

# Capacitance-Voltage Characteristics of MIS Capacitors Using Polymeric Insulators

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## Abstract

In this study, we investigate the capacitance-voltage ( $C-V$ ) characteristics of metal-insulator-semiconductor (MIS) capacitors consisting of pentacene, as an organic semiconductor, and polymeric insulators such as poly(4-vinylphenol) (PVP) or polystyrene (PS) prepared by spin-coating process, to analyze the interfacial characteristics between pentacene and polymeric insulators. Compared with the device with PS, the MIS capacitor with PVP exhibited a pronounced shift in the flat-band voltage according to the bias sweep direction. This hysteretic feature in the  $C-V$  characteristics is thought to be attributed to the trapped charges at the interface between pentacene and PVP owing to the hydrophilicity of PVP. From the experimental results, we can conclude that surface polarity of polymeric insulator has a critical effect on the interfacial properties, thereby affecting the bias stability of organic thin-film transistors.

**Keywords :** Hysteresis, Interface, Polymeric insulators

## 1. Introduction

Recently organic semiconductors have drawn much attention in frontier electronics mainly focused on simplifying processes and their unique properties. There have been many investigations done and accordingly the performance of organic electronics has remarkably advanced toward the promising applications in the fields of large-area displays, solar cells, flexible electronics, and other various sensors [1-3]. *Klauk et al.* have reported about the excellent characteristics of organic thin-film transistors (TFTs) with polymeric insulator, which are comparable or even surpassing those of amorphous silicon TFT [4].

However, organic TFTs with polymeric gate insulators often exhibit a shift in the threshold voltage depending on the sweep direction of the gate-source voltage, making organic TFTs impractical. Yet, it still ensures outstanding

electrical performance. Therefore, research into polymeric insulators for organic TFTs has recently received increasing attention and is rapidly establishing itself as a line of research equally important to organic electronics as the traditional organic TFT research themes that address the organic semiconductor itself, and circuit engineering [5].

In this study, the effects of the surface polarity of polymeric insulator on the capacitance-voltage ( $C-V$ ) characteristics are reported. Different polymeric insulators such as poly(4-vinylphenol) (PVP) and polystyrene (PS), are used to explain the bias-induced instabilities of organic TFTs.

## 2. Experiments

For the measurement of  $C-V$  characteristics, the metal-insulator-semiconductor (MIS) structured capacitors were fabricated as follows. An Al electrode was thermally evaporated to be of 1500 Å in thickness on a glass substrate using the first shadow mask, and then PVP (6 wt% in ethanol) or PS (2 wt% in chloroform) as an insulator was formed by spin-coating method, followed by appropriate curing processes in a vacuum dry oven. For detailed instructions, PVP film was annealed at 60°C for 10 min and consecutively at 110°C for 60 min. Then, PS film was cured at 80°C for 10 min and subsequently at 100°C for 60 min. The thickness of both polymeric gate dielectrics was controlled to be about

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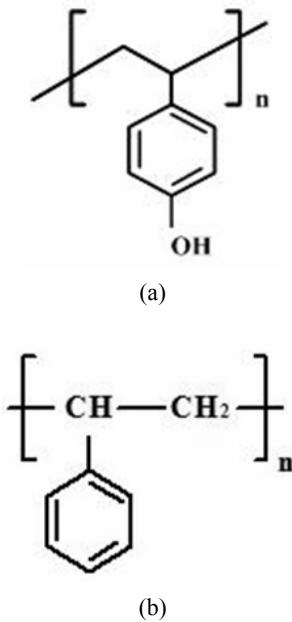
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**Fig. 1.** Chemical structure of the polymeric insulators used in these experiments: (a) PVP and (b) PS.

