Temperature dependence of frequency response characteristics in organic field-effect transistors

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The frequency response characteristics of semiconductor devices play an essential role in the high-speed operation of electronic devices. We investigated the temperature dependence of dynamic characteristics in pentacene-based organic field-effect transistors and metal-insulator-semiconductor capacitors. As the temperature decreased, the capacitance-voltage characteristics showed large frequency dispersion and a negative shift in the flat-band voltage at high frequencies. The cutoff frequency shows Arrhenius-type temperature dependence with different activation energy values for various gate voltages. These phenomena demonstrate the effects of charge trapping on the frequency response characteristics, since decreased mobility prevents a fast charge response for alternating current signals at low temperatures. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4711211]

In recent decades, extensive research has been conducted on organic field-effect transistors (OFETs) due to their attractive features such as low cost, large area, and flexibility.1–4 Most of the work so far has dealt with their static direct current (DC) characteristics such as output and transfer characteristics, through which some of the basic device parameters such as field-effect mobility (μFEET), threshold voltage (VT), and subthreshold swing (S) are extracted. Temperature dependence of these parameters has provided much insight with regard to the operation mechanisms of OFETs, particularly on charge transport in organic semiconductors.5,6 On the other hand, with the increasing use of organic electronic devices in practical applications,7 improvement in the frequency response characteristics (FRC) of OFETs becomes an essential requirement for high-speed operation of these devices.8–13 For instance, the frequency of 13.56 MHz is normally used for RF-ID tags, considered to be one possible application for organic devices.14 The dynamic alternating current (AC) characteristics can also provide fundamental information on charge transport in semiconductors and its limiting factors such as charge trapping at the semiconductor/dielectric interface.15–18 In the past, some works have been done to demonstrate the FRC of OFETs and different factors responsible for their FRC, such as channel length,8,19 parasitic capacitance,20 and contact resistance14, 19 were proposed. However, most of these works have been done through room temperature measurements, although temperature dependence of FRC is expected to provide tremendous information about the important factors of AC characteristics such as an effect of charge transport on the dynamic characteristics.

In this study, the temperature dependence of FRC was demonstrated on metal-insulator-semiconductor (MIS) capacitors and OFETs using pentacene as a p-type semiconductor. We carried out capacitance-voltage (C-V) and capacitance-frequency (C-f) measurements at various temperatures to investigate the dominant factors that affect FRC of organic devices. In particular, we focused on two device parameters, flat band voltage (VFB) and cutoff frequency (fc), and the dependence of these two parameters on the gate voltage (VG), temperature, and the applied frequency was examined. Through the systematic measurements, strong temperature and frequency dependency of the VFB and temperature and VG dependence of fc were observed. The mechanism of these tendencies is discussed with correlation to static measurement results, and charge trapping in organic semiconductor was found to be critical for AC characteristics.

In experiments, pentacene-based OFETs and MIS capacitors were fabricated on the same substrate by the following procedures. A highly doped Si (100) wafer with a 50 nm thick thermally oxidized SiO2 layer was used as a substrate, where the Si wafer is used as the gate electrode and the SiO2 layer acts as a gate insulator. The surface was ultrasonically cleaned in acetone and 2-propanol, followed by UV-O3 treatment for 5 min. To improve the interfacial quality between the gate insulator and organic semiconductor layer, one layer of polystyrene (PS) with ~30 nm thickness was deposited by spin-coating. The PS layer was annealed in air for 5 min at 120°C. Pentacene (Aldrich, purified with temperature gradient sublimation) was vacuum-deposited onto the PS layer-covered SiO2 surface at a growth rate of 0.1 nm/s up to a thickness of 40 nm. The pentacene

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deposition was carried out through a metal mask to isolate the active area of each device. Finally, copper (Cu) electrodes were deposited by vacuum evaporation through a metal mask and thus the OFETs and MIS capacitors were formed on the same substrate. Schematic cross-sections of these devices are shown in Figs. 1(a) and 1(b). An atomic force microscope image of the pentacene surface is shown in the inset of Fig. 1(c). The AC characteristics of the MIS capacitor were measured by an LCR meter (Agilent E4980A) and the static DC characteristics of the transistors were measured using an Agilent 4156C semiconductor device analyzer. All electrical measurements were performed under dark in high vacuum ($<5 \times 10^{-3}$ Pa). The $C-V$ characteristics were measured by applying a DC voltage in the range of 10 to $-10$ V along with a probe AC voltage of 100 mV. The $C-f$ characteristics were obtained by sweeping the frequency with a fixed $V_G$.

