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Preparation of Aluminum Oxide Layer for Gate Insulator Application in Organic Thin-Film Transistors

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The characteristics of organic thin-films transistors (OTFTs) with the anodized aluminum oxide insulator have been investigated. The OTFT with the barrier-type aluminum oxide insulator exhibited the mobility of 0.09 cm^2/Vs, the subthreshold slope of 1.3 V/decade, the threshold voltage of −2.2 V, and the on/off ratio of 7.7×10^4, which are superior to those for the device with the porous-type insulator. The smooth surface of the barrier-type film is found to contribute to a long range hopping of charge carriers in the conducting channel by decreasing the activation energy for the conduction of charge carriers.

Keywords Anodizing aluminum oxide; gate insulator; organic thin-film transistor

Introduction

Organic thin-film transistors (OTFTs) are expected as a switching element for flexible displays, where aluminum gate electrode is usually adopted to reduce the delay time owing to its low electrical resistivity [1]. Anodized aluminum oxide layer is considered as a good insulator because it is inert to various chemical solvents and also has a high dielectric constant enabling a low-voltage operation of OTFTs. Such an anodized aluminum oxide film generally has two types of morphology, i.e., barrier-type and porous-type films, obtained by varying the pH condition of electrolyte. For example, barrier-type films in the pH range of 5 to 7 are utilized as electrolytic capacitors due to thin, compact, and non-porous structure, while porous-type films are applied to nano-templates due to its thick and porous structure [2].

In this work, we fabricated two different types of anodized aluminum oxide films with varying the pH values of electrolyte. The OTFT with the barrier-type

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aluminum oxide insulator exhibited the mobility of about 0.09 cm^2/Vs, the subthreshold slope of 1.3 V/decade, and the on/off ratio of 7.7×10^4, which are superior to those for the device with the porous-type insulator. We found that such characteristic improvements were essentially attributed to a reduction in the activation energy for charge conduction for the OTFT with the barrier-type aluminum oxide insulator.

Experimental

A 300-nm thick aluminum film was thermally evaporated for a gate electrode of our OTFT. In order to grow an anodizing aluminum oxide on the Al gate electrode, an aluminum electrode was connected to the anode of anodizing bath and the platinum mesh connected to the cathode. The electrolyte was a mixture of 0.01 mol^{-1} citric acid monohydrate ($C_6H_8O_7 \cdot H_2O$) in high purity de-ionized water [3]. An electrolyte of pH 4.2 was used to grow porous-type films and an electrolyte of pH 6.3 for barrier-type films. Anodizing was performed at constant current 1 mA/cm^2 and anodizing voltage was increased to 110 V. Anodized aluminum oxide films were baked at 180°C for 1 h in a vacuum condition. The anodized Al_2O_3 films in our study exhibited rough surfaces, which might deteriorate the growth of the subsequent pentacene film. A 60-nm thick pentacene active layer was thermally evaporated at a deposition rate of 1.0 Å/s. Finally, source and drain electrodes were thermally deposited to the thickness of 40 nm. The fabricated OTFTs have a channel length (L) and width (W) of 40 and 200 µm, respectively. All devices were measured in the dark.

Results and Discussion

The atomic force microscopy (AFM) images of anodized aluminum oxide films are shown in Figure 1. It is observed that the surface of the barrier-type film grown at pH 6.3 is smoother than that for the porous-like film grown at pH 4.2. The root mean square (rms) roughness of the porous-like film was about 20 nm, while the barrier-type film exhibited the rms roughness of 10 nm. The capacitance versus voltage characteristics are shown in Figure 1(c), which was obtained at an operating frequency of 10 kHz. The measured capacitance values are 53 and 55 nF/cm^2 for the porous-like and the barrier-type Al_2O_3 films, respectively. Considering the relative dielectric constant of Al_2O_3 to be about 9, the thicknesses of two different types of Al_2O_3 films are estimated to be about 150 and 145 nm, respectively [4].

