



Study on Modification of SiO₂ Thin Film Surface Morphology and Its Electrical Performance

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Silicon dioxide thin film was prepared by thermal growth method and PMMA film layer was prepared by spin coating. Silane coupling agent octadecyltrichlorosilane (OTS) be used to modified the surface of SiO₂. The result of atomic force microscope (AFM) showed that using PMMA and OTS modified SiO₂ layer can effectively improve the surface flatness and reduce the surface energy. The PMMA insulating layer reduces the contact barrier between the organic semiconductor insulating layer and the growth of thin films of copper phthalocyanin became more consistent. Compare with the single SiO₂ insulation layer, thin film transistor devices on SiO₂/PMMA and SiO₂/OTS compound insulation layer with improved carrier mobility and lower threshold voltage. Experimental results showed that using double insulation layer structure can effectively improve the performance of organic thin film transistor.

Keywords: SiO₂, PMMA, Octadecyltrichlorosilane, Insulator.

INTRODUCTION

In recent years there has been considerable interest in organic-based thin film electronics for low-cost and large area electronic application¹⁻³. The mechanical flexibility of organic materials makes them compatible with plastic substrates for light weight and foldable products⁴⁻⁸. SiO₂ is widely used in organic thin film transistor (OTFT) as insulation material, but its high surface energy characteristics, making the mobility of the device has been affected. To reduce the surface energy is a research focus. In the present work, we use PMMA and octadecyltrichlorosilane modify the SiO₂ thin film. PMMA are particularly promising materials for flexible and low-cost fabrication such as spin coating, spray coating and printing⁹⁻¹². A SiO₂ thin film can be used as the first insulator¹³ and a PMMA been used as the second insulator. Octadecyltrichlorosilane is particularly interesting material because it can bond to oxide surfaces and has the ability to form Si-O-Si bond which can influence the characteristics of insulator films. The high-quality octadecyltrichlorosilane monolayer can also be spontaneously formed insulation. Here, a solution technique was used to form octadecyltrichlorosilane monolayer.

EXPERIMENTAL

The equipment of thermal grown shown in Fig. 1, high purity oxygen ($\geq 99.99\%$) be used as reactive gases, wet oxygen get by dry oxygen through the water which temperature

about 95 °C in water bottles. Before thermal grown, heavily doped *p*-Si (100) wafers treated followed by acetone, hydrochloric acid, ethanol (all analytical grade) and deionized water and then processed in the ultrasonic about 20 min. Thermally grown by dry oxygen wet oxygen-dry oxygen oxidation, the temperature control between 1100 and 1150 °C. In the end of the experiment, high purity nitrogen ($\geq 99.99\%$) pass into and then holding for 1 h. Finally, silicon dioxide film been made as the inorganic insulating layer, film thickness be tested by the ST2000-DLXn thickness measurement equipment.

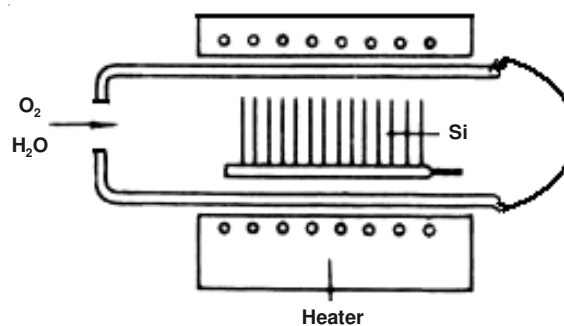


Fig. 1. Schematic setup for the thermal growth system

Preparation of PMMA films: In the experiment, PMMA film been prepared by spin-coating (Fig. 2). PMMA reagents dissolved in chloroform solution, filtered after the solution is

