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# Effects of Ambient Gases on the Electrical Performance of Solution-Processed C8-BTBT Thin-Film Transistors

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## Abstract

We performed a systematic study of the influence of environmental conditions on the electrical performance characteristics of solution-processed 2,7-dioctyl [1] benzothieno[3,2-b][1]benzothiophene (C8-BTBT) thin-film transistors (TFTs). Four environmental exposure conditions were considered: high vacuum (HV), O<sub>2</sub>, N<sub>2</sub>, and air. The devices exposed to O<sub>2</sub> and N<sub>2</sub> for 2 h performed in a manner similar to that of the device kept in HV. However, the device exposed to air for 2 h exhibited significantly better electrical properties than its counterparts. The average and highest carrier mobility of the 70 air-exposed C8-BTBT TFTs were 4.82 and 8.07 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup>, respectively. This can be compared to 2.76 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup> and 4.70 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup>, respectively, for the 70 devices kept in HV. Furthermore, device air stability was investigated. The electrical performance of C8-BTBT TFTs degrades after long periods of air exposure. Our work improves knowledge of charge transport behavior and mechanisms in C8-BTBT OTFTs. It also provides ideas that may help to improve device electrical performance further.

**Keywords:** Solution process, C8-BTBT, Thin-film transistors, Air stability, Ambient gases

## Introduction

Due to the advantages of low deposition temperature, high mechanical flexibility, low cost, and large area production, organic semiconductor materials have recently been widely investigated for various electronic device applications such as organic light-emitting diodes, organic photovoltaic devices, and organic field-effect transistors [1–4]. Organic semiconductors can be divided into two main categories: conjugated polymers and small molecule organic semiconductors [3]. Compared with conjugated polymers, small molecule organic semiconductors offer high degrees of ordering, stacking density, and material purity. These advantages facilitate the fabrication of high-performance devices [5–8]. C8-BTBT is a representative small molecule organic semiconductor material [5]. Extensive research has been performed to study its charge

transport mechanisms [9], low-cost fabrication methods [10, 11], growth and microstructure formation on various substrates [12–14], metal/semiconductor contact characteristics [15, 16], and strategies to increase its carrier mobility [11, 17–19]. Thus far, there is no systematic study on the impact of ambient gases on the electrical performance of C8-BTBT-based devices. On the one hand, environmentally induced changes to the electrical performance characteristics of such organic devices are a critical problem that must be solved to provide stable operation for future commercial applications. On the other hand, such effects imply the potential for use of C8-BTBT-based devices as gas sensors.

In this study, C8-BTBT organic semiconductor films were fabricated via solution processing. The electrical properties of the C8-BTBT-based OTFTs were investigated in various ambient gases. The C8-BTBT OTFTs exhibited their highest carrier mobilities (~ 8 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup>) after exposure to air for 2 h. This is assumed to be closely related to the moisture in the air. The study also revealed that changes in the internal molecular structure play important roles in the electrical performance of the

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OTFTs. The present work not only deepened the understanding on the charge transport mechanisms and structural changes in C8-BTBT films but also provides new ideas to further improve their electrical performances.

## Methods

### C8-BTBT Deposition and OTFT Device Fabrication

A highly doped p-type silicon (100) wafer with a 50 nm thermally oxidized SiO<sub>2</sub> layer was used as the substrate for organic thin-film transistor preparation. The Si wafer was used as the bottom gate electrode, and the SiO<sub>2</sub> layer acted as the gate insulator. The substrates were cleaned with acetone, isopropanol, and deionized water for 5 min each using an ultrasonic cleaner. To ensure that the substrate surfaces were clean and dry, the substrates were dried on a hot plate in air for 15 min at 120 °C. In order to change the surface hydrophobicity, all the samples received a UV-ozone treatment for 1 min. This treatment time was chosen based on our previous results [10]. In a previous study, a C8-BTBT OTFT exposed to 1 min of UV surface treatment exhibited better electrical performance than those exposed to other UV treatment durations or non-UV treatment. The organic semiconductor layer was made from high-purity C8-BTBT (≥ 99%) (Sigma-Aldrich) and PMMA (Aladdin) dissolved in chlorobenzene. The solution (0.5 wt% C8-BTBT and 0.5 wt% PMMA) was spin-coated onto 50 nm SiO<sub>2</sub> covered p++ substrate (2000 rpm for 40 s). Each spin-coating cycle produced one 45 nm layer of C8-BTBT film. After annealing at 60 °C for 2 h in air, MoO<sub>3</sub> (5 nm) was deposited via thermal evaporation through a metal mask. This buffer layer was designed to reduce the contact barrier between the Au electrode and C8-BTBT semiconductor and to improve charge injection. Finally, Au source and drain electrodes (40 nm) were fabricated via thermal evaporation using the same MoO<sub>3</sub> shadow mask. The resulting transistor devices had various channel lengths that ranged from 50 to 350 μm, but the same channel width of 1200 μm.

