# Self-Assembled TiO<sub>2</sub> and Polyelectrolyte Multilayer as OTFT Gate Insulator

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### **Abstract**

Modified self-assembled TiO<sub>2</sub> and polyelectrolyte multilayer film have been used as OTFT insulator. Both films were used as gate insulator and their thickness were reduced to the order of 10nm. The operating voltage of OTFT was substantially reduced due to nanoscale thickness of titanium oxide and polyelectrolyte multilayer. Pentacene-based OTFT characteristics will be discussed.

## 1. Introduction

Titanium oxide films can be produced with the application of vacuum techniques, such as chemical vapor deposition, sputtering or flame hydrolysis. The sol-gel method can be an alternative for those techniques. Inorganic TiO<sub>2</sub> has often been used as OTFT insulator due to much higher gate capacitance than polymers whereas it has relatively high leakage current. In the case of polymer film, main drawback is a high operating voltage in device due to its thickness and low dielectric constant. In the sol-gel method, dielectric films are produced from liquid phase at ambient temperature[1].

In this sense, TiO<sub>2</sub> self-assembled film may be deposited as gate insulator with nano-scale thickness. This "surface sol-gel process" is composed of four steps: chemisorptions of alkoxide, rinsing, hydrolysis of the chemisorbed alkoxide, and drying[2]. Also, polyelectrolyte can be also self-assembled using oppositely charged polyelectrolytes and the film may be further chemically modified. In this work, self-assembled titanium oxide or/and polyelectrolyte multilayer film may have a property to overcome their limitations and have been utilized to apply for gate insulator of organic thin film transistor (OTFT).

## 2. Experimental

PAH, PAA and cyclohexyl isocyanide (CIC) were purchased from Aldrich Chemical. Glutaraldehyde (GA) as crosslinker was phrchased from Sigma-Aldrich. 20mM polyeletrolyte solutions were prepared from deionized water.

The desired number of layers had been assembled and then, the surface was treated with GA and CIC. In order to be gate insulator, the PEM film was further treated with GA and CIC. To stabilize PAH/PAA film, it was immersed in aqueous glutaraldehyde(GA) 2.5% solution for the desired time at room temperature. After surface treatments, the substrates were dried with filtered air and subsequently dried at 200 ℃ oven at least 2h.

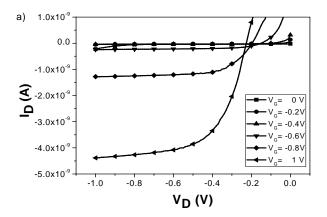
For  $TiO_2$  films, titanium tetrabutoxide 97% as  $TiO_2$  precursor was purchased from Aldrich Chemical. 100mM titanium tetrabutoxide solutions were prepared from mixed solvent(EtOH:Toluene=1:1).  $TiO_2$  film was prepared by spin-coating using 100mM titanium tetrabutoxide solutions and further treated with EtOH and  $H_2O$ . Then substrates were dried at  $100\,^{\circ}\text{C}$  oven.

For OTFT device fabrication, polymer dielectric layer was spin-coated on Si wafer. Pentacene was vacuum deposited on top of the gate dielectric through a shadow mask. The surface of films was investigated by AFM. LCR meter was used for capacitance measurements. Probe station system was used to obtain the characteristics of OTFT devices.

### 3. Results and Discussion

Self-assembled TiO<sub>2</sub> film with titanium tetra

butoxide was deposited using spin-coating and surface treatment. To make self-assembled polyelectrolyte multilayer, polyallylamine hydrogenchloride(PAH) and polyacrylic acid(PAA) were used on Si wafer and the multilayer was chemically treated to cross-link between the layer. According to the surface morphology of (a)TiO<sub>2</sub> and (b)[PAH/PAA]<sub>6.5</sub>, the surface roughness of spin-coated method was decreased for titanium oxide, which gave the denser film.



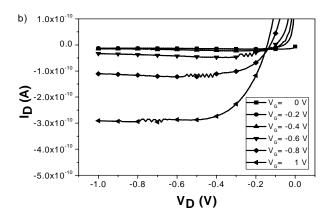
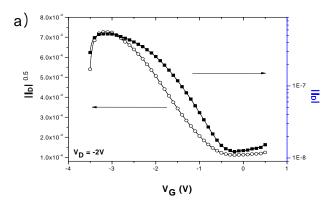


Fig. 1. Current-voltage characteristics as a function of gate voltage of pentacene based OTFT. a) TiO2 using sol-gel reaction b) [PAH/PAA]<sub>6.5</sub>

Figure 1 shows the current-voltage characteristics as function of the gate voltage. The devices with both films worked at very low operating voltage. This is due to the fact of 10 nm scale thickness of gate insulator

As shown in Figure 2, even at -2 source-drain voltages both devices performed as OTFT using self-assembled TiO2 and polyelectrolyte multilayers. The electric characteristics of OTFT with pentacene as organic semiconductor were listed in Table 1.



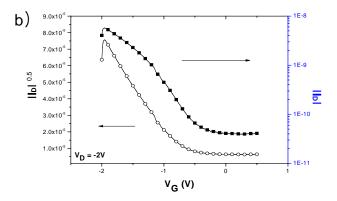


Fig. 2. Electric characteristics of pentacene-based OTFT using 2 differenctt gate insulator: a)TiO2 using sol-gel reaction, b)[PAH/PAA]<sub>6.5</sub>.

TABLE 1. Electric characteristics of pentacenebased OTFT with 3 different types of gate insulator

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Gate insulator	$TiO_2$	[PAH/PAA] <sub>6.5</sub> + surface treatment	Poly(phenyl maleimide- co-MMA)
$I_{\rm on}/I_{\rm off}$	$4.2*10^{1}$	$9.9*10^{1}$	$6.8*10^3$
slope	-3.1*10 <sup>-4</sup>	-5.7*10 <sup>-5</sup>	-1.1*10 <sup>-4</sup>
SS (V/dec)	1.13	0.57	6
$V_{T}(V)$	$-3.1*10^4$	-0.62	-13
Mobility (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )	6.3*10 <sup>-2</sup>	2.7*10 <sup>-3</sup>	2.0*10 <sup>-1</sup>
Insulator permittivity (ε)	41	5.2	2.8

## 4. Summary

Titanium oxide and polyelectrolyte multilayer were used as dielectrics for OTFT in order to reduce the

thickness to nano-scale film fabrication. With sol-gel method, titanium oxide film became inorganic from organic materials by water and thermal treatment. The polylectrolyte multilayer was converted to be chemically cross-linked and non-electrolyte. Both films were successfully fabricated as OTFT gate insulator and the devices worked at low voltage. Using titanium butoxide solution, the titanium oxide film was deposited 10nm scale by sol-gel reaction. The device with 80nm thickness can be operated at -Compared with polyelectrolyte multilayer dielectrics, the surface roughness of Ti dielectrics was somewhat reduced and need to improve on/off ratio and properties of electric insulation. Titanium oxide film known to be high-k material can be deposited with solution process coating. Furthermore, titanium oxide with PEM is expected to have better characteristics as organic-inorganic dielectrics.

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#### 5. References

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