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Organic Phototransistors With All-Polymer Bulk Heterojunction Layers of *p*-Type and *n*-Type Sulfur-Containing Conjugated Polymers

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Abstract—**All-polymer phototransistors were fabricated using both glass and flexible plastic film substrates by employing bulk heterojunction channel layers of p-type polymer (P3HT) and n-type polymer (THBT-ht). The devices could detect the entire visible light because the n-type polymer could sense photons in the deep red parts (>650 nm). The responsivity of devices was higher at the lower light intensity, while it could be controlled by varying the gate and/or drain voltages. Similar performances were measured for flexible all-polymer phototransistors with a bottom-source/drain and top-gate electrode configuration.**

Index Terms—**Organic phototransistor, all-polymer, bulk heterojunction, n-type polymer.**

I. INTRODUCTION

OPTICAL sensors have played a critical role in the development of various electronic devices and systems including lux meters, bar-code scanners, high definition cameras, security vision systems, industrial automatic control systems, cleaning robots, etc. [1]–[3]. Most conventional optical sensors such as photodiodes, charge-coupled devices, and complementary metal oxide semiconductor detectors are fabricated with inorganic semiconductors and can deliver high performance following a substantial development effort [4]–[6]. However, such optical sensors have poor flexibility and small sensing areas [7], which become a hurdle for sophisticated applications in the growing flexible and wearable electronics sector [8]. To meet the ideal requirements for flexible/wearable electronics, it is mandatory for optical sensors to be bendable and readily fabricated in a variety of shapes and sizes. Medical diagnostic applications also stand to benefit from low-cost photodetector-instrumented microfluidic lab-on-a-chip structures [9] and wrap-around X-ray detectors [10], [11].

In this regard, organic optical sensors have recently been spotlighted [12], [13]. To date, organic photodiodes have been relatively much studied because of the simple diode-type device structure (electrode/sensing layer/electrode) leading to large sensing area [9]–[11], [14]–[23] and wrap-around X-ray detectors [10], [11], [24]. However, organic photodiodes need additional driving circuits such as transistors for 1-D or 2-D array sensors [25]. To overcome this complexity, organic phototransistors (OPTRs) have been introduced, combining both sensing and control functions in a single device [26]–[36].

Most OPTRs have employed a single material as sensing (channel) layer as also used in many normal transistors [35], [37]–[46]. However, considering the basic function of phototransistors to efficiently convert photons to individual charges (holes and electrons) and quickly transport the charges for detection, a bulk heterojunction (BHJ) type channel layer rather than a single channel material is expected to be more effective in terms of high charge separation efficiency [36], [47]–[51]. BHJ layers consisting of n-type (electron-accepting) and p-type (electron-donating) polymers may be preferable for improved flexibility, and to control phase segregation. We have previously reported devices of this type, composed of poly(9, 9-dioctlyfluorene-*co*-benzothiadiazole) (F8BT; n-type) and poly(3-hexylthiophene) (P3HT; p-type) [52]. However, the F8BT in these all-polymer phototransistors has a relatively wide optical gap (∼2.4 eV) such that its absorbance doesn't enhance the long wavelength response beyond that of P3HT.

In this work, we report the use of poly(3-hexylthiophene-*co*-benzothiadiazole) (THBT, c.f. Fig. 1(a) for chemical structure) as a replacement n-type polymer. THBT has relatively narrow band gap (1.8 eV) and deep-lying lowest unoccupied molecular orbital (LUMO) energy (3.9 eV) [53]. In particular, hexylthiophene-end capped THBT polymers, namely THBT-ht, were newly synthesized to avoid the influence of the un-reacted bromine and/or boron end groups in the standard Suzuki-coupled THBT polymer. The blend fraction of THBT-ht to P3HT in this study was fixed as 30 wt%. Additionally, a high work-function (∼5.0 eV) nickel (Ni) interfacial layer (∼10 nm) was introduced to increase charge injection from the aluminum (Al) source/drain electrodes into the P3HT:THBT-ht BHJ layer. The

