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Electrical Characteristics of Organic Thin-Film Transistors with Polystyrene/Sol-Gel Derived Titania Composite Insulator

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In this study, a sol-gel process is introduced to fabricate the composite film of polystyrene and titania, using titanium tetraisoproxide as a titania precursor. Pentacene-based organic thin-film transistor (OTFTs) with the composite film as a gate insulator operates at a low voltage below −4 V and exhibits high performances, such as threshold voltage of −0.9 V, subthreshold slope of 0.2 V/decade, and mobility of 4.7 cm^2/Vs, which are mainly attributed to a high dielectric property of the fabricated composite insulator. These results demonstrate that sol-gel derived composite insulators are essentially promising for low-voltage OTFTs with high performances.

Keywords Composite; gate insulator; organic thin-film transistor; sol-gel

Introduction

Organic thin-film transistors (OTFTs) have enormous market potential in a wide range of applications, such as the driving elements for flexible displays, radio-frequency identification tags, and large-area sensors [1]. During the past two decades, there has been intensive research into developing materials and processing techniques for enhancing the performance of OTFTs, and encouraging device performances, comparable or even superior to those of amorphous silicon TFTs, have been reported [2]. Nevertheless, high operating voltages larger than −10 V are still required for OTFTs owing to low charge carrier mobilities of organic semiconductors. Accordingly, high-dielectric-constant (high-*k*) insulators are inevitably necessary to enhance the field-induced carrier density in OTFTs. Recently, a number

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of studies has focused on developing organic/inorganic composite insulators with high-*k* properties, such as poly(methyl methacrylate)/Ta_2O_5 [3] and poly(4-vinylphenol)/TiO_2 (titania) [4], because they promise both the superior dielectric properties of inorganic particles and the processability of organic materials. However, their use still faces critical issues associated with the aggregation and non-uniform dispersion of the inorganic particles, which deteriorate the device performance. One way to circumvent these drawbacks is to utilize a sol-gel process to obtain a uniform distribution of the inorganic material in the composite film.

In this work, we report the characteristics of OTFTs with the polystyrene/titania composite film as a gate insulator, which was formed by a sol-gel process using titanium tertraisopropoxide (TTIP) as a precursor of titania. The composite insulator exhibits an increased dielectric property compared to that of the bare polystyrene and contributes to low-voltage operating OTFTs with high performances.

Experimental

For the preparation of a sol-gel precursor solution, polystyrene was dissolved in chloroform and then TTIP was mixed with the polystyrene solution. The composition ratio of sol-gel precursor solution by weight percentage is approximately 20:1 (polystyrene solution : TTIP). The molecular structures of polystyrene and TTIP are shown in Figures 1 (a) and (b), respectively. To fabricate OTFTs, a 150-nm-thick Al gate electrode was thermally evaporated on a glass substrate using the first shadow mask. Two different gate insulators, such as 500-nm-thick polystyrene layer and 550-nm-thick polystyrene/titania composite layer, were formed by spin-coating in ambient air of the relative humidity of about 40%. The sol-gel derived titania was immediately formed in the composite film by the hydrolysis reaction of TTIP as shown in Figure 1 (c). Note that solvent elimination was carried out under a base pressure of about 2×10^{-3} Torr. Pentacene was thermally evaporated through the second mask onto the gate insulator at a deposition rate of 1.0 Å/s, up to a thickness

