CHIN.PHYS.LETT. Vol. 25, No. 2 (2008) 758

Performance Improvement of Organic Thin Film Transistors Based on Gate Insulator Polymethyl-Methacrylate-co-Glyciclyl-Methacrylate *

LIU Xue-Qiang(刘雪强)[1], ZHANG Tong(张彤)[1*], WANG Li-Jie(王丽杰)[1], LI Ming-You(李明友)[1], FENG Chen-Gang(冯陈刚)[1,2], MA Dong-Ge(马东阁)[2]

[1]*State Key Laboratory on Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, Changchun 130012*

[2]*State Key Laboratory of Polymer Physics and Chemistry, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, Changchun 130022*

(Received 20 November 2007)

Organic thin transistors (OTFTs) on indium tin oxide glass substrates are prepared with polymethyl-methacrylate-co-glyciclyl-methacrylate (PMMA-GMA) as the gate insulator layer and copper phthalocyanine as the organic semiconductor layer. By controlling the thickness, the average roughness of surface is reduced and the OTFT performance is improved with leak current decreasing to 10^{-11} A and on/off ratio of 10^4. Under the condition of drain-source voltage −20 V, a threshold voltage of −3.5 V is obtained. The experimental results show that PMMA-GMA is a promising insulator material with a dielectric constant in a range of 3.9–5.0.

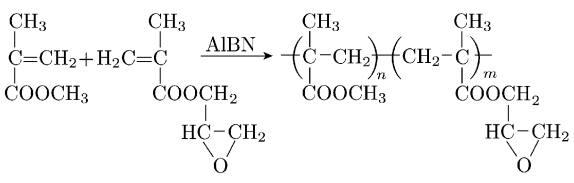
PACS: 85.30.Tv, 81.15.Ef

Organic thin film transistors (OTFTs) have been extensively studied because of their advantages such as low cost process, large-scaled product, room-temperature manufacturing and compatibility with flexible substrate.[1,2] They have been applied in display products, identification tags, and sensors.[3−5] However, there are still some fatal issues, such as low mobility, high leakage current and high driving voltage, that restrict applications of OTFTs. As is known, the electrical characteristics of OTFT devices depend on the material characteristics of the semiconductor and gate insulator layers.[6] A perfect gate dielectric material should have the properties of high dielectric constant, resistivity and well interface quality.[7]

Silicon dioxide SiO_2 and silicon nitride Si_3N_4 obtained by ion-beam deposition and chemical vapour deposition are two kinds of inorganic insulator materials usually used in electronic device. Recently they are being replaced by organic materials in the fabrication of OTFTs, such as polyvinyl alcohol, polyimide, polyvinyl phenol, and polyvinyl carbonate, which generally possess high dielectric constant and high dielectric strength.[8,9]

We use polymethyl-methacrylate-co-glyciclyl-methacrylate (PMMA-GMA) deposited by the method of spin-coating as the gate insulator of OTFTs, and copper phthalocyanine (CuPc) as the active layer obtained by thermal deposition. Source and drain electrodes are MoO_3 and Al films. This fabrication process simplifies the techniques and optimizes the performances of OTFTs significantly.

The cross-section view of OTFT with top contact structure is shown in Fig. 1, which has demonstrated better device performance.[10] The indium tin oxide (ITO) coated glass was used as the device substrate. Firstly, ITO as the gate electrode was formed by photolithography, and the substrates were successively processed by sonicleaning with acetone and ethyl alcohol, soaked in dry air. Subsequently PMMA-GMA with an average molecular weight of 115500 g/mol was dissolved in butyl acetate to obtain the gate insulator film. The PMMA-GMA solutions with different concentrations (2, 2.5 and 3 wt%) were prepared by sonication for 30 min at room temperature in order to obtain the good dispersion. Butyl acetate is a good solvent for PMMA-GMA, which results in a homogeneous solution, easily utilized for spin-coating at room temperature. The gate insulator PMMA-GMA film was deposited on the surface of ITO by spin-coating. The spin-coating speeds and time was controlled by two steps: 300 rpm for 6 s and 4000 rpm for 60 s. The gate dielectric was cross-linked by baking at 120°C for 3 h under ambient conditions and then well solidified. The reaction equation is shown in the following relation. Actual dielectric film thicknesses were about 500 nm, 760 nm and 1000 nm measured by a profilometer (DEKTAK 6 M, made in U.S.A). However, the film would be dissolved when the ITO substrate with the cross-linked polymer film was put in acetone or N-methyl pyrrolidone.



(1)

*To whom correspondence should be addressed. Email: zhangtong@jlu.edu.cn

© 2008 Chinese Physical Society and IOP Publishing Ltd

Azo-bis-isobutryonitrile acts as the activator. The n/m ratio in the above equation is about 10. The cross-linked dielectrics will have radicals left behind, but this effect is not very evident. The devices stored in air 72 h exhibited a similar performance as the fresh one.