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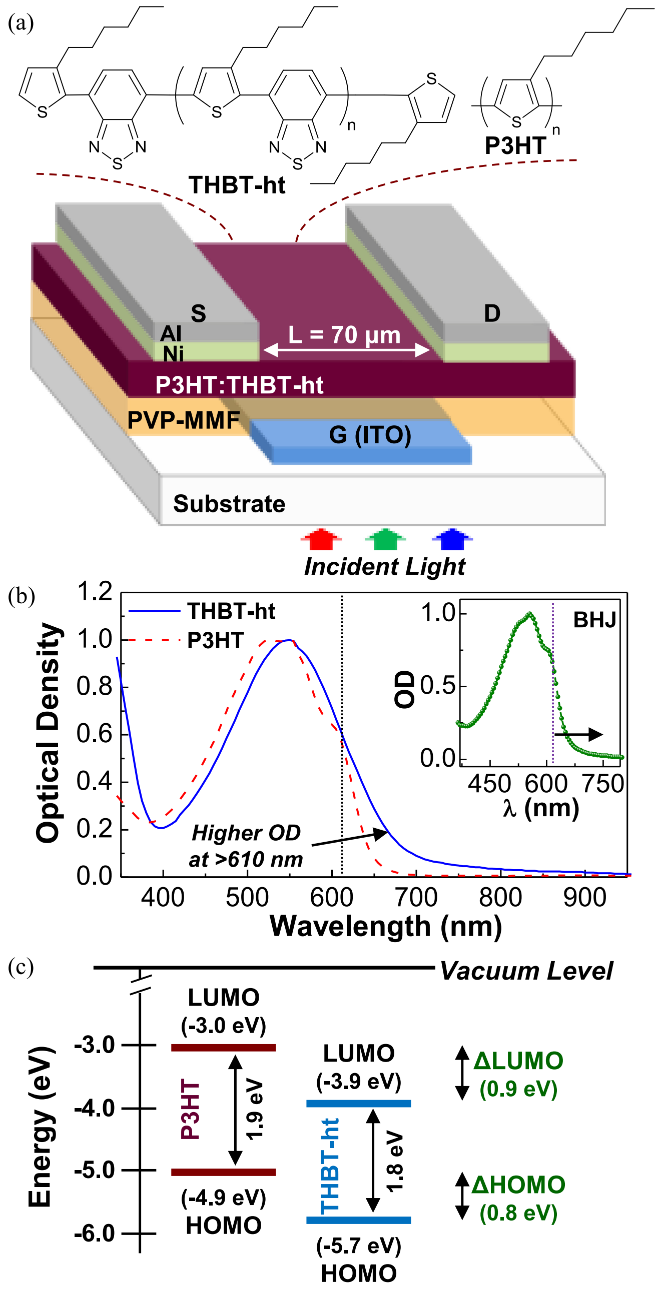


Fig. 1. (a) Materials and device structure for all-polymer phototransistors with the P3HT:THBT-ht layer (glass substrates). (b) Optical density (OD) as a function of wavelength (λ) for pristine polymers and BHJ (P3HT:THBT-ht) layer. (c) Comparison of HOMO and LUMO levels for P3HT and THBT-ht.

performance of our phototransistors was examined for varying incident light intensities (P_{IN}) and wavelengths.