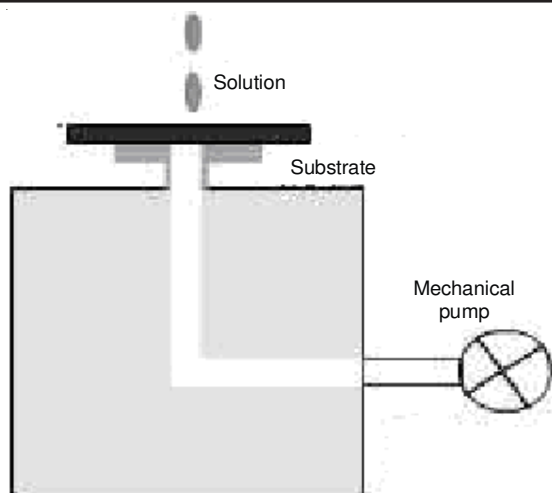
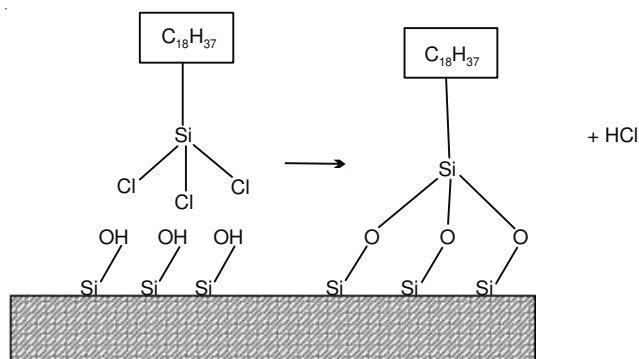


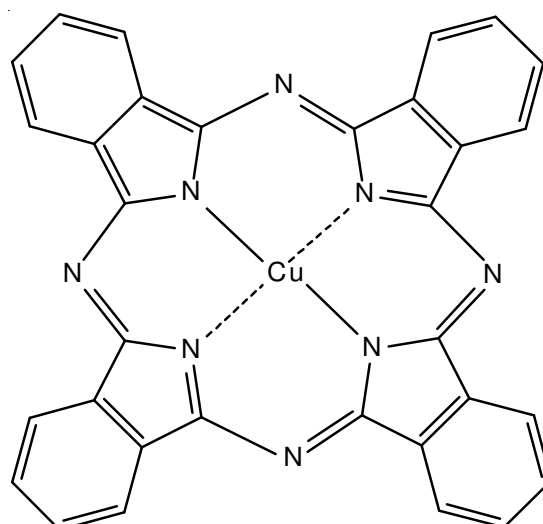
Fig. 2. Schematics of spin-coating process

made. Spin coating machine set rotation speed of 2000 rpm units/min, spin-around time is 40 s. After spin coating, the substrate into the oven baking at 80 °C for 5 h. The thickness of PMMA film been tested about 50 nm.

Preparation of octadecyltrichlorosilane films: The SiO₂ surface was ultrasonically cleaned with acetone, ethanol and deionized water in that order respectively, subsequently the wafer was treated by UV for 10 min. Then the SiO₂ surface was treated with octadecyltrichlorosilane by immersing the sample in a 2 % solution of CHCl₃:C₆H₁₄ = 1:3 for 12 h at room temperature in an anhydrous ambient. This process is shown in Fig. 3.

Fig. 3. Chemical process between octadecyltrichlorosilane and surface of SiO₂

Preparation of organic thin film transistor: Organic thin film transistor based on SiO₂, SiO₂/PMMA and SiO₂/OTS been made (Fig. 4). The active layer material is copper phthalocyanin. Copper phthalocyanin was purchased from Shanghai Chemical Reagent Company J & K, more than 95 % purity. Copper phthalocyanin was purified twice and prepared by vacuum deposition at a rate of 2 nm/min under a pressure of 6 × 10⁻⁴ Pa at room temperature and the thickness of resulting films between 35 and 45 nm. Organic thin film transistors with channel length L = 0.035 mm and channel width W = 5 mm were fabricated by a mask. On top of this surface, gold was deposited through this mask to give the source (S) and drain (D) electrodes. All test conditions were at room temperature and open air environment.



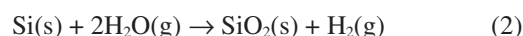
Structure of copper phthalocyanin

Au	Au
CuPc	
PMMA (OTS)	
SiO ₂	
Highly doped Si	

Fig. 4. Schematic structure for organic thin film transistors

RESULTS AND DISCUSSION

Equation of silicon react with oxygen or water vapor in thermal oxidation:



The growth of silicon dioxide is a surface reaction only. However, after the SiO₂ thickness begins to build up, the arriving oxygen molecules must diffuse through the growing SiO₂ layer to the silicon surface in order to react. As silicon dioxide film grown in the surface of silicon, the silicon dioxide film prevent water vapor contact to the silicon, water molecules through the gap of silicon dioxide layer, reach the surface of silicon, silicon reacts with the new layer, increasing the thickness of the silicon dioxide layer. Because water molecules in silica is the diffusion coefficient of oxygen atoms in the silica diffusion coefficient thousand times more, so wet oxygen oxidation rate faster than dry oxidation rate of oxygen. Although the time of dry oxidation reaction is longer, the resulting of silica layer is dense. Wet oxygen oxide growth is faster, but the oxide film is looser. In this study, wet oxygen and dry oxygen used in conjunction with dry oxygen-oxygen wet-dry oxygen oxidation in order. Low-resistance *p*-type boron doped

silicon been used in our experiments for thermal oxidation, segregation of boron to silica in the silica bonding weakened the oxidant in the oxide layer easily spread and thus oxidation rate increases, by reducing the oxidation time.

