Characteristics of Pentacene Organic Thin-Film Transistors with PVP-TiO_2 as a Gate Insulator

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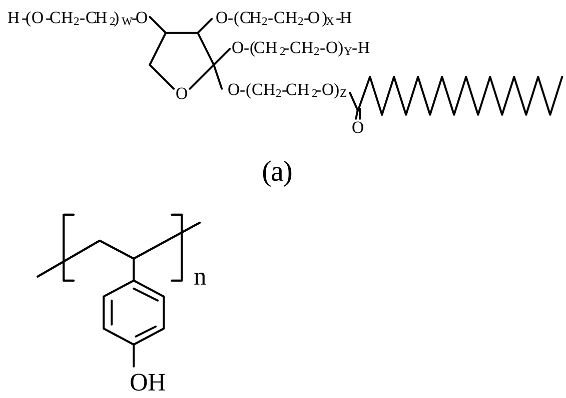
Abstract
The performance of OTFT with PVP-TiO_2 composite, as a gate insulator, is reported, including the effect of surfactant for synthesizing the composite material. According to our investigation results, it was one of critical issues to prevent the aggregation of TiO_2 particles during the synthesis process. From this point of view, TiO_2 particles were treated using Tween80, as a surfactant, and we could reduce the aggregated TiO_2 clusters. As a result, the OTFT with the composite insulator showed the threshold voltage of about -8.3 V and the subthreshold slope of about 1.5 V/decade, which are the optimized properties compared to those of OTFTs with bare PVP, in this study. It is thought that these characteristic improvements are originated from the increase in the dielectric constant of the PVP-based insulator by compositing with high-k particles.

1. Introduction
During the past few years, the performances of organic thin-film transistors (OTFTs) have been improved drastically through focused research efforts [1-3]. At the early stage of research for OTFTs, many groups had made studies of OTFTs with inorganic insulators, such as SiO_2, SiN_x, and so on. But recently, inorganic insulators are being replaced by polymeric materials due to some problems associated with manufacturing complexity, compatibility with plastic substrate, and processing cost. *Klauk* et al. have reported the excellent electrical characteristics of OTFTs with cross-linked poly(4-vinylphenol) (PVP) gate insulator, providing the field-effect mobility as large as 3 cm^2/Vs, the subthreshold swing as low as 1.2 V/decade, and the on/off current ratio of 10^5, which are comparable or even surpassing those of TFTs with hydrogenated amorphous silicon (a-Si:H) [4]. However, in spite of eye-opening progress in the performance of OTFTs with polymeric gate insulators, high threshold voltage due to low dielectric properties of polymeric insulators is one of subjects to be solved.

In the present study, PVP-TiO_2 composite was synthesized. The particle size of TiO_2 is about 30~50 nm and poly(oxyethylene)(20)-sorbitane monooleate (Tween80) is used as a surfactant to treat the surface of TiO_2 particles for their uniform distributions in the solution. The dielectric properties of PVP-TiO_2 composite and the performance of OTFTs with the PVP-TiO_2 composite insulator are presented. The molecular structures are shown in Figure 1.



(a)

(b) (c)

Figure 1. The molecular structures of (a) Tween80 [W+X+Y+Z=20], (b) PVP and (c) pentacene.

2. Experimental Details
In order to synthesize PVP-TiO_2 composite solution, 1 wt% of TiO_2 particles (Ishihara Sangyo Kaisha) and Tween80 were dispersed into ethanol and then sonicated for 2 hrs. Tween80, which is nonionic, was used as a surfactant to treat the surface of TiO_2 particles. The as-received TiO_2 particles show hydrophobic property and the hydrophobic group of Tween80 molecule is generally attached to the surface of TiO_2 particle as shown in figure 2(a). Therefore, the surface-treated TiO_2 particles in ethanol can be

uniformly distributed by the hindrance of the attached Tween80 molecules among the vicinity surface-treated TiO_2 particles. And PVP (Sigma Aldrich) dissolved in the ethanol solvent mixed with TiO_2, and the composite solution was stirred via magnetic stirring for 12 hrs at room temperature.