II. EXPERIMENTAL DETAILS

A. Preparation of Materials and Solutions

The THBT-ht polymer (weight-average molecular weight = ∼10 kDa, polydispersity index = 2.2) was synthesized according to the procedure described in our previous report [53], but 3-hexylthiophene (ht) groups were attached to the end of the THBT polymer chain through an end-capping reaction by adding 2, 1, 3-benzothiadiazole-4, 7-bis(boronic acid pinacol ester) (2B-BT) and 2-bromo-3-hexylthiophene (Br-HT). The P3HT polymer (weight-average molecular weight = 70 kDa, polydispersity index = 1.6, regioregularity = 95%) was purchased from Solaris (Canada) and used without further purification. The binary blend solutions of P3HT and THBT-ht (P3HT:THBT-ht = 70:30 by weight) were prepared using 1, 2-dichlorobenzene as a solvent. Poly(4-vinylphenol) (PVP) and poly(melamine-*co*-formaldehyde) methylated (MMF) were mixed in propylene glycol monomethyl ether acetate (PGMEA) for 72 h at 60 °C, leading to partly cross-linked yellowish solutions from which the gate insulator was subsequently deposited.

B. Fabrication of Phototransistors and Pristine Films

First, indium-tin oxide (ITO)-coated glass substrates were patterned using conventional photolithography process to produce 1 mm × 12 mm stripes for use as the gate electrodes. These were next cleaned using acetone and isopropyl alcohol before the PVP-MMF gate insulator solution was coated on top and prebaked at 120 °C for 10 min. The insulator was further cured at 250 °C for 30 min leading to an insoluble 550 nm thickness gate dielectric. On top of the PVP-MMF layer, the P3HT:THBT-ht BHJ layer (thickness = 50 nm) was spin-coated and thermally annealed at 150 °C for 30 min. Finally, Ni (10 nm) and Al (50 nm) electrodes were deposited on the P3HT:THBT-ht BHJ layer through a shadow mask using a thermal evaporator housed within an argon-filled glove box. The channel length and width were 70 μm and 2 mm, respectively (see Fig. 1(a)). To fabricate flexible phototransistors, bilayer Al/Ni source/drain electrodes were deposited on the poly(ethylene terephthalate) (PET) films (200 μm, thickness). The BHJ layers, followed by a $\sim$1 μm thickness poly(methyl methacrylate) (PMMA) gate insulator, were then spin-coated and an Al gate electrode thermally evaporated on top. Measurements of optical absorption and/or ionization potential were performed on spin-coated P3HT and THBT-ht films on quartz and/or glass substrates.