### Material and Device Characterization

An Agilent B1500A semiconductor device analyzer was used to measure device electrical performance. Surface morphologies and roughnesses were observed via tapping mode atomic force microscopy (Asylum Research). Raman spectroscopy characterizations were performed using a Renishaw Via Raman Microscope. The C8-BTBT layer thickness was measured using an ellipsometer.

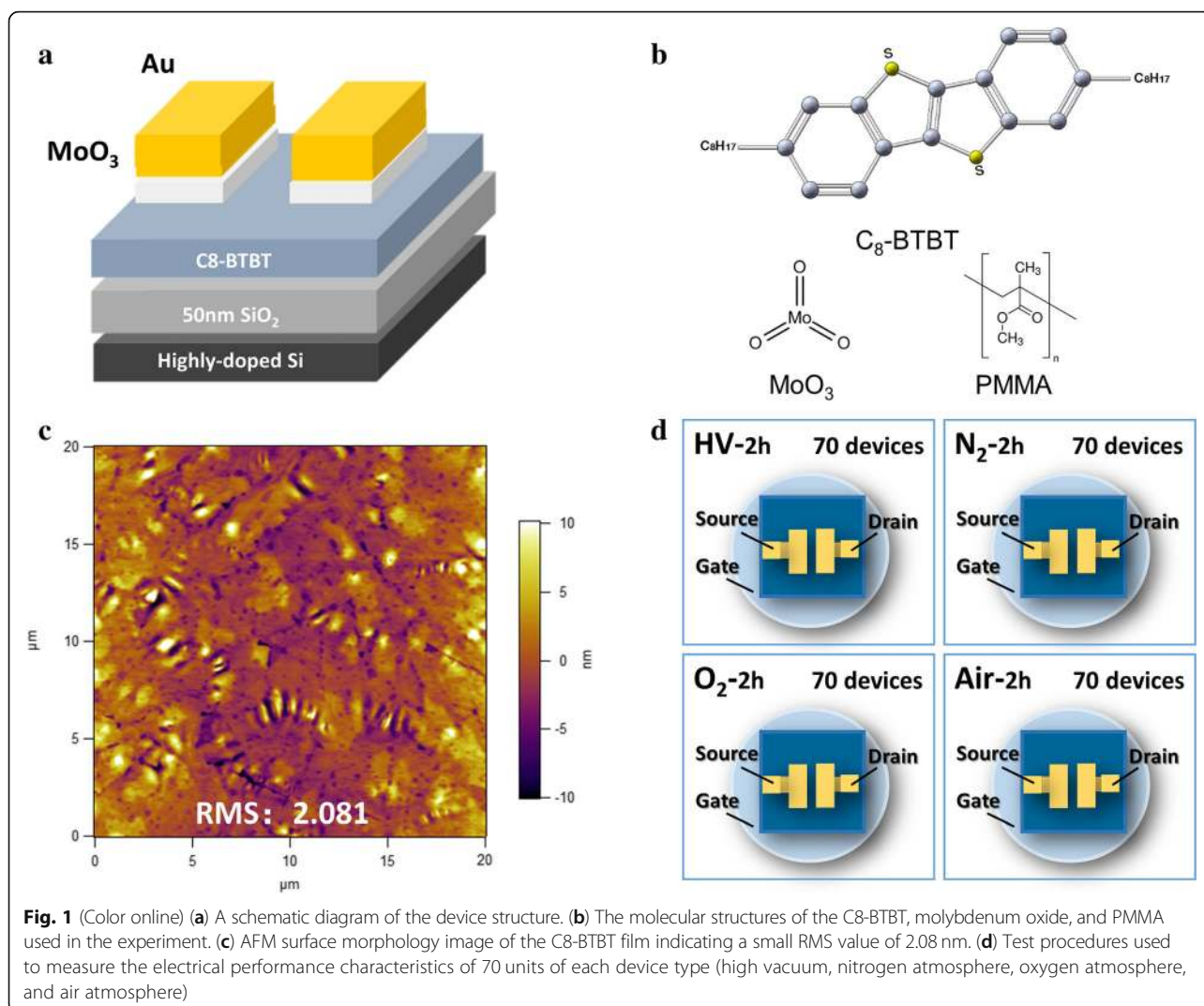
Before their electrical performance measurements, the devices were stored in specific environmental conditions (high vacuum, N<sub>2</sub>, O<sub>2</sub>, air) for 2 h so that they would be fully exposed to the desired gases. For convenience, the devices exposed to high vacuum ( $1.3 \times 10^{-5}$  Torr), N<sub>2</sub>, O<sub>2</sub>, and air will be referred to as the HV, N<sub>2</sub>, O<sub>2</sub>, and air devices, respectively. For each environmental condition or