We first examined the static DC characteristics of the pentacene-based OFETs at various temperatures. The measured OFET had a channel length ($L$) and width ($W$) of 100 $\mu$m and 750 $\mu$m, respectively. Figure 1(c) represents the transfer characteristics of the OFET at a drain voltage of $-10$ V at various temperatures from 295 K to 80 K. One can observe a decrease in the drain current and a negative shift in the turn-on voltage with lowering of the temperature. These tendencies indicate thermally activated charge transport, which agrees well with the previously reported trends for OFETs with a polycrystalline or amorphous semiconductor film.\textsuperscript{21-23} The field-effect mobility ($\mu_{\text{FET}}$) was determined to be 0.58 cm$^2$/Vs at 295 K. An Arrhenius plot of the $\mu_{\text{FET}}$ is shown in Fig. 1(d), and the activation energy ($E_a$) was determined to be 56 meV. This value also corresponds well with previously reported $E_a$ values for OFETs based on a polycrystalline pentacene film.\textsuperscript{24}

The $C-V$ characteristics of the MIS capacitor were measured under various frequencies from 100 Hz to 100 kHz at different temperatures. Figures 2(a) and 2(b) show the characteristics obtained at 295 K and 120 K, respectively. In the case of $p$-type organic semiconductor devices, charge carriers were injected into the semiconductor layer from the upper electrode when a negative $V_G$ was applied, and these accumulated at the semiconductor/insulator interface. In the figures, the $C-V$ curves exhibit a typical output from accumulation to depletion when the voltage sweeps from $-10$ V to $+10$ V. The most important feature observed in the present $C-V$ curves is the large dispersion in the capacitance for varying frequencies and a shift in $V_{FB}$ observed at low temperatures. At a temperature of 295 K, the maximum capacitance ($C_{\text{max}}$) was obtained at high negative $V_G$, indicating the formation of a charge accumulation layer at the insulator/organic semiconductor interface and $C_{\text{max}}$ corresponds to the
capacitance of the gate insulator. The C-V characteristics of the MIS capacitor for various frequencies are fully overlapped at 295 K, and almost no capacitance-voltage frequency dispersion (CVFD) was observed in the temperature range down to 200 K. The $C_{\text{max}}$ started to decrease at high frequencies and the C-V characteristics tended to show CVFD when the temperature was down to 160 K or lower (Fig. 2(b)). Significantly large CVFD was observed at a temperature of 80 K (not shown). The change in the $C_{\text{max}}$ as a function of frequency at various temperatures is summarized in Fig. 2(c). The dependence of $C_{\text{max}}$ on temperature and frequency can be understood by the thermally activated charge transport and charge response for the AC signal in the semiconductor layer. With lowering of the temperature, the charge mobility decreases due to charge trapping in the pentacene film, as seen in the temperature dependence of $\mu_{\text{FET}}$ in Fig. 1(d). The lower mobility prevents charges from responding to AC signals at low temperatures. Thus, the semiconductor layer acts as a dielectric for the AC signals at higher frequencies, which decreases $C_{\text{max}}$. This tendency is clearly observed at a temperature of 80 K, where $C_{\text{max}}$ has almost the same value as the minimum depletion mode capacitance ($C_{\text{min}}$) of 3.3 nF at high frequencies (Fig. 2(c)), indicating that the semiconductor layer is fully depleted for the probe signals.

In silicon-based devices, extensive studies have been carried out on the $V_{\text{FB}}$ of MIS structures, which directly affects the $V_T$ of transistors, particularly for high-$k$ gate dielectric devices.\textsuperscript{24,25} We extracted the $V_{\text{FB}}$ values from the C-V curves of the pentacene-based MIS capacitor at various temperatures by using an experienced method derived by the Motorola Digital DNA laboratory as the voltage corresponding to $C_{\text{FB}} = (C_{\text{max}} - C_{\text{min}}) \times 0.66 + C_{\text{min}}$, where $C_{\text{FB}}$ is the flat-band capacitance.\textsuperscript{26} The frequency dependence of $V_{\text{FB}}$ is summarized in Fig. 3. Similar to $C_{\text{max}}$, the $V_{\text{FB}}$ is found to be dependent on applied frequency and temperature. The $V_{\text{FB}}$ is almost constant for a temperature at 280 K, but varies from $-0.5$ V at 100 Hz to $-2.8$ V at 100 kHz at a temperature of 160 K, and an even larger shift was observed at lower temperatures. This large shift in $V_{\text{FB}}$ may greatly change the $V_T$ value of the corresponding OFET devices when they operate under various frequency situations, which would lead to the unstable device properties. To realize $V_T$ stability under high-frequency operation of OFETs, it is critical to understand the mechanism of this $V_{\text{FB}}$ frequency dispersion.

