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Investigation of Top-Contact Organic Field Effect Transistors by the Treatment Using the VDP Process on Dielectric

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Abstract : 이 논문에서는 게이트 절연막 위에 vapor deposition polymerization(VDP)방법을 사용하여 성막한 유기 점착층을 진공 열증착하여 유기 박막 트렌지스터(OTFTs)소자를 제작할 수 있음을 증명하였다. 우리가 제작한 Staggered-inverted top-contact 구조를 사용한 유기 박막 트랜지스터는 전기적 output 특성이 포화 영역안에서는 포화곡선을, triode 영역에서는 비선형적인 subthreshold를 확실히 볼 수 있음을 발견했다. 0.2㎛ 두께를 가진 게이트 절연막위에 유기 점착층을 사용한 OTFTs의 장 효과 정공의 이동도와 문턱전압, 그리고 점멸비는 각각, 약 0.4 cm2/Vs, -0.8V, 106 이 측정되었다. 게이트 절연막의 점착층으로써 폴리이미드의 성막을 위해, 스핀코팅 방법 대신 VDP방법을 도입하였다. 폴리이미드 고분자막은 2,2bis(3,4-dicarboxyphenyl)hexafluoropropane dianhydride(6FDA)와 4,4′-oxydianiline(ODA)을 고진공에서 동시에 열 증착 시킨 후, 그리고 150℃에서 1시간, 다시 200℃에서 1시간 열처리하여 고분자화된 막을 형성하였다. 그리고 점착층이 OTFTs의 전기적 특성에 주는 영향을 설명하기 위해 비교 연구하였다.

Keywords : organic thin-film transistor (OTFT), vapor deposition polymerization (VDP), adhesion layer.

1. Introduction

The electrical performances of organic thin-film transistors (OTFTs) have been improved for the last decade. Inorganic materials have been generally used as gate insulating layer, such as silicon oxide that has properties of a low electrical conductivity and a high breakdown field.1)~4) However, surfaces of inorganic insulating layers that have hydrophilic property, may affect most of organic semiconductor materials. It caused the field effect mobility and drain current to decrease due to the mismatching of insulator

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and semiconductor layer. Since the interface between the inorganic insulator and organic semiconductor layer was formed by two different properties material, such as hydrophilic and hydrophobic, it is of poor quality and may cause a defect and disorder of semiconductor layer. To avoid this problem, various materials are studied in previous report of OTFTs, such as hexamethyldisilazane (HMDS). Other researcher found that the characteristics of pentacene TFTs can be improved by using a self-organizing material like octadecyltrichlorosilane (OTS) between the SiN_X gate dielectric and the pentacene active layer.[4] But these materials fabricated film by wet process methods, such as spin-casting, dipping and self-assembly. In this works, we have investigated OTFTs with the fabricating polyimide adhesion layer formed by vapor deposition polymerizatioin (VDP) of 6FDA and ODA. VDP is mainly used for polyimide, the membrane or the conducting polymers is introduced to form the gate dielectric film.[5] This method is appropriate to mass production in various end-user applications, for example, flat panel displays, because it has the advantages of shadow mask patterning, no need of catalysts to polymerize, ease of preparation conditions, ease of making polymers with another chemical structure and *in-situ* dry process with flexible low-cost large area displays. Since it is also dry-processing method, it prevent from contamination of wet processing.

2. Experimental

OTFTs were fabricated to demonstrate that thermally evaporated polyimide can be used as an adhesion layer. All our devices were fabricated on glass substrates using a staggered-inverted structure as shown in Figure 1. In this structure, two different voltage sources were used, one across the dielectric layer (V_G), which generates the charges (charge injection into the channel occurring), and the second one (V_D) along the active layer (channel) to drive them from source to drain. Here, 100 nm-thick Indium-tin-oxide (ITO) as a gate electrode was sputtered. Second, 0.2 ㎛-thickness SiN_X as gate insulator was deposited by plasma enhanced chemical vapor deposition (PECVD) in the LG-Phillips LCD company. To improve the quality of the organic/dielectric surface, polyimide film using as adhesion layer on the SiN_X was co-deposited by high-vacuum thermal-evaporation from 6FDA and ODA at 5×10^{-7} Torr with deposition rate of 4 Å/s, and cured at 150℃ for 1 hr followed by 200℃ for 1 hr in the vacuum oven at 5×10^{-3} Torr. For adjusting balance of monomer's deposition rate to one to one, monomers kept up preheating for 2 hr before co-evaporation. Polyimides are polymers made from the polymerization of an acid dianhydride (6FDA) and a diamine (ODA). They are characterized by the presence of the imide functionality, a cyclic tertiary amine bound by two carbonyl groups, and either an aliphatic or aromatic group in the main chain. The curing process and the associated link will affect mechanical, thermal, and electrical properties of polyimide. This heat treatment, known as "curing," usually has been done at temperature range of 150 ~ 300℃. Figure 2 shows the preparation of polyimide via the condensation of carboxylic dianhydride and dianiline; 6FDA-ODA. In result, we showed Fourier transform infrared (FT-IR) analysis profiles of polyimide. Pentacene as active layer was deposited by thermal evaporation at 7×10^{-7} Torr, deposition rate of 0.3 Å/s, and total thickness of 60 nm after material purification by vacuum gradient sublimation. During the deposition of pentacene, the substrates were held at room temperature. To improve the purity of the semiconductor material, moderate heating and low deposition rates have been used to improve the performance of organic thin-film transistor. The devices were completed by thermal deposition of gold (Au) to form source and drain contacts through a shadow mask.