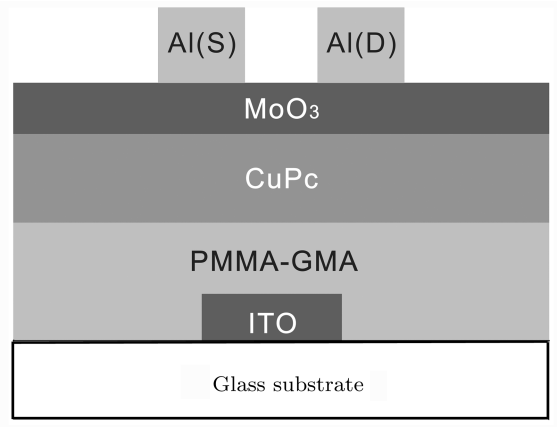


Fig. 1. Schematic device structure of organic thin film transistors.

Organic semiconductor CuPc was thermally evaporated onto the PMMA-GMA surface to the thickness of 500 Å at the rate of 0.8 Å/s. The deposition rate and final film thickness were controlled and tested by a quartz microbalance. The surface morphology of the layers were described by an SPA-400 atomic force microscope (AFM, Seiko, made in Japan) in non-contact mode. Finally MoO_3 (about 10 nm) layers and Al (about 120 nm) layers, which worked as the source-drain contacts, were thermally deposited onto the surface of the CuPc layer through a shadow mask. The configuration of channel length and width of the OTFTs were 400 μm and 1000 μm, respectively. All the deposition processes were carried out under the condition of 3×10^{-4} Torr.

The characteristics of the fabricated OTFTs were measured by Keithley Source Meters (Keithley 2400). To investigate the permittivity of PMMA-GMA film, the capacitance structure of ITO/PMMA-GMA/Al was fabricated under the same condition and investigated by a smart impedance-capacitance-resistance analyser (ZL-5 model, made in Shanghai, China). The test frequency varied from 10 Hz to 10^4 Hz at 1 V. All the measurements were carried out at room temperature under ambient conditions.

The breakdown characteristics of the PMMA-GMA were determined on ITO/PMMA-GMA/Al capacitor with an area of $2 \times 2\,\mathrm{mm}^2$. The curve about dielectric constant of PMMA-GMA capacitor under ac electric fields with the frequency varying from 10 Hz to 10^4 Hz is given in Fig. 2. The value of dielectric constant ranges from 3.9 to 5.0 at overall frequency change. I–V characteristic was also measured by changing the voltage from 0 V to 100 V and dielectric breakdown happened at 94 V. Thus the dielectric strength of 854 kV/mm was obtained from the calculation of the above data (see Fig. 3).

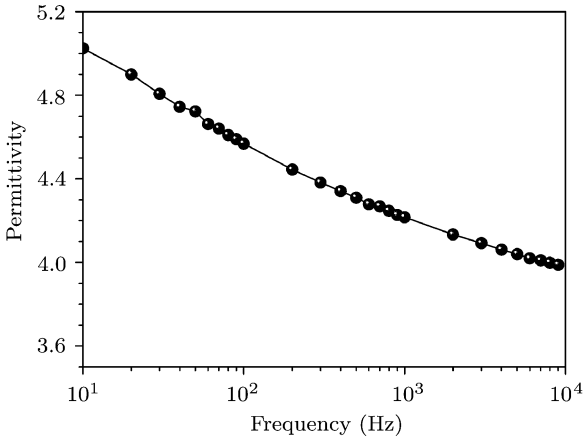


Fig. 2. Permittivity of PMMA-GMA at different frequency.

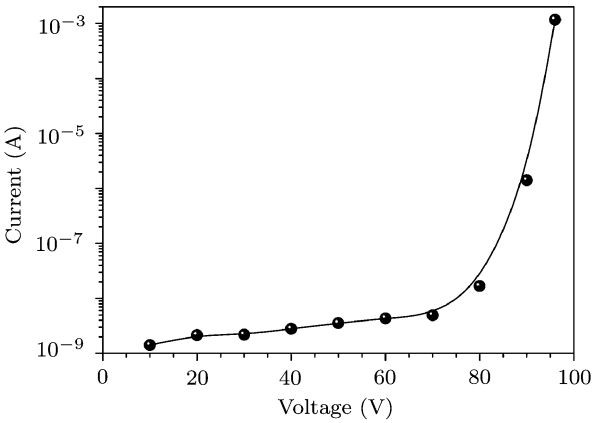


Fig. 3. Breakdown of ITO/PMMAGMA/Al structure capacitor.