Morphology of SiO₂, PMMA and octadecyltrichlorosilane be observed using an atomic force microscope (AFM) (Fig. 5). From Fig. 5 the surface roughness of PMMA and octadecyltrichlorosilane are smaller and smoother than the surface of SiO₂. The smoother the surface insulation, flatness better film in less defects and traps, it is easy to carriers through.

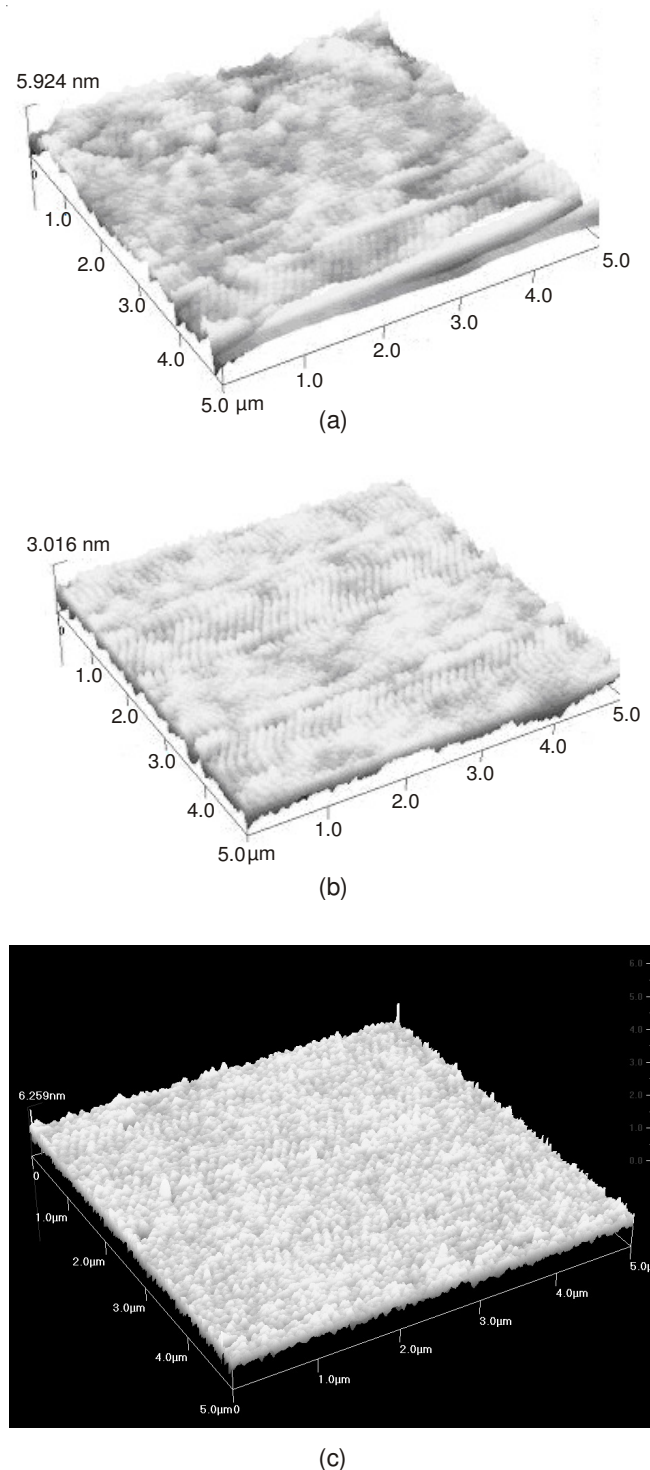


Fig. 5. 5 μ \times 5 μ atomic force microscopy image of SiO₂ (a) and PMMA (b) and octadecyltrichlorosilane (c) surface

