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Effect of morphological and physicochemical properties of dielectric-organic semiconductor interfaces on photoresponse of organic phototransistors

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ABSTRACT

We investigate the effect of the interfacial properties between a polymer dielectric and an organic semiconductor (OS) layers on the photoresponse properties of an organic phototransistor (OPT). Three different polymer materials having different interfacial properties, poly(methyl methacrylate), amorphous fluoropolymer CYTOP, and poly(4-vinylphenol), were used as gate insulators for the OPTs in a bottom-gate top-contact configuration. For the trapping and detrapping processes of minority carriers, the physicochemical nature and the density of functional group of the dielectric material were found to more dominantly govern the photoresponsive properties of the OPT than the morphological effect. Our molecular level description provides a useful guideline to select a proper dielectric material, being used as a gate insulator, for optical applications of the OPTs such as optical memory devices and optical sensors.

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1. Introduction

Organic phototransistors (OPTs) have attracted enormous attention due to their potential for low-cost, light-weight, flexible, and multifunctional optical transducers [1]. Moreover, they exhibit higher photosensitivity and signal-to-noise ratio than two-terminal organic photodiodes owing to the signal amplification capability by the field effect [2]. Since the transport of photo-generated charges primarily takes place within a first few monolayers of an organic semiconductor (OS) film at the interface with a gate insulator, the interfacial properties of the dielectric layer play a critical role on the photoresponse of the OPT [3]. In particular, the interfacial characteristics determine the amount of trapped charges at the dielectric/OS interface under light illumination so that they should be properly tailored for specific applications of the OPTs ranging from optical memory to optical sensing devices [4]. Among a number of previous studies on the OPTs with inorganic insulators such as silicon dioxide (SiO₂) and tantalum pentoxide [1–2,4–8], the modification of the insulator surface by the oxygen content [5] or the treatment with a self-assembling monolayer [6] was found to significantly change the trap density at the interface. However, the effect of the interfacial properties of a polymer insulator, allowing the mechanical flexibility and low-cost fabrication process, on the photoresponse of a polymer insulator-based OPT (p-OPT) has not been fully addressed

from the viewpoint of the charge trapping mechanisms involved in organic field-effect transistors (OFETs) [9]. Therefore, it is very important to obtain a rather complete picture of the relationship between the interfacial characteristics of the polymer insulator and the charge carrier dynamics of the p-OPTs under light illumination.

In this work, we investigated the dependence of the photoresponse of the p-OPT on the morphological and the physicochemical properties at the dielectric/OS interface including the surface roughness, the surface energy, and the functional group of the polymer insulator. In examining the relationships between the photoresponse and the interfacial characteristics, we fabricated the p-OPTs with three different types of polymer gate insulators of poly(methyl methacrylate) (PMMA, the dielectric constant $k = 3.5$), amorphous fluoropolymer (CYTOP, $k = 2.1$), and poly(4-vinylphenol) (PVP, $k = 4.5$), respectively. The schematic diagram of our p-OPT is shown in Fig. 1(a). The three polymer gate insulators we used have been widely employed in organic electronic devices due to their excellent dielectric properties [10,11]. As seen from the chemical structures in Fig. 1(b)–(d), PMMA has the ester group acting as a trapping site for holes [12,13] and PVP has the hydroxyl group which dominantly traps the electrons by the electrostatic interaction [14,15]. In contrast, CYTOP has no specific chemical group for electrical traps [16].

2. Experimental

Each p-OPT was fabricated on a glass substrate with a 165 nm-thick indium-tin-oxide layer serving as a gate electrode. All the

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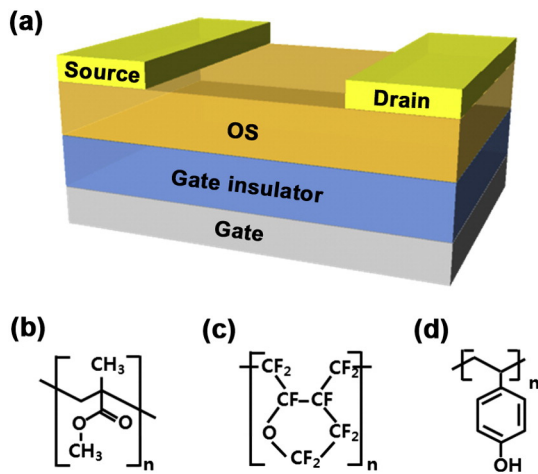


Fig. 1. (a) Schematic diagram of the p-OPT configuration. Chemical structures of (b) PMMA, (c) CYTOP, and (d) PVP.

substrates were cleaned with acetone, isopropyl alcohol, methanol, and deionized water in sequence. For gate insulators, PMMA ($M_w = 996,000$ g/mol, Sigma-Aldrich Korea) dissolved in anisole at the concentration of 8 wt.%, CYTOP (CTL-809 M, Asahi Glass) at 9 wt.% as received, and PVP ($M_w = 25,000$ g/mol, Sigma-Aldrich Korea) mixed with poly(melamine-co-formaldehyde) (PMF) (100 wt.% of PVP) in propylene glycol methyl ether acetate at 10 wt.% were used. In preparing a gate insulator of 500 nm thick in all cases, each solution of PMMA, CYTOP, or PVP was spin-coated at 3000 rpm for 60 s, 5000 rpm for 60 s, or 3000 rpm for 30 s, respectively. The PMMA and CYTOP films were annealed at 110 °C for 2 h and 100 °C for 1 h, respectively, under ambient condition to remove the residual solvent. For the PVP film, it was first soft baked at 100 °C for 30 min and then thermally treated at 200 °C for 50 min for crosslinking. As the OS layer, a 50 nm-thick pentacene layer was evaporated on the top of each dielectric layer at the rate of 0.5 Å/s under the pressure of 1×10^{-5} Torr. Note that the mobility of pentacene (about $1 \text{ cm}^2/\text{Vs}$) is comparable to that of hydrogenated amorphous silicon [17,18]. For the source and drain electrodes, 50 nm-thick gold layers were thermally deposited through a shadow mask at the rate of 1.0 Å/s under 1×10^{-5} Torr. The channel length and the channel width of the p-OPT were 150 μm and 1 mm, respectively.

