Flexible Thermal Sensors Based on Organic Field-Effect Transistors with Polymeric Channel/Gate-Insulating and Light-Blocking Layers

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ABSTRACT: Here, we report flexible thermal sensors based on organic field-effect transistors (OFETs) that are fabricated using polymeric channel and gate-insulating layers on flexible polymer film substrates. Poly(3-hexylthiophene) and poly-[methyl methacrylate] were used as the channel and gate-insulating layers, respectively, whereas indium-tin oxide-coated poly(ethylene naphthalate) films (thickness = 130 μm) were employed as the flexible substrates. Aluminum-coated polymer films were attached on top of the channel parts in the flexible OFETs to block any influence by light illumination. The present flexible OFET-based thermal sensors exhibited typical p-type transistor characteristics at a temperature range of 25−100 °C, while the hole mobility of devices was linearly increased with the temperature. The drain current could be amplified at various temperatures by adjusting the gate and drain voltages. In particular, stable sensing performances were measured during the repeated approaching/retreating cycle with a heat source. The flexible OFET thermal sensors attached on human fingers could sense heat from human fingers as well as from approaching objects.

1. INTRODUCTION

Temperature is one of the most important indicators in our daily life because it provides useful information on environmental changes, human body conditions, and so forth.1−3 Recently, the temperature sensing has become crucial for the safety control of various mobile electronic devices with rechargeable batteries such as smart watches, smart phones, smart pads, and notebook computers.4−6 Since early attempts for the measurement of temperature, a variety of thermal (temperature) sensors have been invented because of the development of the material and device technology.7−11

Thermal (temperature) sensors can be classified into two categories, contact type and noncontact type, according to the method of temperature measurement. The contact-type thermal sensors need to directly contact the sensing parts to target objects, whereas the noncontact-type thermal sensors measure the heat radiated from the objects.12−15 However, most of the conventional thermal sensors are fabricated with metals and/or inorganic materials, which in principle lack inherent flexibility and require high-temperature processes, so that they have limitations as a flexible thermal sensor for various applications in the flexible electronics era.

In this regard, organic electronic devices are considered to be a good approach for flexible thermal sensors because they can be fabricated using flexible plastic film substrates at low temperatures.16−20 Of various organic electronic devices, organic field-effect transistors (OFETs) have been widely applied for sensors and detectors because of their advantages in signal amplification by adjusting gate voltages in the presence of the third (gate) electrode.21−30 However, no detailed study has been so far reported on the flexible thermal sensors based on OFETs.

In this work, we have attempted to fabricate the flexible OFET-based thermal sensors using 130 μm thick poly(ethylene naphthalate) (PEN) film substrates. In particular, the present flexible OFET thermal sensors consist of polymeric channel and gate-insulating layers, poly(3-hexylthiophene) (P3HT) and poly[methyl methacrylate] (PMMA), whereas aluminum-coated polymer films were employed to prevent any influence of light from the surrounding environments. The performance of the flexible OFET thermal sensors was examined by varying the temperature of heat sources, while their sustainability was tested through the continuous measurement of drain current by repeating the approach of heat sources to the channel region. The possibility of practical applications was demonstrated by attaching the flexible OFET thermal sensors on human fingers to measure the internal heat from the human body as well as external heat from heat sources away from the fingers.

2. RESULTS AND DISCUSSION

The flexible OFET thermal sensors were fabricated by employing the transistor configuration of bottom-gate and...
top-source/drain electrodes, as shown in Figure 1a. The PMMA gate-insulating layers were first spin-coated on the indium-tin oxide (ITO)-coated PEN substrates, followed by thermal annealing to make a hard layer that can withstand the attack of toluene (solvent) from the P3HT solutions during the channel layer coating (see details in the Experimental Section). After coating the P3HT channel layers, the source and drain electrodes were formed by the thermal evaporation of silver in a vacuum chamber. Finally, the aluminum-coated PEN films were attached on top of the OFETs to block light from outside because the P3HT channel layer is very sensitive to the light illumination.31−35 As shown in Figure 1b, the flexible OFET thermal sensors exhibited a typical p-type transistor behavior when it comes to the output and transfer curves. The output curves clearly showed a saturation in drain current ($I_D$) as the drain voltage ($V_D$) increased negatively at a fixed gate voltage ($V_G$). The transfer characteristics of the present flexible devices delivered the hole mobility ($\mu_h$) of $\sim 7.3 \times 10^{-3}$ cm$^2$/V·s and the threshold voltage ($V_{TH}$) of 5.4 V (see Figure S1).