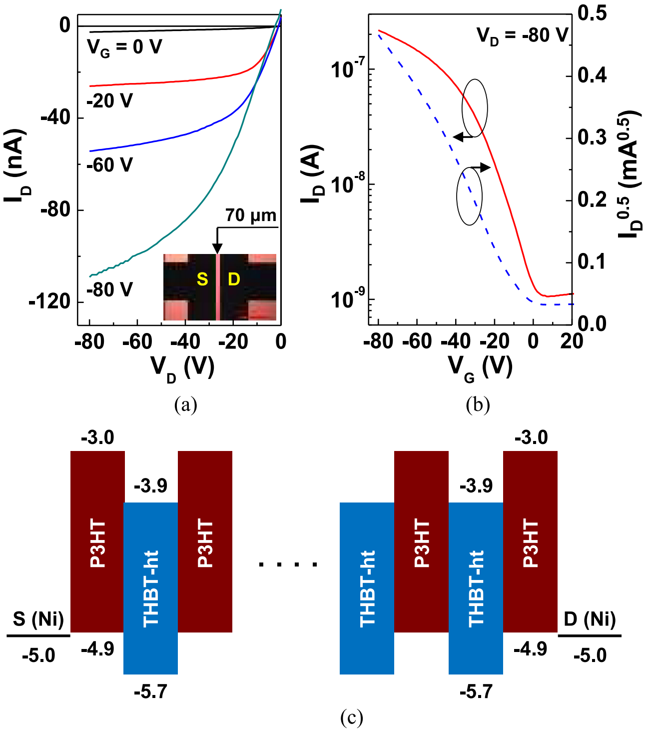
C. Measurements

A UV-visible spectrophotometer (Optizen 2120+, Mecasys Co., Ltd) was used to measure the optical absorption spectra of film samples, while the ionization potential of the polymer films was measured using a photoelectron yield spectrometer (AC-2, Hitachi High Tech). The transistor characteristics of devices were measured using a semiconductor parameter analyzer (Keithley 2636B). Monochromated (CM110, Spectral Products) light from a tungsten-halogen lamp (150 W, ASBN-W, Spectral Products) was used to measure the phototransistor performance: The incident light intensity (P_{IN}) was controlled by neutral density filters and monitored with a calibrated photodiode. Apparent (R_A) and corrected (R_C) responsivities are calculated with $R_A = I_{DP}/P_{IN}$ and $R_C = (I_{DP} - I_{DD})/P_{IN}$, where I_{DD} and I_{DP} are the drain current in the dark and under illumination, respectively.

III. RESULTS AND DISCUSSION

A. Energy Band Structure and Transistor Characteristics

As shown in Fig. 1(b), the optical absorption spectrum of THBT-ht extends to slightly longer wavelength than that of

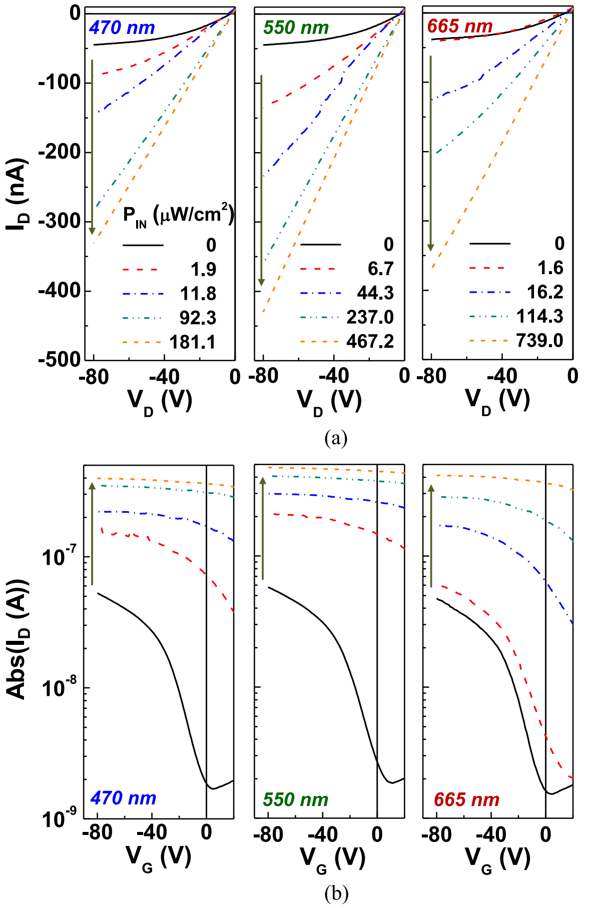


(a) (b) (c)

Fig. 2. Output (a) and transfer (b) characteristics under dark condition for the device (glass substrates) with the P3HT:THBT-ht BHJ layer: The inset in (a) shows the optical microscope image on the channel area. (c) Flat energy band diagram to show the possibility of charge blocking by the THBT-ht component in the P3HT domain in the case of ideally mixed state (note that the "eV" unit is omitted in the energy value).

P3HT with a higher optical density beyond 610 nm (vertical line). In particular, the absorption tail (not scattering corrected) for THBT-ht reaches as far as $\sim$750 nm whereas P3HT alone should not absorb photons past $\sim$650 nm (red light). The measured ionization potential of THBT-ht was similar to that reported previously for THBT [53], leading to the highest occupied molecular orbital (HOMO) energy of –5.7 eV. Depending on blend microstructure, this deep-lying HOMO might be expected to interrupt hole-transport (c.f. Fig. 2(c) for an illustrative extreme). The corresponding LUMO energy was estimated by subtracting the optical band gap (see Fig. 1(c)). The LUMO offset between P3HT and THBT-ht is then $\sim$0.9 eV, which is more than sufficient to drive charge separation between them [12], [53].