ambient gas, 70 devices were measured in order to produce reliable and statistically meaningful electrical performance results. In addition, the electrical performance of one sample was monitored as a function of the air exposure time to study its stability in air.

## Results and Discussion

The cross-sectional structure of the OTFT device is shown schematically in Fig. 1a. From bottom to top, it consists of a highly doped Si substrate, 50 nm of silicon oxide, 45 nm of C8-BTBT film, and Au(40 nm)/MoO<sub>3</sub>(5 nm) electrodes. Au/MoO<sub>3</sub> source/drain electrodes were used to reduce the contact barrier between the Au electrodes and C8-BTBT, which can help to increase the charge injection efficiency and produce high-mobility devices [10]. Figure 1b shows the molecular structures of C8-BTBT, MoO<sub>3</sub>, and PMMA. It should be noticed that PMMA was added into C8-BTBT to make a mixed solution in our work. Blending a polymer into a small molecule organic semiconductor is a common method to improve electrical performances of an organic semiconductor. It helps to form a smooth, continuous semiconductor film. In addition, differences in mass induce vertical phase separation, which is expected to reduce the number of surface traps in the semiconductor [19]. An AFM surface morphology image of the C8-BTBT thin film is shown in Fig. 1c. It indicates large grain-size, good surface continuity, and a smooth surface morphology (RMS value 2.081 nm). Figure 1d shows schematic diagrams of the test procedures used with samples that had been exposed to HV, nitrogen, oxygen, and air. For each ambient gas, 70 devices were measured after 2 h of exposure.