For the fabrication of OTFTs, an Al gate electrode was thermally evaporated to be of 1500 Å thickness on a glass substrate using the first shadow mask. A 2000-Å-thick gate insulator was formed by spin-coating and baked at 60℃ for 10 min. and consecutively at 100℃ for 20 min. in a vacuum dry oven. Pentacene layer as an organic semiconductor was thermally evaporated through the second mask onto the insulator at a rate of 1.0 Å/s and its thickness was about 600 Å. Subsequently, a 500-Å-thick Au layer was thermally evaporated through the third mask for the source and drain contacts. All the deposition processes were carried out at a base pressure of about 1.6×10^{-6} Torr. All the OTFTs have the top-contact geometry, where the channel length (L) and width (W) are 90 μm and 300 μm, respectively. The molecular structures and the cross section of the fabricated OTFT are shown in figure 2(b).

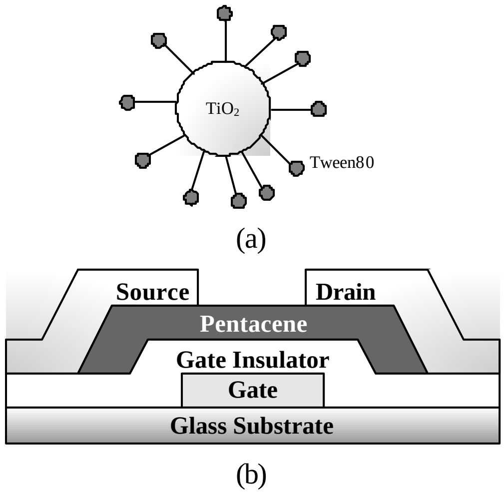


Figure 2. The schematic of (a) the surface-treated TiO2 with Tween80 and the cross-sectional image of (b) the fabricated OTFT.

3. Results and Discussion

3.1 The Composite without Tween80

First of all, the PVP-TiO_2 composite was synthesized without Tween80 and utilized as a gate insulator for OTFT. The dielectric constant of the composite without Tween80 was measured about 6.7, which is relatively increased value compared to that of PVP (ε_r ~ 4.0). It is considered that the TiO2 (ε_r ~ 50) particles induce the increase of dielectric constant of the composite film. Typical output and transfer characteristics of OTFTs adopted the composite without Tween80 are shown in figure 3. It is clearly seen that the device performance was deteriorated due to the critical gate leakage current. Atomic force microscopic (AFM) images showed that the deteriorated device performance might be resulted from the aggregated TiO_2 particles (not shown here). Therefore, we can confirm that it is necessary to utilize a surfactant for preventing the aggregation of TiO_2 particles.

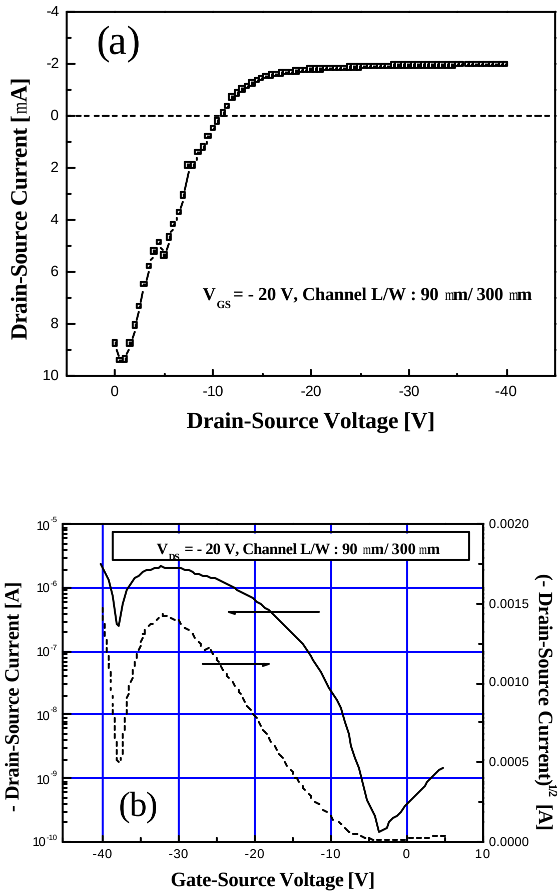
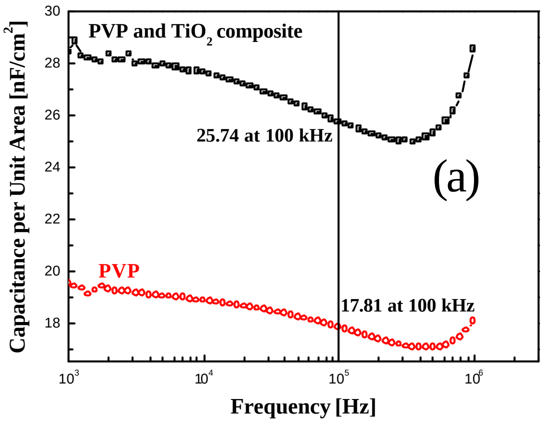