Figure 2(a) shows the transfer characteristics measured at a drain voltage (V_D) of −40 V, where the gate voltage (V_G) was swept from 5 to −40 V, of the fabricated OTFTs. The field-effect mobility in the saturation regime is extracted using Eq. (1):

$$I_D = \frac{W}{2L}\mu C_i (V_G - V_T)^2 \tag{1}$$

where I_D is the drain current, C_i is the capacitance per unit area of the insulator layer, and V_T is the threshold voltage [5]. The insets in Figure 2(a) show that the pentacene grain size on the rough porous-like Al_2O_3 gate insulator is smaller than that on the barrier-type Al_2O_3 gate insulator. The threshold voltage was extracted from the curve of the square root of drain current versus gate voltage by tangent at a zero drain current and the subthreshold slope (S), $S = [\partial V_G/\partial(log I_D)]$), is the change in gate bias required for a one-decade change with varying drain current. The on/off

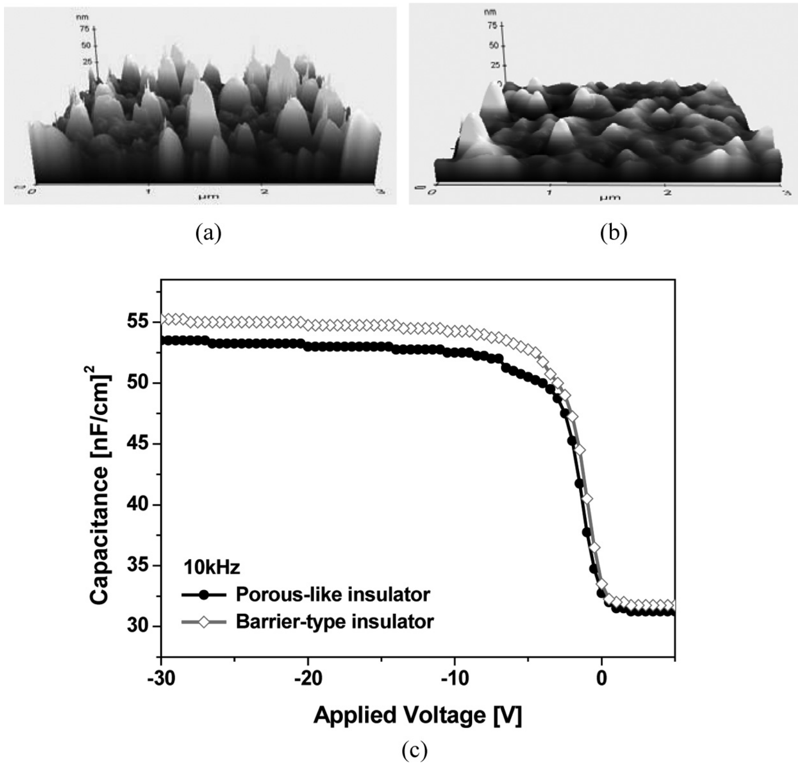

Figure 1. AFM images of two different types Al_2O_3 films; (a) porous-like film and (b) barrier-type film. (c) The capacitance versus voltage characteristics.

ratio of about 7.7×10^4 was obtained for the OTFT with the barrier-type insulator and 1.3×10^5 for the device with the porous-like insulator. The subthreshold slope, field-effect mobility, and threshold voltage for the OTFT with the barrier-type insulator are about 1.3 V/decade, 0.09 cm^2/Vs, and −2.2 V, while those for the device with the porous-like insulator are 1.5 V/decade, 0.03 cm^2/Vs, and −5.1 V.

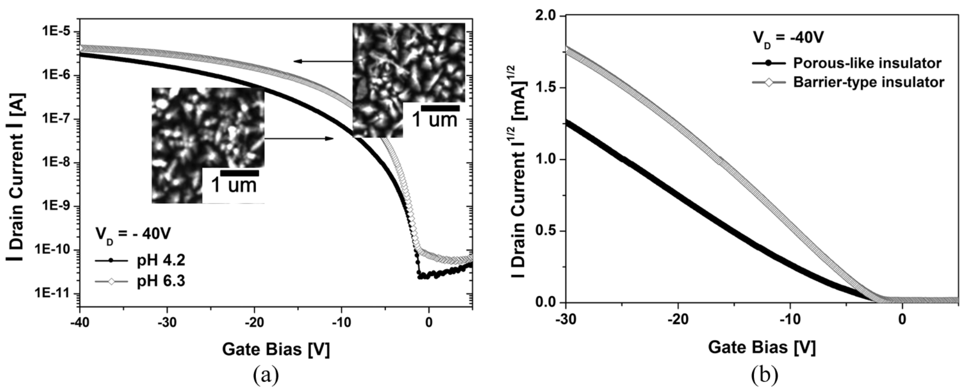

Figure 2. The transfer characteristics of the fabricated OTFTs @ $V_D = -40$ V according to the condition of electrolyte; (a) log-scale and (b) square-root curves.