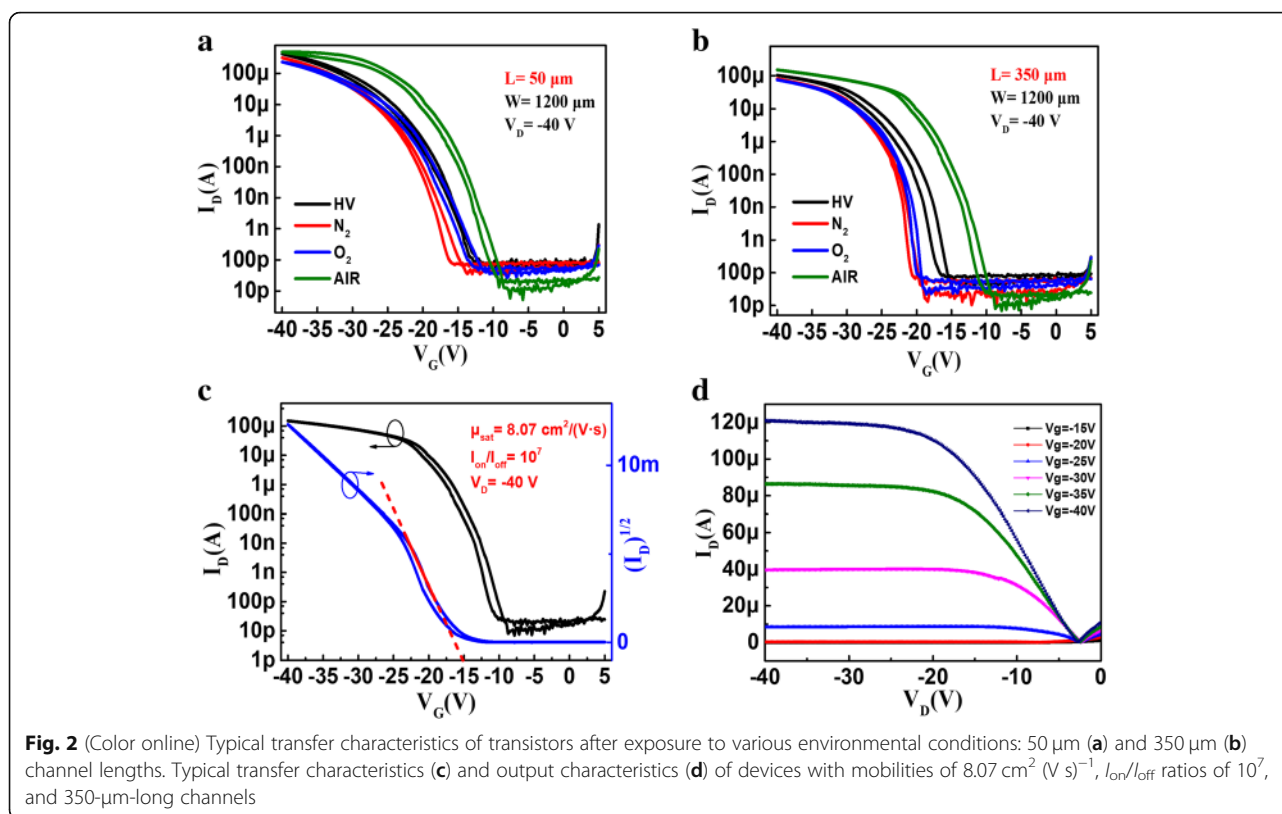
To clarify how the different ambient gases affect device electrical performance, the transfer characteristics of these four device types were compared. Figure 2a and 2b show typical drain current-gate voltage ( $I_D$ - $V_G$ ) curves of short channel ( $L = 50 \mu\text{m}$ ) and long channel ( $L = 350 \mu\text{m}$ ) devices, respectively. All of the devices have the same channel width of 1200 μm and were measured using the same -40 V drain voltage. No significant hysteresis loops are observed regardless of gas exposure or channel length. An obvious decrease in the off-state drain current ( $I_{\text{off}}$ ) and increase in the on-state drain current ( $I_{\text{on}}$ ) are observed for the device exposed to air. Its on/off drain current ratio is as high as 10<sup>7</sup>, while those of HV devices, O<sub>2</sub> devices, and N<sub>2</sub> devices are 10<sup>6</sup>. In addition, the air device exhibits carrier mobility that is almost twice as high as those of the other devices and a  $V_{\text{TH}}$  that is 5 to 8 V lower. The results shown in Fig. 2a and 2b demonstrate that the device exposed to air for 2 h exhibits better electrical properties than those exposed to other ambient gases. Typical transfer ( $V_D = -40 \text{ V}$ ) and output characteristics of air devices with a channel length of 350 μm are



shown in Fig. 2c and 2d, respectively. These figures show the outstanding electrical performance characteristics of the solution processed C8-BTBT transistors. A well-saturated  $I_D$ - $V_G$  curve, large  $I_{on}/I_{off}$  of  $10^7$ , and high carrier mobility of  $8.07 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  are observed. The small hysteresis loop shown in Fig. 2c indicates that an imperfect interface is present between the C8-BTBT and  $\text{SiO}_2$ . The non-linear  $I_D$ - $V_D$  curves at low drain voltage shown in Fig. 2d indicate that the potential barrier at the contact interface is still not low enough for ohmic conduction, despite the use of a  $\text{MoO}_3$  layer to reduce the interfacial barrier between the S/D electrodes and semiconductor. The electrical performance of the air device can be further improved via future interface optimization.

In order to get reliable and statistical data, we measured a total of 280 devices (70 devices for each environmental condition). The carrier mobility and threshold voltage experimental results are summarized and plotted as histograms in Fig. 3a and 3b. In addition, the

average carrier mobilities, highest carrier mobilities, and average threshold voltages of devices exposed to various ambient gases are shown in Table 1. The highest average carrier mobility ( $4.82 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ ) and lowest threshold voltage ( $-20.16 \text{ V}$ ) are observed with devices exposed to air. Thus, air-exposed devices exhibit the best electrical performances of the device types tested. The HV device,  $\text{N}_2$  device, and  $\text{O}_2$  device histograms indicate only slight differences in average carrier mobility, highest carrier mobility, and threshold voltage. It is known that air is composed of nitrogen (78%), oxygen (21%), moisture, etc. The HV,  $\text{N}_2$ , and  $\text{O}_2$  devices exhibit similar electrical characteristics, which indicates that exposure to  $\text{N}_2$  and  $\text{O}_2$  does not produce significant performance differences relative to an HV device. One can assume that moisture plays a key role in improving the electrical performance of the air device. The relative humidity range during these experiments was 40–59%. Consequently, it is likely that  $\text{H}_2\text{O}$  in the air affects device performance.