Figure 3. The electrical characteristics of OTFTs adopted the composite without Tween80: the output (a) and transfer (b) characteristics.

3.2 The composite with Tween80

Capacitors with Al/insulator/Au MIM structure were fabricated to investigate the dielectric properties of the bare PVP and composite insulators, and the measured capacitances as the function of the frequency are shown in figure 4 (a). It is evident that the capacitances of the composite insulator are larger than those of the bare PVP. The calculated dielectric constant of the composite insulator is about 5.8 at 100 kHz, while that of the bare PVP is calculated about 4.0. Therefore, it is expected that OTFT with the composite insulator can operate at relatively lower voltage compared to device with the bare PVP because more charges can be accumulated at the conduction channel with increasing the dielectric constant of a gate insulator at the same voltage. And it is thought that the relatively lower dielectric constant of the composite with Tween80 compared to that of the composite without Tween80 might be attributed from Tween80. The dielectric constant of PVP with Tween80 was calculated about 1.1 from figure 4 (b).



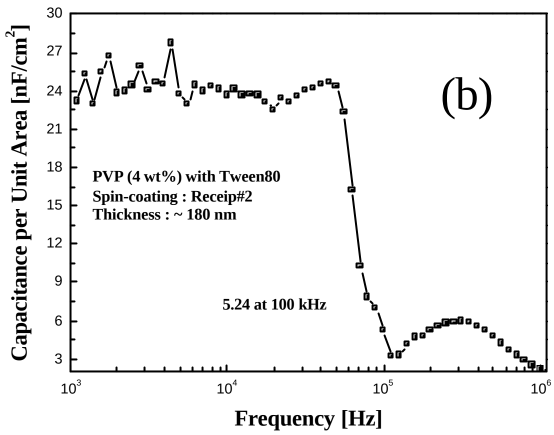
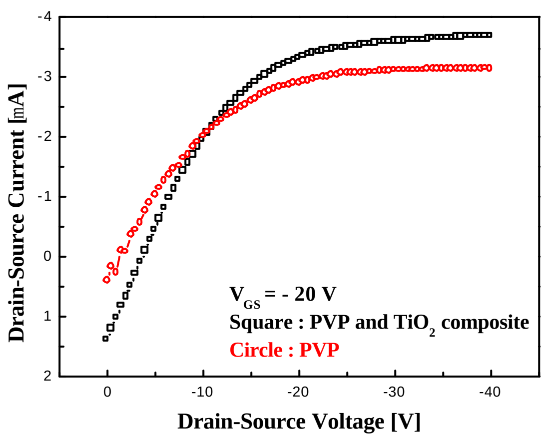


Figure 4. The capacitance-frequency plots according to the insulators.