With the above energy level information in mind, the transistor characteristics were first measured in the dark in order to check that the devices function as expected. We found typical p-type behavior (note that n-type transistor performance could not be obtained because of poor n-channel feature and low-loading of THBT-ht). In particular, the output curves in Fig. 2(a), showed an obvious saturation in drain current (I_D) as the drain voltage (V_D) increased negatively, with higher saturation currents as the gate voltage amplitude ($|V_G|$)) increased. In addition, I_D (at negative V_D) increased substantially as $|V_G|$ increased (see Fig. 2(b)). We note, however, that the drain current is relatively low compared to typical OFETs with a P3HT channel layer on its own [54]–[56]; albeit similar to that for F8T2 [35]. This relatively inferior performance in the dark may be attributable to the trapping effect of the THBT-ht component mixed with the P3HT for which there is a significantly deeper ($\Delta E \sim 0.8$ eV) HOMO level (see Fig. 2(c)) leading to charge blocking effects [57]. If true, this may also affect photodetector response time but it is not necessarily a problem for many applications, provided other factors, including responsivity and dynamic range, are sufficiently competitive.



(a) (b)

Fig. 3. Output (a) and transfer (b) characteristics of devices (glass substrates) under illumination of three major visible lights. The gate voltage was –80 V in (a), while the drain voltage was –80 V in (b). The incident light intensity P_{IN} is given for each light in (a), which is the same for (b). The arrows denote the increasing direction of drain current.

B. *Phototransistor Characteristics*

To examine the phototransistor performance, three wavelengths spanning most of the visible spectrum (470, 550 and 665 nm) have been selected to probe different parts of the P3HT:THBT-ht BHJ optical absorption spectrum. As shown in Fig. 3(a), the output curve $|I_D|$ values remarkably increased at all three wavelengths as the incident light intensity (P_{IN}) increased. A similar increase in $|I_D|$ was also measured for the transfer curves (see Fig. 3(b)) confirming that our phototransistors do sensitively respond to visible light. Focusing on the response to red light (665 nm) illumination, solely absorbed by

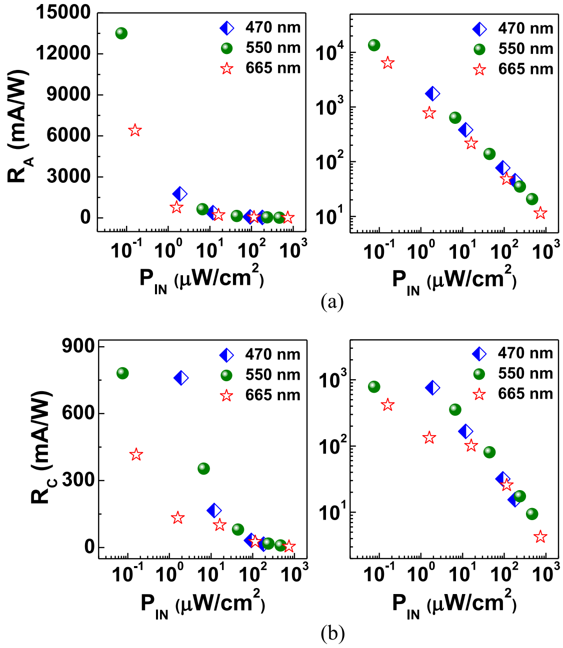


Fig. 4. Responsivity of devices (glass substrates) as a function of the incident light intensity for three major wavelengths: (a) apparent responsivity (R_A), (b) corrected responsivity (R_C) at $V_G = -80$ V and $V_D = -80$ V. Note that R_C was obtained by subtracting the dark current from R_A.

the THBT-ht component (not by P3HT), we find that the low light intensity (<10 μW/cm^2) $|I_D|$ response is relatively small compared to that seen at the other two wavelengths. It is likely that excitation of only the 30 wt% THBT-ht component reduces the probability of charge generation and limits charge collection. To understand this result explicitly, further studies on morphology are needed for the BHJ films with full compositions.