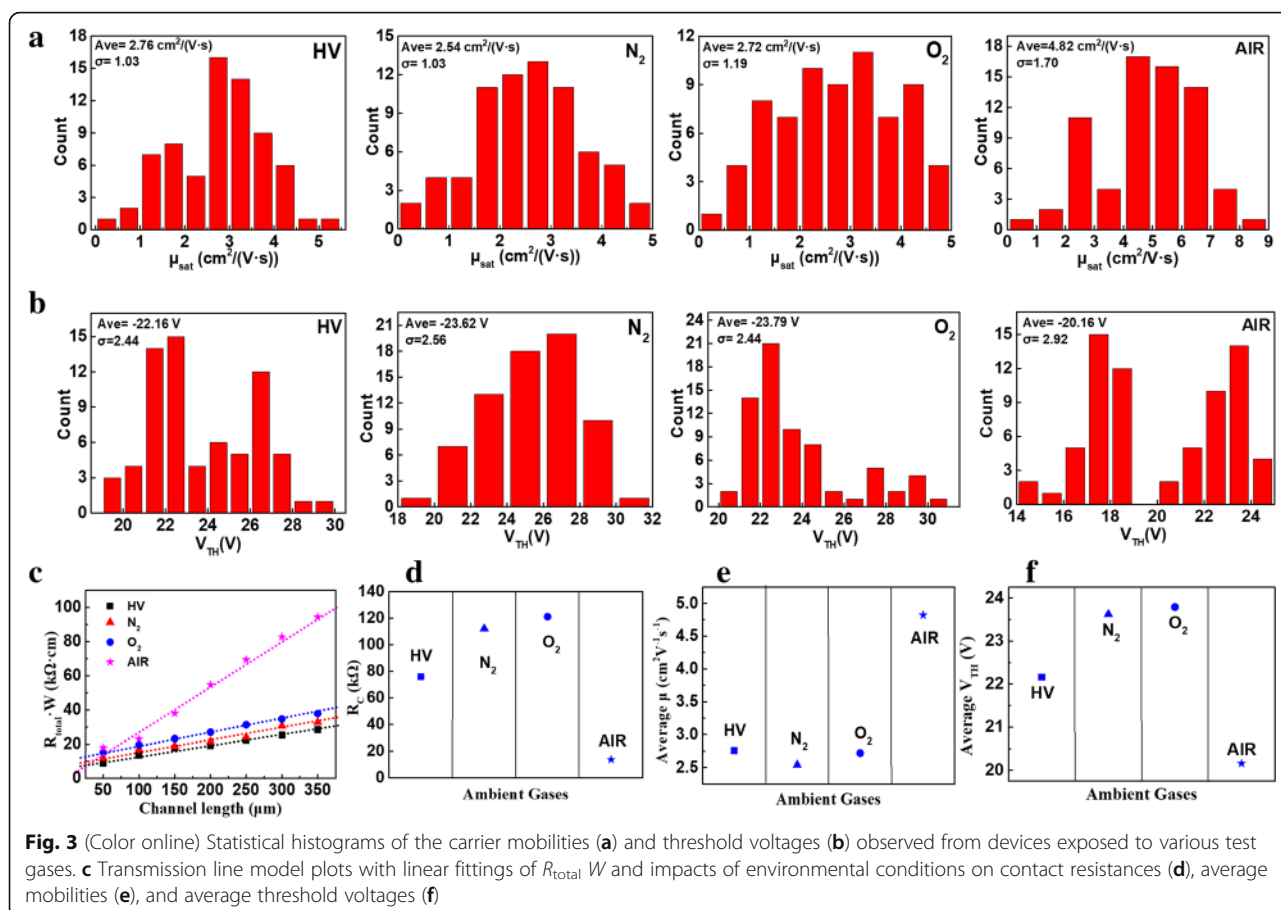
In order to understand gas exposure-based variation in the electrical properties of these C8-BTBT-based transistors, we measured  $I_D$ - $V_G$  curves of devices with channel lengths of 50 to 350  $\mu\text{m}$ . Metal/semiconductor contact resistances ( $R_C$ ) were investigated for all four device types. We performed  $R_C$  extraction using the transfer line method, which is based on the following linear regime equation (1): [20].