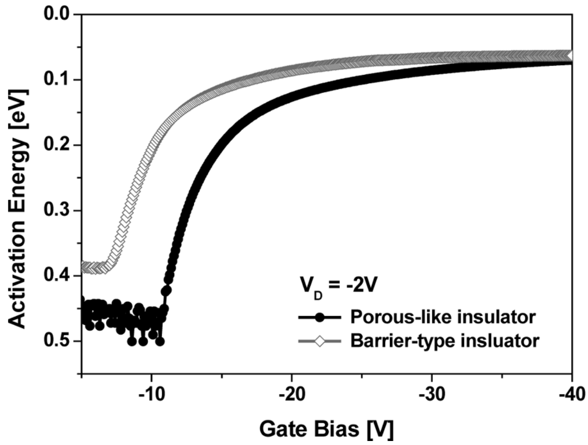

Figure 3. The activation energy curves related with charge carrier conduction.

The activation energy (E_a) related charge carrier conduction is shown in Figure 3. The activation energy, which was derived from the field-effect conductivity and plotted as a function of the gate bias, is extracted using Eq. (2):

$$E_a = -kT \cdot ln\left(\frac{\sigma(V_G)}{A}\right) \tag{2}$$

where $\sigma\ (V_G)$ is the conductivity, $\sigma(V_G) = [(L \cdot I_D)/(W \cdot V_D)]$, k is the Boltzmann's constant of 8.62×10^{-5} eV/K, and kT is room temperature value of 0.0259 eV. A is the constant obtained from the slope of the linear fit in Arrhenius plots [6]. The low drain voltage of -2 V was applied for the measurement in order to determine the important parameters of conductivity and activation energy in the linear regime [7]. Upon a negative bias applied to gate electrode, the charge carriers flow in the organic semiconductor close to the gate insulator. In this case, charge flow can be directly affected by the interface characteristics because pentacene was directly evaporated on the rough aluminum oxide films. Regarding the activation energy, which is required for charge carriers to hop between the energy states in the conduction channel, the smooth roughness would be favorable for a long range hopping [8]. Indeed, the OTFT with the porous-like insulator exhibited larger activation energy than that for the device with the barrier-type insulator. This result demonstrates that the characteristic improvement for the device with the barrier-type structure can be essentially attributed to low activation energy for carrier transport in the conducting channel.

The time-dependant degradation in the drain current of OTFT is shown in Figure 4. The drain current in the linear region were measured by applying a drain voltage of -1 V and a gate voltage of -40 V, while constant gate bias stress was induced by a temporal period of every 10s during 2500s. The fabricated OTFTs with two different types of insulators exhibited rapid decay in the drain current with time. However, the time-dependent decrease in the drain current is slightly slower for the device with the barrier-type insulator. It is thought that a smooth surface of a gate insulator is of primary importance for the device characteristics as well as the stable operation.

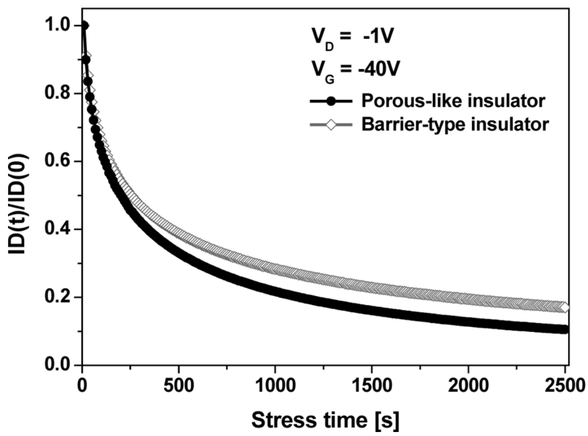

Figure 4. The time-dependant degradation curves of the fabricated OTFTs.

Conclusion

We investigated the electrical characteristics of OTFTs with two types of anodized aluminum oxide insulators which were prepared by varying the pH condition of an electrolyte. The surface morphology of an anodized aluminum oxide insulator is found to be decisive on the activation energy for the charge conduction and thus significantly influence on the electrical properties of OTFTs. We believe that the barrier-type insulator is versatile for the conduction of charge carriers driven by lowering the activation energy for a long range hopping in OTFTs.

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