Since PMF is known to change the number density of the hydroxyl groups in the PVP film [19], the PVP layers of 60 nm thick at two different concentrations of PMF:PVP (0:1 and 1:1 in weight) were also prepared on thermally grown SiO₂ layers of 300 nm thick for gate insulators. In this case, heavily doped p-type silicon wafers were used as gate electrodes.

The electrical characterization of each p-OPT was carried out using a semiconductor parameter analyzer (HP4155A, Hewlett-Packard Co.) under ambient condition. The photoresponse of the p-OPT was measured using a ultraviolet (UV) light source (GL-155, UVSMT) with the intensity of 9 mW/cm² at the wavelength of 365 nm. Note that pentacene-based OPTs have been widely used for detection of UV light [20–22]. The UV light was illuminated from the top side where the source and drain electrodes were located. The surface morphologies of each dielectric layer and the corresponding pentacene film were determined using an atomic force microscopy (AFM) (XE-100, PSIA) and the contact angle of water on the dielectric layer was measured with an optical microscope (Optiphot-Pol, Nikon). Fourier transform infrared spectroscopy (FT-IR) (Tensor 27, Bruker) was used for monitoring the amount of hydroxyl group in the PVP film with or without PMF.

3. Results and discussion

3.1. Interfacial characteristics of PMMA, CYTOP, and PVP layers

In Fig. 2(a)–(c), the morphological and physicochemical properties of three different dielectric/OS layers were characterized using the AFM images and the corresponding morphological profiles of the PMMA, CYTOP, and PVP layers, respectively. From the AFM images, the values of the root-mean-square roughness for PMMA, CYTOP, and PVP were measured to be 0.17, 0.32, and 0.24 nm, respectively. Considering that the surface roughness in the three cases is only a few tenths of a nanometer, being consistent with the previous studies [13,23], the morphological effect on the photoresponse of the p-OPT would be quite similar. From the physicochemical viewpoint, the disparity of the surface energy between the dielectric polymer and the OS will strongly influence the morphology of film grown on the polymer layer [15]. From the contact angles of 66° for PMMA, 107° for CYTOP, and 68° for PVP as shown in the insets of Fig. 2(a)–(c), the surface energies of PVP and PMMA are similar and rather hydrophilic but that of CYTOP is relatively hydrophobic [24,25]. Fig. 2(d)–(f) show the AFM images of the pentacene films on the three dielectric layers. It should be noted that the pentacene film of 10 nm thick was prepared since a first few monolayers on the dielectric surface would contribute significantly to the charge transport as observed in organic field-effect transistors [26]. The values of the average grain size of pentacene on the PMMA, CYTOP, and PVP layers were 0.92, 0.30, and 0.72 μm , respectively. Note that the average grain size of pentacene on the CYTOP layer was smaller than those in the other two cases. This is most likely due to the large difference in the surface energy between the pentacene film and the CYTOP surface [15,27].

3.2. Photoresponse behaviors of p-OPTs with different polymer dielectrics

To understand the influence of the interfacial properties on the photoresponse, the transfer curves of the p-OPTs with three dielectric layers of PMMA, CYTOP, and PVP before and after the light illumination for 1 min were investigated. As shown in Fig. 3(a)–(c), before the light illumination, the transfer curve in the PVP case exhibited a positive onset voltage (V_{on}) in contrast to the PMMA and the CYTOP cases. This is attributed to the building up of negative charges captured by hydroxyl groups at the PVP/OS interface in the presence of oxygen and moisture [28,29]. Note that, in Fig. 3(b), the p-OPT with CYTOP showed a lower current level than those with PMMA and PVP. This is attributed to a relatively high density of grain boundaries of the pentacene film on the CYTOP layer as clearly seen in Fig. 2(e). In Fig. 3(a) and (b), it was observed that, for the PMMA and CYTOP cases, the magnitudes of the threshold voltage shift (ΔV_{th}) before and after the light illumination were 1.04 V and 0.47 V in the negative direction, respectively. On the other hand, the PVP case showed a significantly large value of 10.55 V toward the positive direction as seen in Fig. 3(c). In fact, ΔV_{th} toward the positive (or negative) direction under light illumination is proportional to the amount of trapped electrons (or holes) at the dielectric/OS interface [4,9,30]. For the PVP case, a large amount of electrons were trapped at the interface under light illumination whereas for the PMMA and the CYTOP cases, the values of ΔV_{th} were so small that the amount of trapped charges could be ignored. In addition, the photosensitivity (or equivalently, the ratio of the photocurrent to the dark current) in the PVP case showed a much higher value of 6.26×10^4 than the PMMA (4.00×10^{-1}) and the CYTOP (2.24) cases at the gate voltage (V_G) equal to V_{on} . It should be noted that the photosensitivity of each OPT was extracted from the transfer curves before and after the light illumination.

The dynamic photoresponse of each of the three p-OPTs was described in terms of the drain current (I_D) as a function of time under light illumination for 5 s in every 20 s period. Fig. 4 shows the dynamic photoresponse behaviors of the p-OPTs at $V_G = V_{\text{on}}$. In this case, the

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