$$R_{\text{total}} = R_{\text{channel}} + R_{\text{contact}} = \frac{L}{WC_i(V_g - V_{\text{th}})\mu_{\text{channel}}} + R_{\text{contact}} \quad (1)$$

Figure 3c shows the total resistances ( $R_{\text{total}}$ ) of devices exposed to different environmental conditions as functions of channel length. The  $R_C$  values are extracted from the  $y$ -intercepts of the fitting lines and plotted by exposure gas.  $R_C$  values are compared in Fig. 3d based on the results shown in Fig. 3c. Only small differences between the HV,  $\text{N}_2$ , and  $\text{O}_2$  devices are noted. However, the air device exhibits a significant reduction in  $R_C$ . The average carrier mobilities and average threshold voltages are summarized in Fig. 3e and 3f, respectively. The air devices exhibit much higher carrier mobilities and lower threshold voltages than their counterparts. The  $R_C$  values, average and highest carrier mobilities, and threshold voltages of the four device types are summarized in Table 1. Based on the

results shown in Fig. 3d–f and Table 1, we can conclude that the improved electrical properties exhibited by the air devices are closely related to the reduced contact resistance between the C8-BTBT semiconductor and source/drain electrodes. Furthermore, the  $\text{N}_2$  and  $\text{O}_2$  device electrical properties do not deviate significantly from each other or those of the HV device. This indicates that the reduced  $R_C$  values that drive increased carrier mobilities and decreased threshold voltages are caused by  $\text{H}_2\text{O}$  in air, rather than  $\text{N}_2$  or  $\text{O}_2$  concentrations. The mechanisms of this interaction are not clear, but we assume that hydronium and hydroxyl anions from  $\text{H}_2\text{O}$  may passivate traps and defects in C8-BTBT semiconductors. Our present results provide further insights into the role of air in reducing contact resistances and improving overall electrical performance.

To further understand the mechanisms that drive differences in device electrical performance, we performed Raman spectra measurements of C8-BTBT films exposed to various environmental conditions. Figure 4a compares the Raman spectra of C8-BTBT films exposed to HV and air. Only the  $1300 \text{ cm}^{-1}$ – $1600 \text{ cm}^{-1}$  spectral range is shown since these peaks are typically associated with C8-BTBT molecules and all of the charge sensitive bands lie in this region. Typically, C8-BTBT molecules orient themselves with the long-axis ( $c$ -axis) direction along the  $\text{SiO}_2/\text{Si}$  substrate. A herringbone arrangement of BTBT core parts appears in



