ºC. Gold was thermally evaporated on the pentacene films through a shadow mask to form source and drain electrodes. The OTFTs had a channel length (L) of 50 μm and channel width (W) of 3000 μm.

2.3 Characterization of Films and OTFTs

The electrical properties of the hybrimer thin films including capacitance and leakage current density were measured using Al/hybrimer thin film/Si structures with an HP4194A impedance

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analyzer and a Keithley 236 source-measure unit. The crystallographic ordering and the morphology of pentacene on the hybrimer thin films were measured using X-ray diffraction (XRD) and AFM, respectively. And contact angle analyzer was used to examine hydrophobicity of the hybrimer surface. Electrical characteristics of the OTFTs were measured using an HP 4155A semiconductor parameter analyzer.

3. Results and Discussion

3.1 Characteristics of Hybrimer Films

The formation of totally cross-linked 3-dimensional networks and few hydroxyl groups in the hybrimer gate insulator give the low leakage current density and high dielectric strength of the hybrimer thin film. Figure 2 shows the current densities of the hybrimer dielectric as a function of the hybrimer thin film thickness. The dielectric strength, which is measured at leakage current density of 10^{-6} A/cm^2, is higher than 2.5 MV/cm. The leakage current densities at 1MV/cm increase from 4nA/cm^2 to 15nA/cm^2 as the film thickness decrease. The electrical properties of the hybrimer thin films are much better than those of the commonly used polymer dielectric in OTFTs[6,7]. The dielectric constant of the hybrimer dielectric film is 3.1 at 100 kHz, which is calculated from the film thickness and capacitance.. The water contact angles of the hybrimer thin films are as high as ~80°, which indicates that they have the hydrophobic surface. It is known that surface properties such as surface energy and roughness of gate dielectric are one of the most important factors affecting the performance of the OTFTs. Figure 3 shows that the spin-coated hybrimer thin films have pin-hole free surface and their rms roughness is as low as ~0.5 nm examined by an atomic force microscopy (AFM), which reveals that the hybrimer thin films have the very smooth surface. Since the hybrimer thin films have the good electrical properties such as low leakage current densities and high dielectric strength, and the smooth and hydrophobic surface, the hybrimer can be used as the gate dielectric in OTFTs.

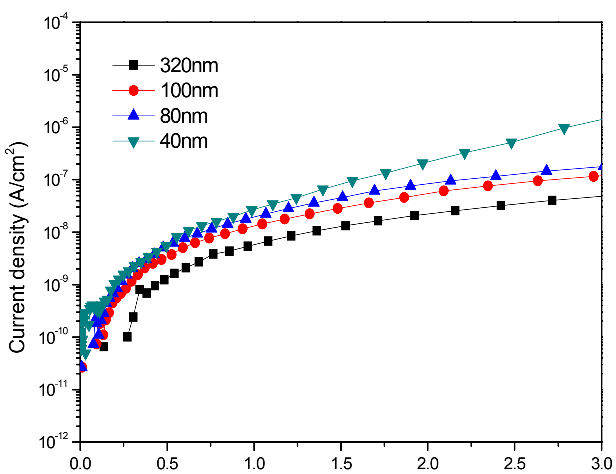

Figure 2 Leakage current densities of the hybrimer thin films with different thickness

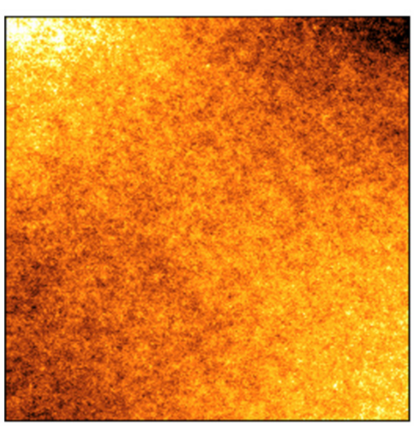

Figure 3 AFM images of spin-coated the hybrimer films. Image is 5 µm × 5 µm in size.