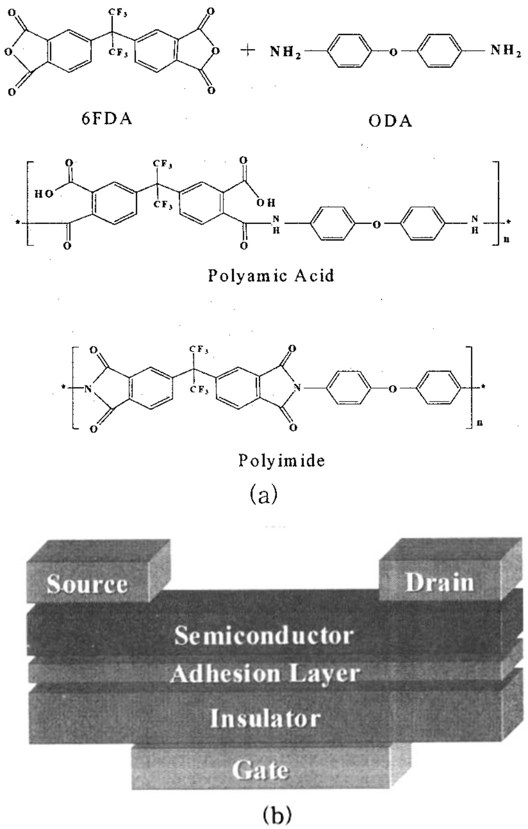


Fig. 1. The simplified mechanism of polyimidazation via the condensation of 6FDA and ODA (a) and schemes of OTFTs (b).

3. Results and Discussion

In order to characterize the organic polymeric film layers, 6FDA and ODA were deposited on a silicon wafer, and analyzed by FT-IR. Figure 2 shows the spectrum of a deposited layer annealed at 150℃ for 1 hr followed by 200℃ for 1 hr after co-deposition. It shows that the first step reaction took place immediately after the deposition in vacuum without annealing and the deposited monomers converted to polyamic acid. But it had not polymerized to polyimide. After annealed at 150℃ for 1 hr followed by 200℃ for 1 hr, the C-N imide peak appeared at 1380 $\text{cm}^{-1}$and the amide acid peak disappeared at 1660 cm^{-1} and there was difference in the intensity of all the imide peaks as compared with the annealing at 150℃ for 1 hr. Besides either peak at 1210 cm^{-1}, C-N stretches at 1300˜1325 cm^{-1}, C=C stretching mode of the ODA moiety (excess ODA) at 1500 cm^{-1} and anhydride stretches at 1780 ˜ 1850 cm^{-1} appeared.[7,10] The electrical characteristics of the individual OTFT using 6FDA-ODA polymeric material as adhesion layer are shown in Figure 3 and 4 for enhancement mode operation. It shows the output I_D-$V_D$characteristics for an OTFT fabricated as described above with channel length of 50 μm and width of 1.25 mm (W/L = 25), and 50 μm and 2.5 mm (W/L=50), respectively. The field effect mobility of the FET is generally determined in the region where the drain current (I_D) saturates i.e.; FET could be evaluated simply from $V_D > V_G - V_T$ where V_G is the gate voltage. In this region, the current can be modeled as $I_{Dsat} = (W/2L)\mu_{FET}C_i(V_G-V_T)^2$, where W and L are the channel width and length, and C_i is the