C. *Responsivity Trend*

From the phototransistor characteristics in Fig. 3, the apparent responsivity (R_A), which includes both dark current (field-effect current) and photocurrent, has been plotted as a function of the incident light intensity in Fig. 4(a). The apparent responsivity was remarkably enhanced for all three wavelengths as the incident light intensity decreased (see Fig. 4(a) left), which supports the reduced charge recombination at low light intensity in the case of BHJ channel layers [51], [52]. The logarithmic plot (see Fig. 4(a) right) shows quite a good linear relationship between the apparent responsivity and the incident light intensity. As shown in Fig. 4(b) left, the corrected responsivity (R_C), which is a genuine sensitivity without any dark current effect, was similarly increased as the incident light intensity decreased. The corrected responsivity data also clearly shows the reduced effect for red light (665 nm) discussed above.

The linear relationship on a logarithmic scale ($\log R_C \sim \log P_{IN}$) is found to be broken as the incident light intensity decreased. This nonlinear trend is particularly pronounced for red light detection so that it can be attributed to the change of both light-absorbing identity (from the P3HT component to the THBT-ht component) and resulting charge separation/transport processes, mediated by the blend microstructure. As summarized in Fig. 5, the present phototransistors can detect across the whole visible spectrum even though the responsivity is relatively low for red light. Considering the responsivity difference (ΔR) between the apparent responsivity and the corrected responsivity, better signal-to-noise ratios are expected when the present phototransistors are operated at a low gate voltage. Finally, it is noted that the reason why the present R_C values are relatively higher than the theoretical maximum values can be attributed to the different (perpendicular) geometry between the current flow and the incident light.

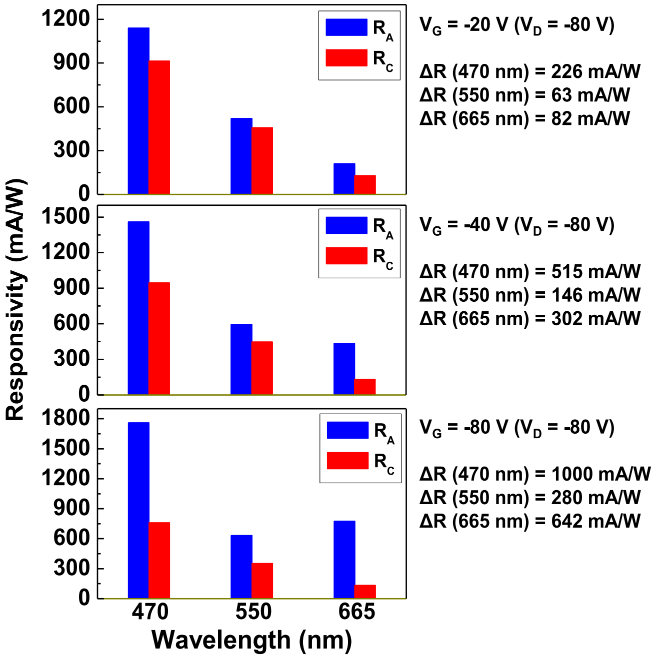
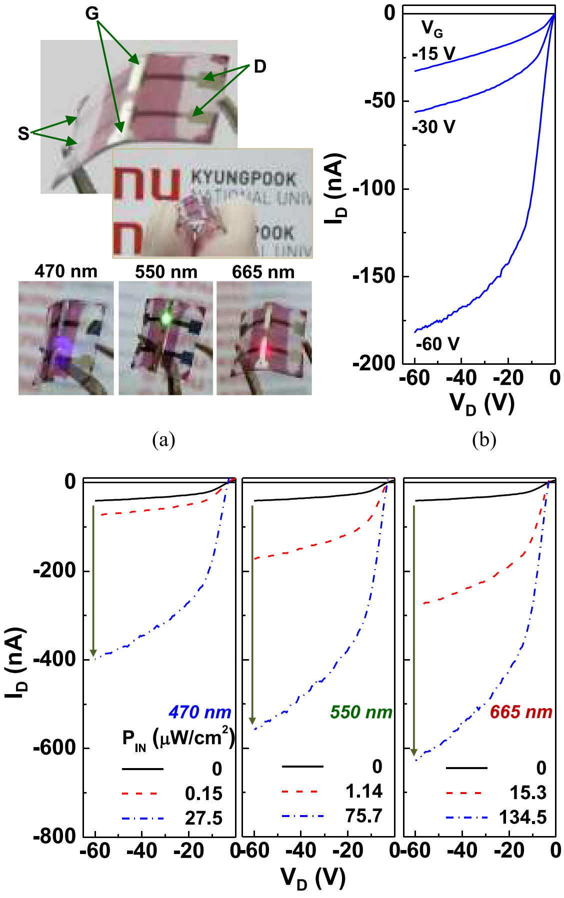