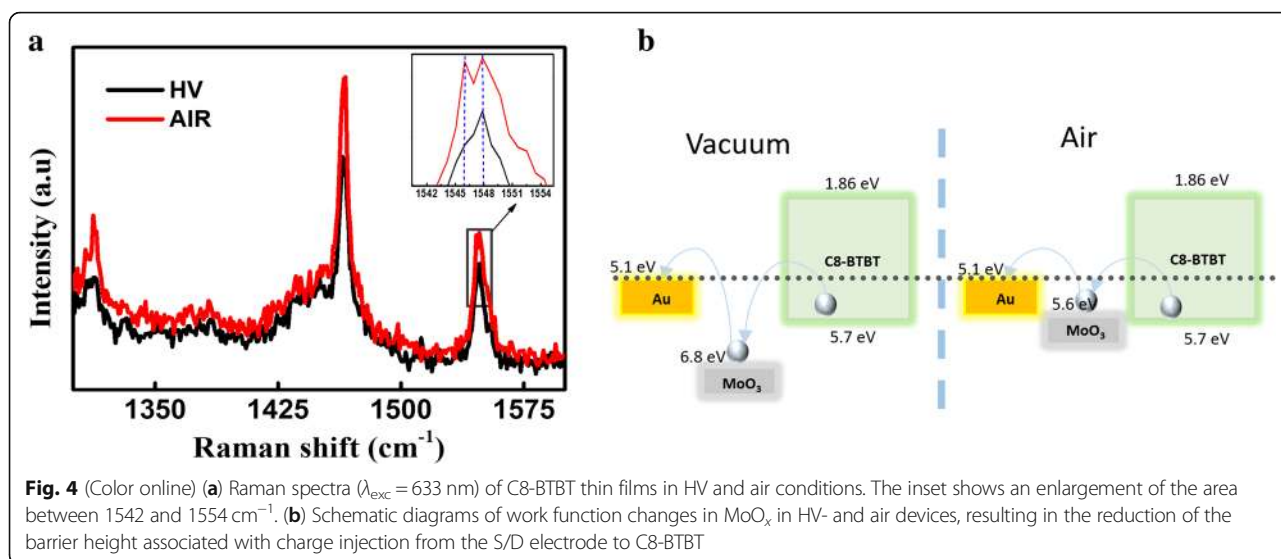
the in-plane direction [14]. Thiophene peaks are located at  $1314\text{ cm}^{-1}$  and  $1465\text{ cm}^{-1}$ , while the C–H in-plane peak appears at  $1547\text{ cm}^{-1}$  [6, 21]. The Raman spectra of C8-BTBT samples exposed to HV,  $\text{O}_2$ , and  $\text{N}_2$  do not exhibit significant differences. When the sample is exposed to air for a period of time, it exhibits Davydov splitting at  $1547\text{ cm}^{-1}$  due to interactions between the hydroxyl anion from water and hydrogen from C–H groups. [22] The C–H bond from stacking of C8-BTBT molecules is typically suspended on the surface [14]. Thus, it can easily interact with moisture in the air and increase the carrier mobility via enhanced  $\pi$ - $\pi$  and van der Waals interactions [5, 9]. This result provides further support for our previous assumption that hydroxyl anions passivate traps in the C8-BTBT films.

**Table 1** Average and highest carrier mobilities, average threshold voltages, and  $R_C$  values of devices exposed to various gases

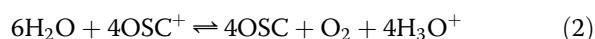
	HV	$\text{N}_2$	$\text{O}_2$	Air
Ave. mobility ( $\text{cm}^2/\text{V s}$ )	2.76	2.54	2.72	4.82
Highest mobility ( $\text{cm}^2/\text{V s}$ )	4.70	4.84	4.88	8.07
Ave. threshold voltage (V)	-22.16	-23.62	-23.79	-20.16
$R_C$ (k $\Omega$ )	76	112.08	121.08	13.75

As Irfan et al. reported [23], the work function ( $W_F$ ) of the thermally evaporated 5.5 nm  $\text{MoO}_x$  is 6.82 eV. However, this decreases by 1.18 to 5.64 eV after 1 h of air exposure. The reduction in  $W_F$  upon air exposure may be due to moisture adsorption on the film surface. Based on the results shown by Irfan et al., we proposed a model that describes the effect of air exposure on C8-BTBT contact resistance and electrical performance (Fig. 4b) [9, 19, 23]. It is assumed that reducing the height of the contact barrier between the metal and the semiconductor would improve the carrier injection efficiency, reduce the contact resistance, and increase the carrier mobility. Another possible mechanism of  $R_C$  reduction is passivation of traps in the interface between C8-BTBT and the Au/ $\text{MoO}_3$  electrode. According to Wang et al., the metal/semiconductor interfacial trap density significantly affects the interfacial contact resistance [24]. In the present work, hydronium from water passivates interfacial traps, producing an  $R_C$  reduction.

Finally, the air stability of the C8-BTBT OTFTs was investigated. We measured the electrical properties of C8-BTBT devices that had been exposed to air for up to 9120 min (~ 1 week). Figure 5a compares  $I_D$ - $V_G$  characteristics of devices with air exposure times of 0 min, 2 h, and 9120 min. The carrier mobility is shown as a function