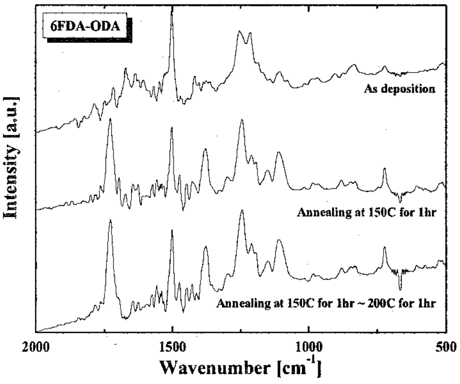
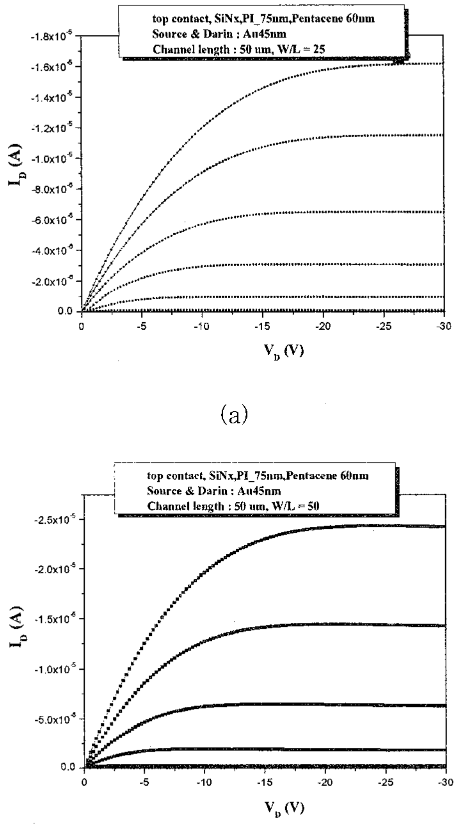


Fig. 2. Fourier transform infrared (FT-IR) spectra of a cured polymeric film

capacitance of the gate dielectric layer.[8] The field effect mobility is therefore estimated from the slope of the square root of the saturation current, $I_D^{1/2}$ as a function of the gate voltage, V_G. As the gate voltage (V_G) and the drain voltage (V_D) increase, drain current (I_D) increased, too.

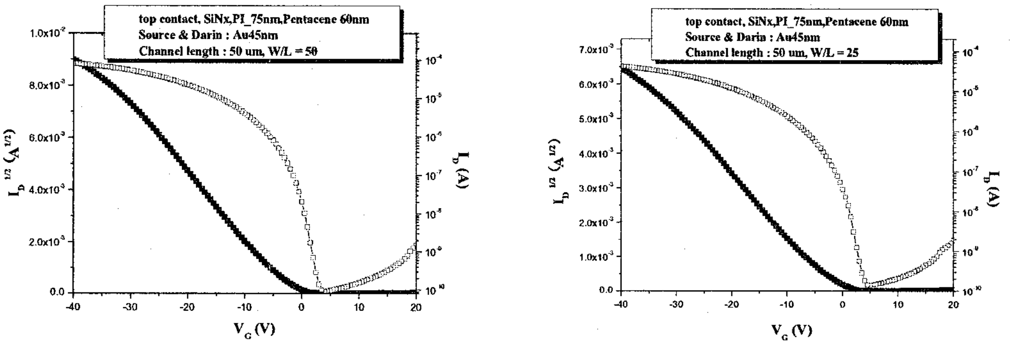


(a)

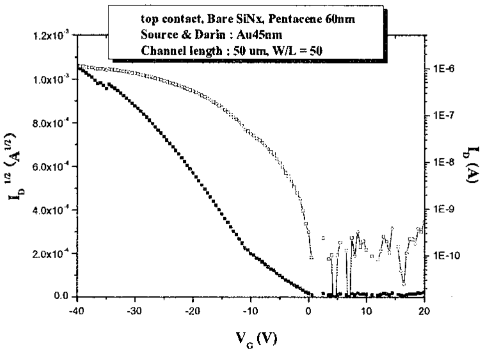
(b)

Fig. 3. Electrical output characteristics of the organic thin film transistors; the ratio of the channel width and length (W/L = 25(a) and 50(b)) of OTFTs.