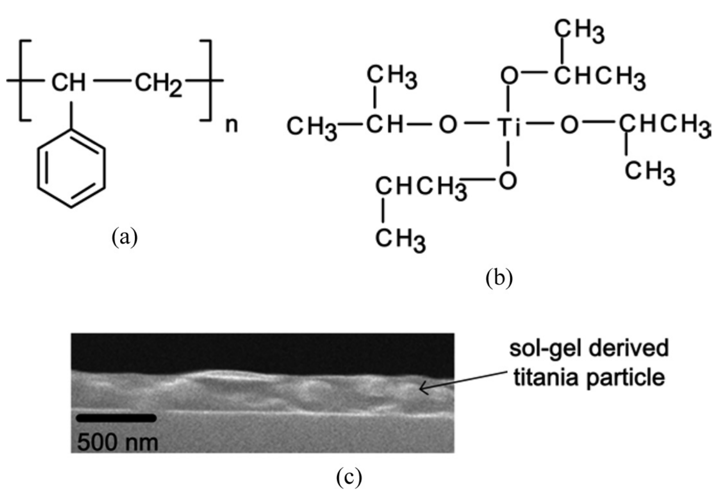

Figure 1. The molecular structures of (a) polystyrene and (b) TTIP, and the cross-sectional scanning electron microscopic image of (c) the polystyrene/titania composite film.

of 60 nm. Subsequently, a 50-nm-thick Au layer was thermally evaporated through the third mask for the source and drain contacts. The channel length and width of OTFTs are 90 μm and 300 μm, respectively. The electrical characteristics of our devices were measured with impedance analyzer (HP 4192LF, Agilent Technology) and semiconductor analyzer (EL 421C, Elecs Co.) in ambient air.

Results and Discussion

The sandwich-structured capacitors of Al/insulator/Au were fabricated to investigate the dielectric properties of the bare polystyrene and the composite insulators, and the experimental capacitance versus frequency plots are shown in Figure 2. It is evident that the capacitance of the composite insulator is larger than that of the bare polystyrene. The calculated dielectric constant of the composite insulator was about 10.6 at 100 kHz, while that of the bare polystyrene was calculated about 2.7, which is attributed to a high-k of sol-gel-derived titania.

Typical transfer characteristics of OTFTs with different gate insulators are shown in Figure 3. For the transfer characteristic measurements, the drain currents (I_D) of the fabricated OTFTs were measured at the drain voltage (V_D) of -3 V, where the gate voltage (V_G) was swept from 1.5 to -4 V with the sweep step of 0.05 V. From the curve of the square root of drain current versus gate voltage, the mobility (μ_{eff}) in the saturation region is estimated using Eq. (1):

$$I_{D,sat} = \frac{W \mu_{eff} C_i}{2L} (V_G - V_T)^2 \quad (1)$$

where C_i is the capacitance of the gate insulator per unit area, V_T is the threshold voltage [5]. The threshold voltage was extracted from the corresponding plot of $(I_D)^{1/2}$ versus V_G by extrapolating to $I_D = 0$ and the subthreshold swing, the inverse of the subthreshold slope $[\partial \log I_D / \partial V_G]$, represents the change in V_G needed to change I_D by a factor of 10. As can be seen in Figure 3(a), the device with the

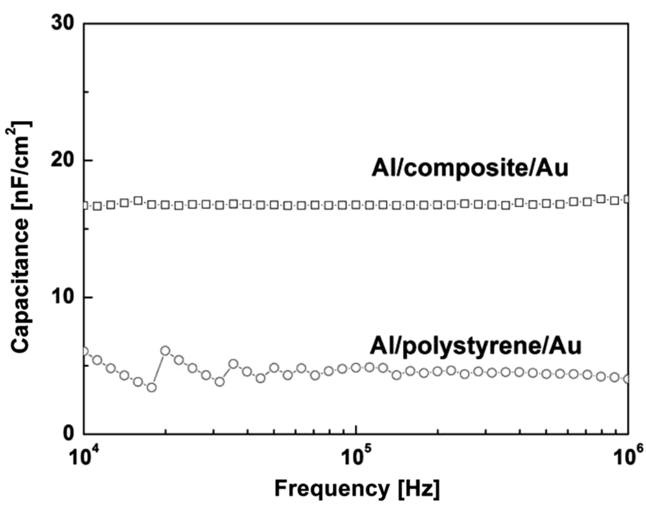

Figure 2. The capacitance-frequency plots of MIM capacitors with two different insulators.