Typical output and transfer characteristics of OTFTs with different gate insulators are shown in figure 5. For the output characteristic measurements, the drain current (I_D) of the fabricated OTFTs, where the drain-source voltage (V_D) was swept from 0 to - 40 V with the sweep step of -0.5 V at the gate-source voltage (V_G) of -20 V. And for the transfer characteristic measurements, the V_G was swept from 5 to - 40 V with the sweep step of - 0.5 V at the V_D of -20 V, and from the curve of the square root of I_D versus V_G, field effect mobility (μ_{eff}) in the saturation region is estimated using eq. (1):

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

where C_i is the capacitance of the gate insulator per unit area, V_T is the threshold voltage [5]. In accordance with our expectations, it is observed that the composite insulator presents the increased saturation current and contributes to lowering the threshold voltage and enhancing the subthreshold slope of OTFTs due to its high dielectric constant. However, relatively large gate leakage current slightly deteriorates the device performance in the low drain voltage region for the output characteristics and hindered the drain current from further increasing in the high gate voltage region for the transfer characteristics. Though the gate leakage current could be reduced by using Tween80, it is thought that the aggregated TiO_2 clusters still exist and deteriorate the device performance. The important device parameters are listed in Table 1.



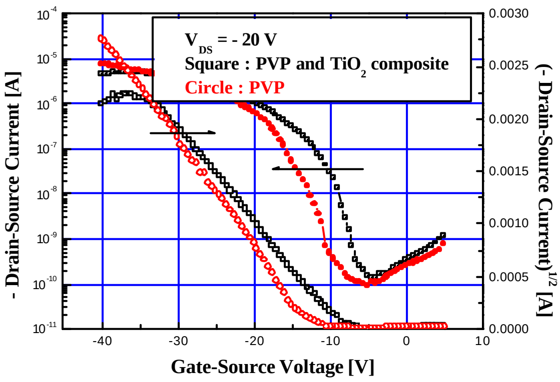


Figure 5. The electrical characteristics of OTFTs: (a) output characteristics and (b) transfer characteristics.

Table 1. The properties of the fabricated OTFTs according to the gate insulators.

	OTFT with the bare PVP	**OTFT with the composite**
V_T	-11.5 V	-8.3 V
Subthreshold Slope	2.8 V/decade	1.5 V/decade
I_{on}/I_{off}	8.5×10^4	3.6×10^4
μ_{eff}	0.32 cm^2/Vs	0.18 cm^2/Vs

According to AFM images, it is revealed that the partially aggregated TiO_2 clusters, which are the main cause of the large gate leakage current, still exist in the composite film in spite of the surface treatment of TiO_2 particles with Tween80 (see figure 4). Currently, our investigations are focused on modifying the process to eliminate the aggregated TiO_2 clusters and prevent the aggregation of TiO_2 particles.

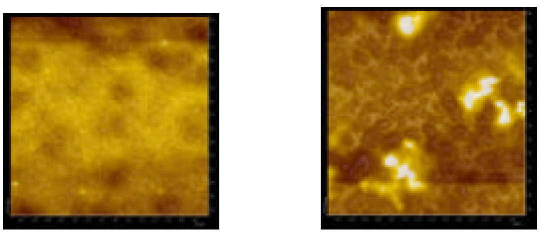


Figure 6. The AFM images of the gate insulators: (a) the bare PVP film and (b) the composite film.

4. Summary

Previously, we have reported that the dielectric constant of polyvinylacetate (PVAc) could be increased with high-k clays, and the performance of OTFTs with PVAc-clay composite as an insulator could be modified with increasing the amount of clays [6]. But it was very difficult to disperse clays (a few microns) uniformly into PVAc solution and it was observed that vertically aligned clays in the composite film even deteriorated device performance. In this paper, we have synthesized the PVP-TiO_2 composite and investigate the performance of OTFTs with the composite insulator. It was observed that TiO_2 particles were generally aggregated in the solution. Therefore, Tween80 was used as a surfactant to treat TiO_2 particle for uniform distribution. As a result, the composite material showed the increased dielectric constant. Thereby, for the OTFT with the composite, V_T was reduced and the subthreshold slope was improved, compared to those of device with the bare PVP. Consequently, it is convinced that the composite can contribute for OTFTs to be feasible for real applications. However, there are still some problems associated with preventing the aggregation of TiO_2 particles during the synthesis process. We make an effort to develop a high-performance composite insulator without such defects and investigations are in progress.

5. Acknowledgements

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6. References

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