Low dielectric surface roughness is an important demand for obtaining high charge carrier mobility in OTFTs.[5] A smooth uniform PMMA-GMA surface (500 nm) image was observed by AFM before CuPc deposition, as shown in Fig. 4. The cross-linking reaction increases the surface roughness lightly, because the average surface roughness of PMMA-GMA film increases from 3.4 Å to 3.8 Å before and after it (see Figs. 4(a) and 4(b)). The CuPc film deposited on the surface of PMMA-GMA was measured by the same way and it has a surface roughness of 4.0 Å, as shown in Fig. 4(c).

Figure 5 shows the drain-source current I_{ds} as a function of the drain-source voltage V_{ds} with different insulator thicknesses (500 nm, 760 nm and 1000 nm). At low V_{ds}, it is observed that I_{ds} increases linearly

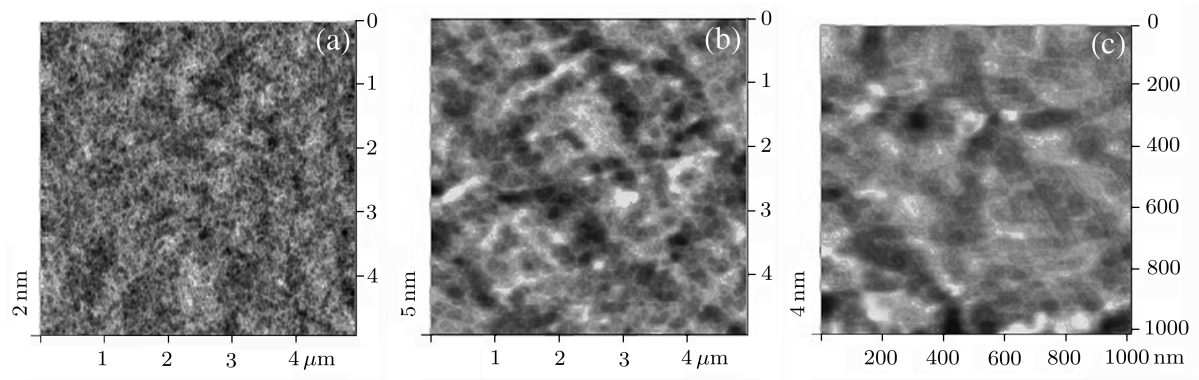


Fig. 4. AFM images of the PMMA-GMA layer (500 nm) surface and CuPc film (50 nm), i.e. 5 μm × 5 μm AFM images of the PMMA-GMA layer before (a) and after (b) cross-linking reaction, (c) 1 μm × 1 μm AFM image of the CuPc film grown on the gate insulator layer (C).

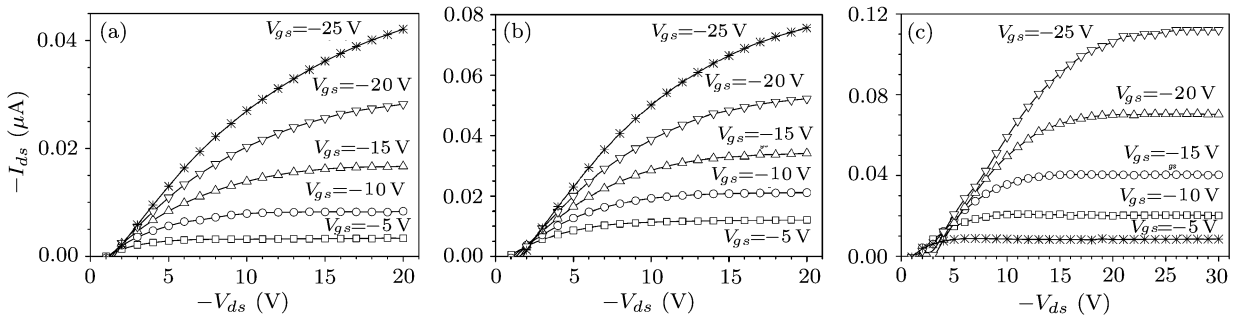


Fig. 5. Output characteristics of the OTFT with varying thicknesses of PMMA-GMA layer from 1000 nm (a), 760 nm (b) and 500 nm (c).