PMMA influences growth behavior of copper phthalocyanin thin films at the gate dielectric interface and adhesion of copper phthalocyanin to the substrate, for the interface has lower energy boundary and PMMA layer has a low-dielectric constant similar to organic semiconductor. Morphology of copper phthalocyanin thin films is observed using an atomic force microscope (AFM) (Fig. 6). The PMMA insulating layer reduces the contact barrier between the organic semiconductor and insulating layer, so that the growth of thin films of copper phthalocyanin is more consistent. The mobility is strongly dependent on the order and orientation of copper phthalocyanin¹⁴⁻¹⁶. Copper phthalocyanin thin films on the top of PMMA layer are polycrystalline. From Fig. 6 we can see that the surface roughness of copper phthalocyanin on PMMA is smaller and more smooth than the surface of copper phthalocyanin on SiO₂.

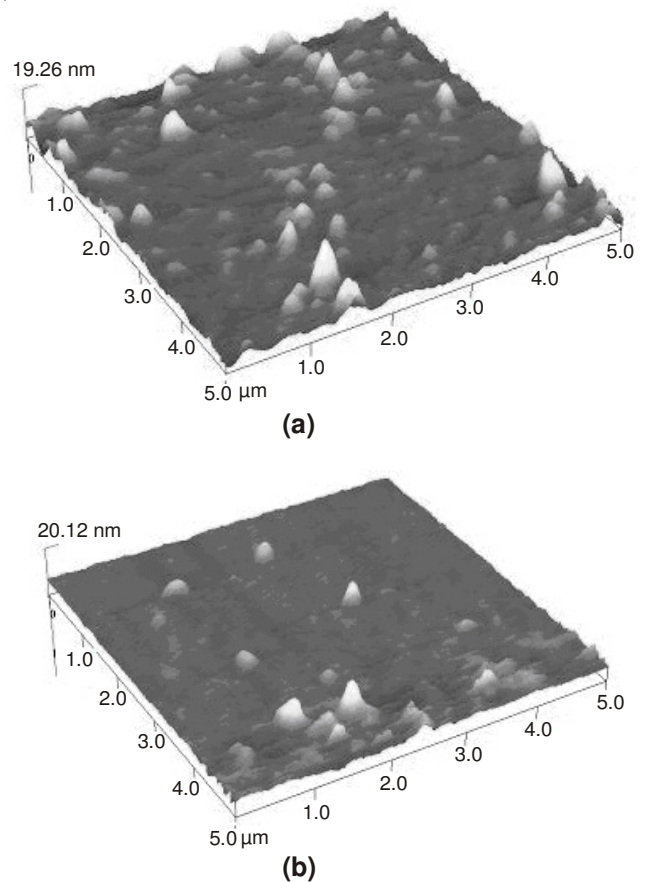


Fig. 6. 5 μ \times 5 μ atomic force microscopy image of copper phthalocyanin on SiO₂ (a) and copper phthalocyanin on PMMA (b)

The mobility was measured in the saturation regime, which is modeled by the equation:

$$I_{DS} = \frac{WC_i}{2L} \mu (V_{GS} - V_T)^2 \quad (2)$$

where μ is the field-effect mobility, L and W are channel length and width, C_i is the insulator capacitance per unit area and V_T is the extrapolated threshold voltage. Field-effect mobility of the device with SiO₂/PMMA insulator is 4.0×10^{-3} cm²/Vs and threshold voltage is -9.9 V. Field-effect mobility of the device with SiO₂ insulator is 2.1×10^{-3} cm²/Vs and threshold voltage is -11.2 V. Field-effect mobility of the device with SiO₂/OTS insulator is 4.3×10^{-3} cm²/Vs and threshold voltage

is -9.5 V. So the mobility of SiO₂/PMMA and SiO₂/OTS insulator almost twice than that of the device with single SiO₂ insulator. The results showed the leakage current also been reduced after modified by PMMA layer.

Conclusion

SiO₂/PMMA and SiO₂/OTS composite insulation film have been fabricated. After modification, the surface of the SiO₂ films become more smooth, with reduced surface energy. The PMMA insulating layer reduces the contact barrier between the organic semiconductor insulating layer and the growth of thin films of copper phthalocyanin became more consistent. Field-effect mobility of the device with PMMA modified insulator is $4.0 \times 10^{-3} \text{ cm}^2/\text{Vs}$, which is twice than that of the device without PMMA insulator. The leakage current also been reduced after modified by PMMA layer.

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