Fig. 5. Comparison between the apparent responsivity (R_A) and the corrected responsivity (R_C) of devices (glass substrates) for three major wavelengths at three different voltage conditions: (top) $V_G = -20$ V and $V_D = -80$ V, (middle) $V_G = -40$ V and $V_D = -80$ V, (bottom) $V_G = -80$ V and $V_D = -80$ V. All data were taken at the same (similar) incident light intensity (~6.7 μW/cm^2) for each wavelength. The responsivity difference (ΔR) between R_A and R_C is given on the right part for each case.

D. *Flexible All-Polymer Phototransistors*

Finally, flexible all-polymer phototransistors were fabricated employing PET film substrates (see Fig. 6(a)). As shown in Fig. 6(b), the flexible devices exhibited good output characteristics with clear saturation, comparable to the performance of glass-based devices (see Fig. 2(a) above). They also detected each of the three visible wavelengths (470, 550 and 665 nm), with the photocurrent controllable by changing the drain voltage (see Fig. 6(c)). The corrected responsivity reached ~4 A/W at 470 nm and ~2 A/W at 550 nm, considerably higher than obtained from our phototransistors on glass substrates. The improvements are expected to be attributable to the different geometry (bottom-source/drain and top-gate electrodes for the flexible phototransistors) as well as the different gate insulator (PMMA).



(a) (b)

(c)

Fig. 6. (a) Photographs of flexible all-polymer phototransistors with the P3HT:THBT-ht BHJ layer (device structure: PET/Al/Ni/BHJ/PMMA/Al): Bottom images show the flexible devices under illumination three major visible lights. (b) Representative output characteristics of the flexible device under dark condition. (c) Representative drain current change upon illumination of three different monochromatic lights for the flexible phototransistors at $V_G = -60$ V (note that the incident light intensity was different each other).

IV. CONCLUSION

All-polymer phototransistors were fabricated using both glass and PET film substrates by employing bulk heterojunction layers comprising P3HT (p-type) and THBT-ht (n-type) polymer blends. The present all-polymer transistors exhibited clear p-type transistor characteristics with current saturation, but the resulting drain current was relatively low, likely due (at least in part) to the hole-trapping role of the n-type component (THBT-ht). Under illumination with 470, 550 and 665 nm light, the device drain current was significantly increased. The photocurrent could be controlled by varying the gate and/or drain voltages. The corrected responsivity was relatively low for red light (665 nm), probably related to the low loading of THBT-ht (30 wt%). An understanding of the role of the blend microstructure and its control will be important to enhancing the device performance and allowing such flexible all-polymer phototransistors to contribute to the further development of highly-sensitive ultrathin/flexible plastic image sensors in the coming flexible electronics era. Finally, it is suggested to maximize the benefit of all-polymer phototransistors by using n-type conjugated polymers with strong optical absorptions in the longer wavelengths covering near infrared (NIR) range.

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