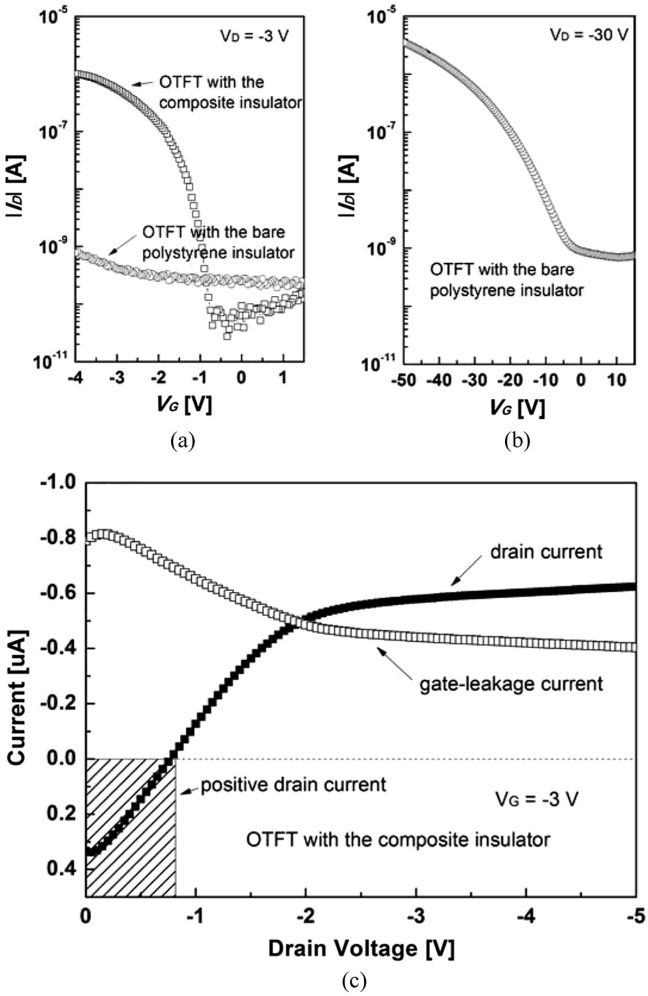

Figure 3. Typical transfer characteristics of OTFTs with different gate insulators (a) at low applying voltages and (b) at relatively large applying voltages. (c) Typical output characteristics and gate-leakage currents for the device with the composite gate insulator.

composite insulator operates even at low applied voltage and shows the extremely low threshold voltage of $-0.9\,\mathrm{V}$ and the pronounced subthreshold slope of $0.2\,\mathrm{V/}$ decade, while the device with the bare polystyrene still stays at off-state in this region. In addition, the mobility for the device with the composite insulator is calculated to be about $4.7\,\mathrm{cm}^2/\mathrm{Vs}$. Note that the device with the bare polystyrene insulator shows comparable characteristics at relatively large-voltage region (see Fig. 3(b)). However, the gate-leakage current of the device with the composite insulator is acute at low drain voltage region below $-1\,\mathrm{V}$, which led to undesired positive currents in the output characteristics as shown in Figure 3(c). It is thought that the thicknesses of polystyrene and titania layers in the composite insulator should be optimized by modifying the composition ratio of sol-gel precursor solution to reduce the gate-leakage current. Comparisons of parameters of two devices are listed in Table 1.

Table 1. Performance parameters for the fabricated TFTs with different gate insulators

Gate insulator	Mobility (cm^2/Vs)	V_T (V)	Subthreshold slope (V/decade)	I_{On}/I_{Off}	V_D
Polystyrene	–	–	–	–	−3 V
	0.2	−17.2	7.9	0.8×10^4	−30 V
Composite	4.7	−0.9	0.2	2.0×10^4	−3 V

Conclusion

From the results so far achieved, it is thought that the sol-gel process can be an effective method to prepare organic/inorganic composite films, which can contribute to improve the performance of OTFTs. However, the properties of composite films are sensitive to the composition ratio of a precursor (such as TTIP) to host polymers, which might cause an increase in the gate-leakage currents in OTFTs. We believe that further optimizations in the composition ratio of the sol-gel precursor solution are essentially necessary to improve the composite film quality for OTFT applications. For further works, stability issues should be investigated within the frame work of the storage stability of the OTFTs with the sol-gel derived composite insulators because such composite films might be influenced by ambient moisture.

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