3.2 Characteristics of Active Layer

It is known that the performance of the OTFTs depends on the crystallinity and grain size of organic semiconductor[8,9]. Figure 4 shows the XRD patterns of the pentacene films deposited on hybriemr gate dielectric with 50nm. The XRD patterns of the pentacene show the thin-film phase of pentacene with its main peak at 2θ =5.7°. Three characteristic peaks which come from the (00l) planes of the thin-film phase are shown in the XRD patterns. Due to the fact that the main peak is sharp and intense, a high crystallinity of pentacene on the hybrimer gate dielectric can be assumed. Also we investigated the morphology of pentacene on the hybrimer gate dielectric using AFM. Figure 5 shows AFM

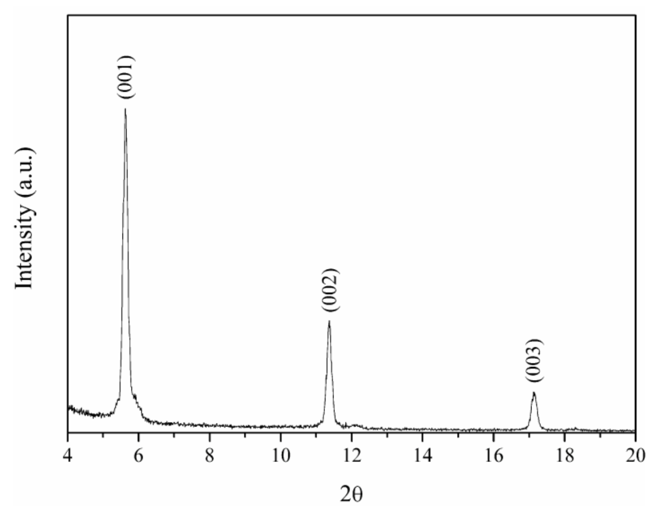

Figure 4 X-ray diffraction patterns of the pentacene films deposited on the hybrimer gate dielectric

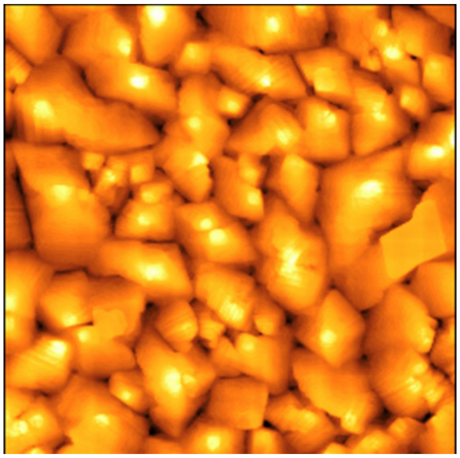

Figure 5 AFM image of pentacene on the hybrimer gate dielectric. Image is 5 μm × 5 μm in size.

images of the morphology of pentacene film on the hybrimer gate dielectric which has large grain size of about 1 μm. Grain size is big enough that charge carriers would flow between pentacene molecules easily.

3.3 Characteristics of Flexible OTFTs

Figure 6(a) shows the output curves of the flexible OTFTs with the hybrimer gate dielectric at various gate voltages (V_G). The flexible OTFTs exhibit good linear/saturation behavior. Figure 6(b) shows the transfer curve of the flexible OTFTs with the hybrimer gate dielectric. The drain-source voltage (V_{DS}) was kept at -40 V. The gate voltage (V_G) was swept continuously with a 0.5 V step, starting from +20V, passing through -40V, and finally returning to +40V. The electrical parameters were determined from a plot of $I_D^{1/2}$ vs. V_G on the basis of the following relationship in the saturation regime:

$$I_D = \frac{WC_i}{2L}\mu(V_G - V_{th})^2 \quad (1)$$

where I_D is the drain current, μ is the field-effect mobility, C_i is the capacitance per unit area, V_{th} is the threshold voltage.