It is known that the $V_{\text{FB}}$ can be determined by the metal-semiconductor work function difference $\phi_{\text{MS}}$ and various fixed or trapped charges in MIS capacitors, as described by the following expression:\textsuperscript{27}

$$V_{\text{FB}} = \phi_{\text{MS}} - \frac{Q_f}{C_{\text{ins}}} - \frac{Q_l (\phi_l)}{C_{\text{ins}}} - \frac{1}{C_{\text{ins}}} \int_{0}^{\text{ins}} x \rho_s (x) dx - \frac{1}{C_{\text{ins}}} \int_{0}^{\text{ins}} x \rho_m (x) dx,$$

(1)

where $Q_f$ is the fixed charge located near the semiconductor/insulator interface and $Q_l$ is the interface trapped charge (occupation of interface trap states is a function of surface potential $\phi_l$). $\rho_s$ and $\rho_m$ are the insulator charges and mobile ions per unit volume, and $C_{\text{ins}}$ and $t_{\text{ins}}$ are the insulator capacitance and thickness, respectively. In the case of organic thin-film devices, the effect of charge traps associated with the organic semiconductor layer seems to be more significant than that of other fixed charges such as dielectric trapped charges and mobile ions. Thus, we consider that $Q_f$ ($\phi_l$) is probably a dominant factor that mainly affects $V_{\text{FB}}$. It should be noted that charge trapping at the interface between the top electrode and the organic semiconductor also causes a large $V_{\text{FB}}$ shift in the case of organic thin-film devices. The effect of the top electrode interface on $V_{\text{FB}}$ can be found in our previous report, where the insertion of an injection-assist layer into the contact interface causes a large positive shift in the C-V characteristics.\textsuperscript{28} Since charge trapping at both semiconductor/insulator and electrode/semiconductor interfaces should be a thermally activated process, it can be concluded that the $V_{\text{FB}}$ can be temperature dependent.

The above results demonstrate that charge trapping in an organic semiconductor layer plays an important role in the $V_{\text{FB}}$ frequency response characteristics. To further investigate the frequency response characteristics of OFETs, we carried out C-f measurements on OFETs at various temperatures for extraction of $f_c$, the most common parameter showing the dynamic response capability of OFETs. $f_c$ can be defined as\textsuperscript{8,19}

$$f_c = \frac{1}{2 \pi} \cdot \frac{g_m}{C_{\text{ins}} \cdot WL} = \frac{\mu (V_G - V_T)}{2 \pi L^2},$$

(2)

where $g_m$ is the transconductance. As seen in Eq. (2), $f_c$ should be a function of $V_G$ and $L$, which has already been clarified for OFET devices.\textsuperscript{19} The C-f measurement configuration used in the present study is shown in Fig. 1(a), where source and drain electrodes are electrically shorted and connected to the LCR meter, as described in Ref. 19. The OFET has comb-shaped source/drain electrodes with $L = 150 \mu$m. The obtained C-f characteristics at a constant $V_G$ of $-20$ V at different temperatures are shown in Fig. 4(a); here, the C-f characteristics obviously depend on the measurement temperature. The $f_c$ values were estimated as the intersection points of the extrapolated fitting lines and the capacitance of the low frequency region (Fig. 4(a)). The extracted $f_c$ values are plotted as a function of the inverse of the temperature with least square fitting lines at various $V_G$ in Fig. 4(b). In this figure, it can be clearly seen that the $f_c$ values show Arrhenius-type temperature dependence as observed for $\mu_{\text{FET}}$ of the OFET in Fig. 1(d). At higher $V_G$, the
These values are very similar to the $E_a$ of 56 meV obtained from $\mu_{\text{FET}}$ in Fig. 1(d), indicating strong correlation between $f_c$ and $\mu_{\text{FET}}$. The observed temperature and $V_G$ dependence of $f_c$ is related to multiple trapping and release model for charge transport. This mechanism results in the temperature and $V_G$ dependence of the mobility, which can be associated with $f_c$ as indicated in Eq. (2). Thus, thermally activated charge transport caused by charge trapping at localized states is a critical factor that limits frequency response characteristics of OFETs.

In summary, the frequency response characteristics of pentacene-based OFETs and MIS capacitors, particularly instability of $C-V$ and $f_c$, are investigated through temperature dependent $C-V$ and $C-f$ measurements. At low temperature, the $C-V$ characteristics tend to show large frequency dispersion and $V_{FB}$ is shifted to a negative voltage. A significant temperature dependence is also observed for the $C-f$ characteristics, where the $f_c$ shows Arrhenius-type temperature dependence with different $E_a$ for variation of $V_G$. These phenomena can be explained by thermally activated charge transport described by a multiple trapping and release model, as decreased charge carrier mobility prevents fast charge response for AC signals at low temperatures.

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$C_f$ values become higher and the slope of the fitting line is smaller (Fig. 4(b)). Accordingly, $E_a$ values of $f_c$ at different $V_G$s were calculated and are summarized as a function of $f_c$ at different $V_G$s. These values are very similar to the $E_a$ of 56 meV obtained from $\mu_{\text{FET}}$ in Fig. 1(d), indicating strong correlation between $f_c$ and $\mu_{\text{FET}}$. The observed temperature and $V_G$ dependence of $f_c$ is related to multiple trapping and release model for charge transport. This mechanism results in the temperature and $V_G$ dependence of the mobility, which can be associated with $f_c$ as indicated in Eq. (2). Thus, thermally activated charge transport caused by charge trapping at localized states is a critical factor that limits frequency response characteristics of OFETs.

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