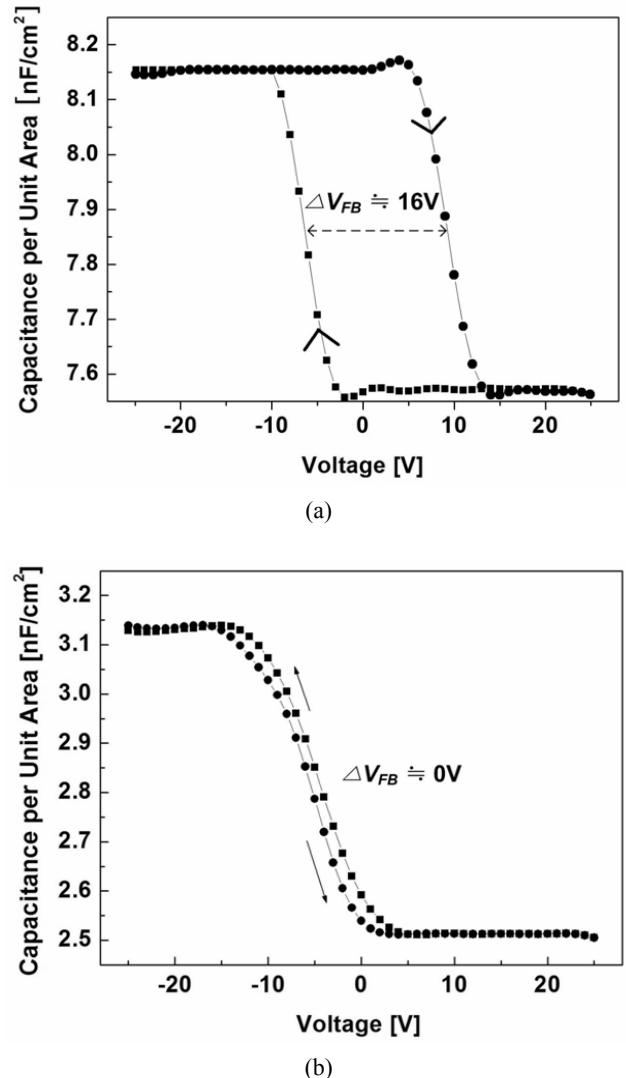
4000 Å. Pentacene (Tokyo Kasei Organic Chemicals, unpurified), which was used as an organic semiconductor, was thermally evaporated through the second mask onto an insulator-coated substrate at a rate of 1.0 Å/s and its thickness was about 600 Å. Then, a 400- Å-thick Au layer was thermally evaporated through the third mask. All the deposition processes were carried out at a base pressure of about  $1.6 \times 10^{-6}$  Torr.

The *C-V* characteristics were performed using HP4192 impedance analyzer in the dark and shielded condition. For the surface characterization, the polymer surface roughness was investigated using atomic force microscopy (AFM) and the contact angle measurements were performed because the wettability seemed to be a suitable indication for comparing the surface polarities of PVP and PS. The molecular structures of polymeric insulators are shown in Fig. 1.

### 3. Results and discussion

The *C-V* characteristics were measured using the fabricated MIS capacitors by applying a small ac amplitude of 10 mV and a frequency signal of 100 kHz. For the device with PVP, the flat-band voltage ( $V_{FB}$ ) in the *C-V* curve exhibited a critical shift of about 16 V depending on the voltage sweep direction and the hysteretic behaviour is quite apparent, as shown in Fig. 2 (a). It is well known that a shift

in  $V_{FB}$  occurs because of the effect of the trapped charges at the interface between insulators and semiconductors [6], and a hysteresis in the *C-V* curve may be due to both traps filled at the polar insulator surface and/or electrical properties of the insulator bulk. But we did not observe any dependency of the hysteretic feature on the thickness of PVP layer in the previous study [7]. On the other hand, a hysteresis in the *C-V* curve was not observed for the MIS capacitor with PS, as shown in Fig. 2 (b), even though the slope is shelving, which may indicate more interfacial trap density than for the device with PVP. This result can be explained by lower dielectric property ( $\epsilon_r \sim 2$ ) and fast de-trap process of PS compared to PVP ( $\epsilon_r \sim 4.1$ ). These



**Fig. 2.** Measured *C-V* curves of MIS capacitors with (a) PVP and (b) PS, respectively.

results confirms that the residual charges, which were trapped at the interface between PVP and pentacene after a prior measurement, could be the main cause of the pronounced shift in  $V_{FB}$  for the MIS capacitor with PVP. The trapped charges at the interface between polymeric gate insulator and pentacene was estimated by the shift of  $V_{FB}$  using equation (1),

$$\Delta V_{FB} = \frac{\Delta Q_{int}}{C_i} \quad (1)$$

where  $Q_{int}$  is the charge density at the interface between polymeric insulator and organic semiconductor [8]. As  $C_i$  is a constant value according to polymeric insulator ( $\sim 12.49$  nF/cm $^2$  for PVP), it can be assumed that  $Q_{int}$  is the only component to cause a shift in  $V_{FB}$ . From  $\Delta V_{FB}$  obtained by the C-V result, the interfacial trapped charge concentration for MIS capacitor with PVP is calculated to be about  $1.2 \times 10^{12}$  cm $^{-2}$ .