The output characteristics of devices were measured by varying the temperature in the channel layer (see Figure 2). As the temperature increased from 25 to 100 °C, the output curve was gradually shifted toward the negative drain current direction irrespective of gate voltages that are negatively higher than the threshold voltage ($V_{TH} = 5.4$ V in Figure 1b) (see Figure 2a). However, the shape of the output curves was well-maintained even in the presence of the large increase in drain current, according to the temperature increase. This result basically indicates that the charge transport in the channel region could be improved by the increased temperature.36−38 In addition, it is considered that the injection of external charge carriers was also enhanced by the improved contact resistance between the P3HT layer and the Ag electrodes.39,40 As shown in Figure 2b (see Figure S2 for all temperatures), the output curves were obviously shifted toward the negative drain current direction by increasing the gate voltages when the channel temperature was kept constant. Here, it is worthy to note that the drain current level at the same gate voltage was gradually increased as the temperature increased.

Then, the transfer characteristics of devices were examined by varying the temperature in the channel layer from 25 to 100 °C. As shown in Figure 3a, the transfer curves were gradually shifted toward the higher drain current direction, irrespective of gate voltages, when the temperature in the channel layer increased. The extent of drain current increase became more pronounced as the gate voltage increased at a constant drain voltage. This result supports that the increased temperature can...
improve the charge transport in the channel region, as discussed above. A close investigation finds that the threshold voltage was almost linearly increased as the temperature increased (see the inset graphs). In particular, it is worthy to note that the shift of threshold voltage \( V_{TH} = 6.8 - 9 \) V was not much affected by the drain voltage but by the temperature (see Figure S3). As observed from the transfer curves in Figure 3b (see Figure S4 for all temperatures), the drain current increase was steeper as the drain voltage increased negatively irrespective of the temperature. This trend reflects that the present flexible devices function properly at the temperature tested in this work.

To understand the overall trend of drain current change according to the temperature change, the drain current from both output and transfer curves was plotted as a function of temperature in Figure 4. Interestingly, as shown in Figure 4a, the change in drain current from the output curves was linearly proportional to the temperature irrespective of drain voltages. However, the slope (ratio of drain current to temperature, \( \Delta I/\Delta T \)) was increased (negatively) as the drain voltage increased at a fixed gate voltage. A similar trend was measured for the trend of drain current from the transfer curves (see Figure 4b). The higher \( \Delta I/\Delta T \) at higher voltages can be attributable to a better charge transport at higher drain and gate voltages, as given by the trend of hole mobility according to the temperature in Figure 4c. In addition, the hole mobility was found to have a linear correlation with the temperature (\( \Delta \mu_h/\Delta T = 2.34 \times 10^{-5} \) cm²/V·s·°C), which may support the major role of temperature on the performance of the present flexible devices (note that the capacitance was also linearly increased with the temperature, as shown in Figure S5). Here, the linear dependence of hole mobility on the temperature can be theoretically explained by the linear relationship of carrier mobility with both the drain current and the capacitance of gate insulators (see the charge carrier mobility equation in Figure S1).

On the basis of the detailed investigation on the temperature-dependent characteristics above, the performance of the present flexible OFET thermal sensors was examined by the repeated measurement of drain current change to the approaching heat sources (see Figure 5a). Three different temperatures in the channel region were controlled by adjusting the power of heat sources, while the approaching distance was fixed 0.5 cm for all cases. As shown in Figure 5b, the drain current was increased when the heat source approached the PEN side of the flexible OFET thermal sensors. Upon retreating the heat source, the drain current was decreased immediately in the presence of a relatively longer tail compared with the drain current shape in...
the approaching case. The longer detail in the retreating event can be ascribed to the heat remaining in the devices after retreating the heat source. It is also noticeable that the drain current signals were quite stable upon the approaching and retreating events of heat sources irrespective of the temperature (see Figure S6 for the extended measurement). To briefly investigate the response of the present devices, the elevated signals in Figure 5b were fitted with a single exponential equation so as to extract the response time. As shown in Figure S7a, the fitting was carried out separately at around the boundary (inflection) point by considering the approaching time (approximately 1.5 s) of heat sources. The extracted response time was linearly decreased with temperature for both parts (see Figure S7b). Considering the data acquisition resolution time (ca. 200 ms) of the present measurement system, the actual response time of approximately 50–100 ms at 100 °C can be obtained for the present flexible OFET thermal sensors.

Finally, the flexible OFET thermal sensors were attached on human fingers to test the feasibility of monitoring body temperature as well as surveilling (sensing) the approaching objects with heat. As shown in Figure 6a, the drain current was negatively increased as soon as the flexible OFET thermal sensor attached on the finger. Interestingly, while the flexible sensor was steadily placed on the finger, the drain current was quite well-maintained for about 200 s. Upon removing the flexible sensor from the finger, the drain current went back to the baseline. Then, as shown in Figure 6b, when the heat source approached the flexible OFET thermal sensor on the finger, the drain current was quickly increased (negatively) from its level by the body temperature. Although the heat source did steadily dwell near the flexible sensor for keeping 50 °C in the channel region, the drain current was well-maintained in the presence of marginal oscillations. Then, the drain current was quickly decreased as soon as the heat source was retreated suddenly. It is worthy to note that the drain current signal was greatly affected by the ambient (room) light for the devices without the light-blocking layers when it comes to the already increased initial current level as well as the continuously increasing trend of the drain current (see Figure S8).