The drain bias (V_D) was swept from 0 to -30 V and back at gate bias (V_G) of 0 to -30 V at -5 V intervals. Charge carrier mobility of an OTFT using the ratio of W/L = 25 in the saturation region and that of an OTFT using the ratio of W/L = 50 were about 0.08 cm^2/Vs in all. These are typical bias conditions for *p*-type channel operation. We hope that carrier transport in field induced channels in pentacene is dominated by the difficulty of transporting carrier from one molecule to the next because of disorder, defects, and chemical impurities which can form trapping site. We believe the most important factor in obtaining the much better mobility reported here is improved quality of the evaporated pentacene layer. According to increasing gate voltage and drain voltage, drain current increased and saturated. Figure 4 shows the log (I_D)-V_G transfer characteristics for the same devices. We extracted field-effect mobility of OTFTs at V_D = -20V and a threshold voltage of about -2 ˜ -1 V. Also, an on/off current ratio and subthreshold swing were obtained about 10^6 and 1.0 ˜ 1.8 V/decade. Figure 4(c) shows the electrical transfer characteristics without an adhesion layer in the OTFTs and poor electrical performances. Several papers proved that carrier transport in field effective channels of the semiconductor is dominated by the difficulty of moving carriers from one molecule to the next because of residual doping in the semiconductor, fixed charges, dipoles at the dielectric interface (impurities) and molecular ordering of semiconductors at induced channel, especially.[3,8~10] We have found that pentacene molecular is more strong densely packed crystals and well films deposited by evaporation onto an adhesion layer than on to a bare SiNx dielectric. Some researches investigated that the grown crystalline of pentacene is correlated with the surface energy for dielectrics.[11,12] especially polymeric dielectric. The surface energies of the silicon based dielectrics and PI on dielectric layer are listed in Table 1. In case of SiNx, the PI deposited on dielectric shows the decrease of surface energy and polarity compared with the other one. And We demonstrate that the contact angle of the PI deposited on dielectric is higher than bare



(a) W/L = 25 (b) W/L = 50



(c) W/L = 50 (bare SiNx)

Fig. 4. Electrical transfer characteristics of the organic thin film transistors the ratio of the channel width and length (W/L = 25(a), 50(b) and bare SiNx (50)) of OTFTs.

Table 1. Surface Energy of Gate Dielectrics and PI on Bare Insulator Substrates

Gate dielectric	Contact angle [°]		Polar	Dispersion	Surface tension
	Water	Diiodo methane	[mjm^{-2}][a]	[mjm^{-2}][a]	[mjm^{-2}][a]
SiNx	66.00	26.00	9.04	40.55	49.59
SiNx/PI	79.00	31.00	3.56	40.95	44.51

SiNx in figure 4, In case of SiNx, PI deposited on silicon based dielectric and bare silicon based dielectric, however, show the quite similar surface characteristics. In generally, since the surface of inorganic insulating layers has a hydrophilic property which means a higher polarity and surface energy, it may disturb the growth of pentacene with highly ordered.[13] Besides, OTFT involves injecting charge from the electrodes into the organic semiconductor. Because of the difference of the work function of electrodes and semiconductor, contacts are occurred the Schottky barrier and the contact resistance.[9] Effect of the contact resistance is visible at low drain voltage and is relatively more important as the treatment of the dielectric surface. The large grain size requires substantial surface mobility for the depositing pentacene molecules. A large grain size reduce the grain boundaries, in which carrier scattering occur and the carrier movement is disturbed, so the field effect mobility might be enhanced. Since the interface between the organic active material and the gate dielectric material is a critical part of the field-effect device, control of this interface is important. Figure 5. shows the atomic force microscope (AFM) images of 60 nm thickness pentacene deposited onto the adhesion layer. It was found that the 2 ㎛ -thick-crystal grain size on 6FDA-ODA polymeric film, and the depth and width of grain boundaries were narrow and dense. In the result, it improved the pentacene molecular ordering onto polymeric films using as an adhesion layer and the field effect mobility enhanced to transfer of charge. To improve the electrical characteristic of OTFT, we have to reduce thickness of adhesion layer up to a few nanometers.

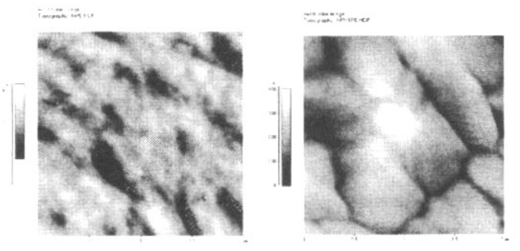


Fig. 5. Surface morphologies of pentacene film on the adhesion layer.

4. Conclusions

We proposed the new dry-processing method of preparing organic adhesion layer on the gate dielectric film in organic field-effect transistors. Vapor deposition polymerization (VDP) is introduced to form the organic adhesion layer. This method is appropriate to mass production in various end-user applications, for example, flat panel displays, because it has the advantages of shadow mask patterning and *in-situ* dry process with flexible low-cost large area displays. Proposed method can be applied to in-*situ* solution-free process to fabricate organic thin-film transistors during overall fabricating steps.

Acknowledgment

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