In order to compare the surface polarities of PVP and PS, contact angles were measured by using distilled water. Among the techniques for measuring contact angles, the direct measurement of contact angles from sessile drops was used and the angle was obtained by aligning a tangent with the drop profile at the point of contact with the solid surface, in this study. The photographs of sessile drops according to the different polymeric insulators are shown in Figs. 3 (a) and (b). It is observed that the contact angle for

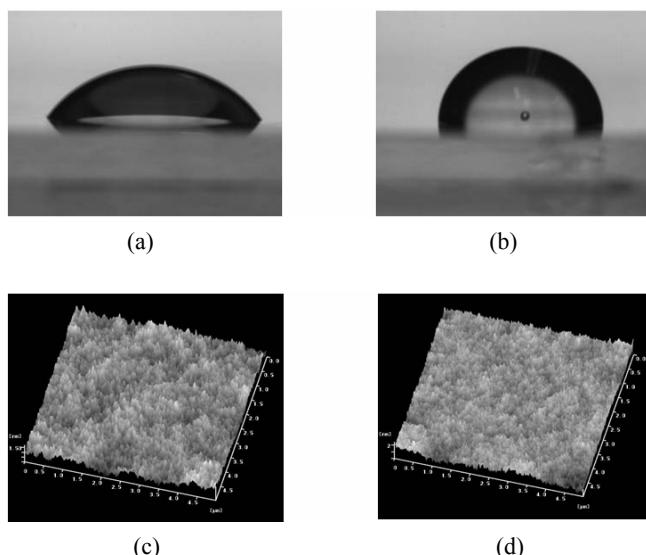
PS ( $\theta_0 = 88^\circ$ ) is much larger than that for PVP ( $\theta_0 = 56^\circ$ ), which means that the solid surface of PVP is more wettable than that of PS. The surface free energy of the polymeric insulators ( $\gamma_p$ ) could be calculated by using equation (2), where  $\theta_0$  is the contact angle at the equilibrium and  $\gamma_w$  is the water surface free energy (73 mJ/m $^2$ ) [9].

$$\gamma_p = \frac{\gamma_w}{4} (1 + \cos \theta_0)^2 \quad (2)$$

The  $\gamma_p$  of PVP is estimated to be about 44.3 mJ/m $^2$  and 19.5 mJ/m $^2$  for PS. It is well known that the wettability is a complex phenomenon related to both physical and chemical effects. Therefore, the surface topography of the insulator was also investigated by atomic force microscopy (AFM) to elucidate the influence of the surface roughness of the insulator on the contact angles. But we did not observe any significant difference in the surface roughness (see Figs. 3 (c) and (d)). The root-mean-square roughness values are about 21 Å and 23 Å for PVP and PS, respectively, which indicate that the surface roughness of polymeric insulator had little effect on the aforementioned critical difference in the contact angles according to polymeric insulators. From this study, we could find out that the wettability and  $\gamma_p$  are not affected by the surface roughness but the polarity of polymeric insulator. Accordingly, it is believed that the hydrophilicity of PVP induced trapped charges at the interface between PVP and pentacene, thereby resulting in the pronounced shift in  $V_{FB}$  for the MIS capacitor with PVP. Thus, we can conclude that the trapped charges at the interface between the PVP and pentacene layers are strongly related with the hydrophilic surface of the PVP layer owing to hydroxyl groups and are the main cause of the OTFT characteristic degradations and instabilities. Our conclusion is in pretty good agreement with the results of the previous report, where the hydroxyl groups in organic dielectrics are considered to be the origin of the hysteresis because hydroxyl groups act as trap sites [10].

#### 4. Conclusions

Polymeric gate insulators often cause a large hysteresis in the transfer characteristics of organic TFTs and a shift in threshold voltage. These unstable characteristics deteriorate the reliability of organic TFTs and, thereby, hinder them from being adopted for future electronics. Recently, many



**Fig. 3.** The side views of a sessile drop on (a) PVP and (b) PS, and the AFM images of (c) PVP and (d) PS, respectively.

researches focus on the problems associated with the electrical stability of organic TFTs with polymeric insulators. In this regard, we have successfully investigated the origin of hysteretic feature in the  $C$ - $V$  characteristics, which can explain the bias-induced instabilities of organic TFTs with polymeric gate insulators. For future works, we will conduct investigations to understand the trap/de-trap mechanism of polymeric gate insulators, systematic studies on the effects of frequency and delay time on the  $C$ - $V$  characteristics are in progress.

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