3. CONCLUSIONS

The flexible OFET thermal sensors were fabricated by coating polymeric channel and gate-insulating layers (P3HT and PMMA) on the ITO-coated plastic film substrates (PEN), followed by attaching the aluminum-deposited polymer films for light blocking. The flexible devices exhibited a typical p-type transistor behavior with a hole mobility of approximately $7.3 \times 10^{-3}$ cm$^2$/V·s. Both output and transfer curves were gradually shifted to a (negatively) higher drain current direction as the temperature applied increased from 25 to 100 °C, while they were obviously changed by varying the gate and/or drain voltages even at higher temperatures. The change in drain current and hole mobility was linearly proportional to that of temperature. The ratio of drain current to temperature became larger as the gate and drain voltages increased. The present flexible devices exhibited a stable sensing performance during the repeated approaching/retesting with a heat source. In particular, the flexible OFET thermal sensors attached on human fingers could sense heat from the human fingers as well as from the approaching objects. Hence, the present flexible OFET thermal sensors are expected to be used as a sensory component for a variety of applications including medical instruments, artificial skins for humanoid robots, surveillance systems, and so forth.

4. EXPERIMENTAL SECTION

4.1. Materials and Solutions. P3HT (weight-average molecular weight = 30 kDa, polydispersity index = 1.7, regioregularity > 97%) was supplied from Rieke Metals (Lincoln, NE, USA), whereas PMMA (weight-average molecular weight = 120 kDa, polydispersity index = 2.2) was purchased from Sigma-Aldrich (USA). The P3HT powders were dissolved in toluene (concentration = 10 mg/mL). The PMMA solutions were prepared using chlorobenzene (concentration = 85 mg/mL). The P3HT and PMMA solutions were vigorously stirred at 60 °C before spin-coating.

4.2. Device Fabrication. ITO-coated PEN substrates were patterned to make the 30 mm × 1 mm ITO stripe as a gate electrode, followed by cleaning in acetone and isopropyl alcohol. After drying, the ITO/PEN substrates were treated with UV–ozone (28 mW/cm$^2$) for 20 min. Then, the PMMA gate-insulating layers were prepared by spin-coating the PMMA solutions, followed by thermal annealing at 120 °C for 30 min to make them dense for withstanding the attack of toluene (solvent) in the P3HT solutions. Then, the P3HT channel layers were spin-coated on the PMMA layers and further annealed at 120 °C for 15 min. These samples were transferred to a glove box and used as flexible OFET thermal sensors.
into an argon-filled vacuum chamber for the deposition of source/drain electrodes (thickness = 65 nm) through a shadow mask in a vacuum of approximately $1 \times 10^{-6}$ Torr. The top light-blocking films were prepared by depositing aluminum (thickness = 100 nm) on one side of 60 μm thick PEN films. Finally, the sticky adhesive side of the Al-deposited PEN films was attached on the channel parts of devices (see Figure 1a). A diode-type device (ITO/PMMA/Al) was fabricated to measure the capacitance of the gate-insulating layers (PMMA).

4.3. Measurements. The performance of transistors was measured using a semiconductor parameter analyzer (model 4200SCS, Keithley), whereas a surface profiler (Alpha-Step 200, Tencor Instruments) was used for the measurement of the film thickness. Thermal sensing experiments were performed using a home-built sensor measurement system equipped with a probe station (PS-CPSN2, ModuSystems) and a heat control part (TZAST, Autonics). A loadlike heat source was used and controlled to approach the flexible OFET thermal sensors. An infrared thermometer (62 Mini, Fluke) and a temperature-sensing unit (NI 9211, National Instrument) were used for the measurement of accurate temperatures near the channel region in the flexible OFET thermal sensors. The capacitance of diode-type devices (ITO/PMMA/Al) that were placed inside a temperature-controlled sample chamber was measured using a potentiostat (VersaSTAT 4, AMETEK).

ASSOCIATED CONTENT

Supporting Information
The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsomega.7b00494. Illustration for the calculation of hole mobility from the transfer curve, output curves of the flexible OFET thermal sensors at different temperatures, capacitance as a function of temperature for the gate-insulating layers, change in drain current upon repeated stimulations, and change in drain current for the flexible OFET thermal sensor without the light-blocking layer (PDF)

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Notes
The authors declare no competing financial interest.

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