with V_{ds} (in linear region). As V_{ds} becomes increasingly negative, I_{ds} tends to be saturated (saturation region) due to the pinch-off of the accumulation, which is described by the following equation:

$$I_{ds} = \frac{WC_i}{2L}\mu(V_{gs} - V_T)^2, \tag{2}$$

where μ is the field effect mobility, W is the channel width, L is the channel length, and C_i is the capacitance per unit area of the insulating layer, and V_T is the threshold voltage. The results indicates that the channel currents in the saturation region (at $V_{gs} = -25$ V) increases as the thickness of insulator layer decreases from 1000 nm to 500 nm. For the OTFT with the 500 nm PMMA-GMA layer, the field-effect mobility $\mu = 3.1 \times 10^{-2}$ cm^2/Vs can be calculated from the data in saturation region ($V_{ds} = -20$ V and $V_{gs} = -25$ V). When V_{ds} is -20 V, the maximum operating frequency 0.62 MHz can be obtained by

$$f_{\max} \cong \frac{\mu V_{ds}}{2\pi L^2}. \tag{3}$$

Here V_T can be obtained from Fig. 6. characteristics of I_{ds} and $(I_{ds})^{1/2}$ versus V_{gs} for CuPc OTFT with PMMA-GMA dielectric gate insulator at $V_{ds} = -20$ V are shown in Fig. 6. The on/off ratio is about 10^4 and the threshold voltage is -3.5 V. The threshold voltage is related with the thickness of the gate insulator, interface charge and charge in the gate insulator. It has a linear relationship with the thickness if there is no bulk charge in the gate insulator.[11,12] The threshold voltage -3.5 V can be obtained by decreasing the gate insulator thickness varying to 500 nm. Compared with other polymer dielectric CuPc OTFT, the electronic properties such as mobility and on/off ratio of the OTFT device with PMMA-GMA gate insulator have been improved.[13,14]

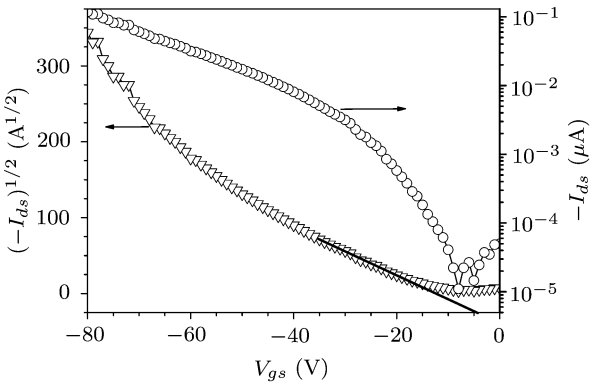


Fig. 6. Electrical transfer characteristics of OTFT with the PMMA-GMA gate insulator.

In summary, we have investigated the electric characteristics of CuPc TFTs based on the polymeric gate insulator PMMA-GMA film, which has low surface roughness and high dielectric strength, fabricated through cross-linking reaction at 120°C. The TFTs also exhibit low threshold voltage and reduce the leakage current. Estimated from the saturation current, the field-effect hole mobility is about $3.1 \times 10^{-2}\,\mathrm{cm}^2/\mathrm{Vs}$. Thus the polymeric material PMMA-GMA has a potential application value in the fabrication of OTFTs.

We gratefully thank Professor Daming Zhang at State Key Laboratory on Integrated Optoelectronics, Jilin University, for supplying the PMMA-GMA materials.

References

[1] Xu M S, Nakamura M K and Kudo K H 2007 *Thin Solid Films* (accepted)
[2] Kwon T G, Baek C H and Lee H H 2007 *Org. Electron.* **8** 615
[3] Kang H, Han K, Park J E and Lee H H 2007 *Org. Electron.* **8** 460
[4] Zan H W, Yen K H, Liu P K, Ku K H , Chen C H and Wang J H 2007 *Org. Electron.* **8** 450
[5] Stallinga P and Gomes H L 2007 *Org. Electron.* **8** 300
[6] Sung J H, Park S J, Park J H, Choi H J and Choi J S 2006 *Synth. Metals* **156** 861
[7] Byun H S, Xu Y X and Song C K 2005 *Thin Solid Films* **493** 278
[8] Lee J H, Kim S H, Kim G H, Lim S C, H. Lee and Zyung T 2003 *Synth. Metals* **139** 445
[9] Lim S C, Kim S H, Lee J H, Yu H Y, Park Y and Kim D 2005 *Mater. Sci. Engin.* B **121** 211
[10] Kim H Y, Lee D H and Kim J K 2006 *Polymer* **47** 5108
[11] Choi Y J, Kwak W K, Cho K S, Kim S K and Jang J 2000 *IEEE Electron Device Lett.* **21** 18
[12] Yuan J F, Zhang J, Wang J, Yan X J, Yan D H and Xu W 2003 *Appl. Phys. Lett.* **82** 3967
[13] Yasud T K and Tsutsui T S 2005 *Chem. Phys. Lett.* **402** 395
[14] Puigdollers J, Voz C, Fonrodona M, Cheylan S, Stella M, Andreu J, Vetter M and Alcubilla R 2006 *J. Non-Crystalline Solids* **352** 1778