The flexible OTFTs have the mobility of 0.15 cm^2/Vs and threshold voltage of -13.4 V, respectively. The subthreshold slope, S, defined as the voltage require to increase the drain current by a factor of 10, was 2 V/decade. The on/off current ratio is higher than 10^6, which is pretty high ratio. Furthermore, the flexible OTFTs with the hybrimer gate dielectric show small hysteresis as shown in Figure 6(b). One major problem in using solution processable gate dielectric is the hysteresis problem. It is desirable that the hysteresis of OTFTs is as small as possible for organic circuit applications. In the case of the hybrimer gate dielectric,

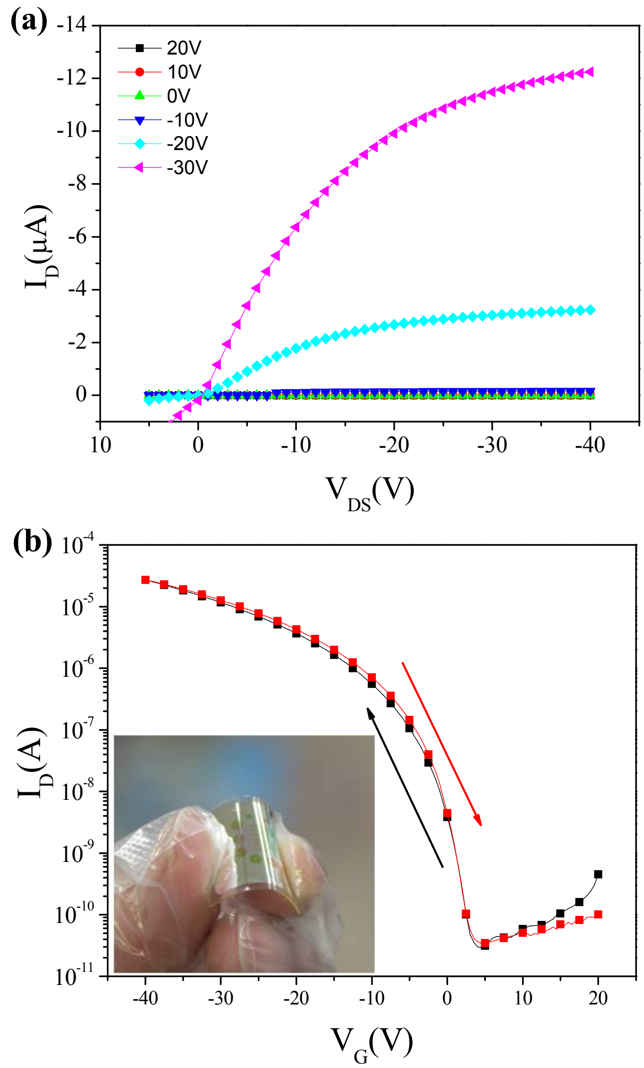

Figure 6 (a) Output characteristics and (b) Transfer characteristics of OTFTs with the hybrimer gate dielectric on PES [inset: photograph of the flexible OTFTs]

small hysteresis is due to the hydrophobic surface and the low leakage current, and few hydroxyl groups of the gate dielectric.

4. Conclusion

Solution-processed the hybrimer thin film showes good electrical properties, including high dielectric strength and low leakage current density down to the 40 nm thickness, and have also smooth and hydrophobic surface. And pentacene forms large grain with high crystallinity on the hybrimer gate dielectric. The electrical properties of the hybrimer thin film are much better than those of the commonly used polymer dielectric in OTFTs. The flexible OTFTs fabricated on the PES substrate with the hybrimer gate dielectric exhibit a mobility of 0.15 cm^2/V s, a threshold voltage of -13.4 V, and high on/off ration of >10^5. In addition, the flexible OTFTs have small hysteresis. Solution-processed hybrimer gate dielectric will make it possible to fabricate the flexible and low-cost OTFTs with high-performance.

5. References

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