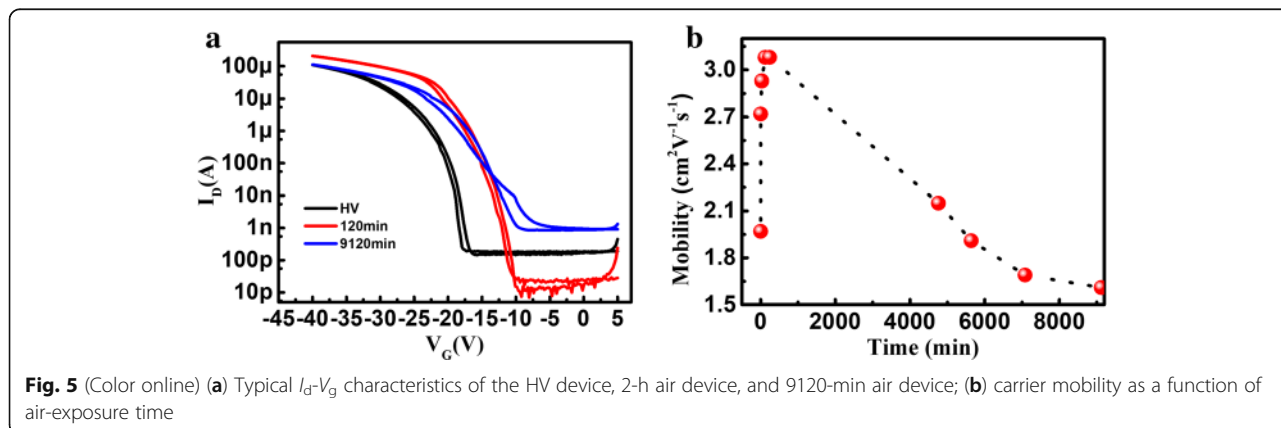
of air exposure duration in Fig. 5b. The carrier mobility of a non-air exposed device is  $1.97 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ . The mobility increases with the air exposure duration until this duration reaches 4 h. The highest carrier mobility ( $3.08 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ ) is achieved after an air exposure time of 2 to 4 h. Further monitoring of the carrier mobility shows that it decreases gradually with additional air exposure. The carrier mobility decreases to  $1.61 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  after the device has been exposed to air for 9120 mins (approximately 1 week). This carrier mobility degradation may occur because the channel is readily oxidized by moisture as shown below in Eq. (2) [25]. In this equation, OSC and OSC+ represent the organic semiconductor and molecular cation, respectively.



After a period of air exposure, moisture adsorption induces unoccupied states above the HOMO and generates deep hole traps, which significantly degrade carrier

transport in the channel and increase the contact resistance [24]. Gomes et al. and Peter et al. have demonstrated that water on the surface of  $\text{SiO}_2$  plays an important role in p-type OTFTs. Due to the  $\text{Si-O-H} \leftrightarrow \text{Si-O}^- + \text{H}^+$  reaction, a significant amount of hydronium is present in the absorbed water layer [26]. In addition, mobile charges in the semiconductor are slowly replaced by immobile charges at the  $\text{SiO}_2$  surface that can reversibly migrate into bulk  $\text{SiO}_2$ . Therefore, exposure to air for a long time, constant absorption and interaction of moisture will lead to increased transistor instability [27] and reduce its carrier mobility.

Using a comparative study of devices exposed to various gas environments, we demonstrated that moisture in the air has a significant impact on the electrical performance characteristics of C8-BTBT-OTFT devices. We also found that an appropriate air-exposure time can improve device electrical performance but a long exposure time degrades it. It is widely believed that exposing organic devices to air is harmful to their



electrical properties. The present work also demonstrates the positive role of moisture in passivating C8-BTBT semiconductor traps and lowering  $R_C$  values. It also provides useful insights into the ideas that may improve C8-BTBT OTFT device performance and improve knowledge of their air stability.

## Conclusions

In summary, we have investigated the effects of ambient gases on the electrical properties of solution-processed C8-BTBT OTFTs. The electrical properties of devices exposed to various ambient gases (HV, O<sub>2</sub>, N<sub>2</sub>, and air) were compared. We observed that the electrical properties of the O<sub>2</sub> device and N<sub>2</sub> device varied little relative to the HV device. However, a significant improvement in electrical properties was observed with the air device. For the 70 devices with 2 h of air exposure, the average and highest carrier mobilities were 4.82 and 8.07 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup>, respectively. This compares to 2.76 and 4.70 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup> for HV devices. The lowest threshold voltages were also observed using the air devices. The improved electrical performance of the air device is thought to be due to reduced contact resistance and decreased MoO<sub>3</sub> work function after air exposure. In addition, C8-BTBT OTFT air-stability was investigated. The electrical performance degraded upon exposure to air for more than 4 h. This work provides a systematic understanding of the influence of environmental conditions on the electrical performance characteristics of solution processed C8-BTBT OTFTs. It aids in the development of high performance, air-stable, printable OTFT devices.

## Abbreviations

AFM: Atomic force microscopy; Au: Gold; C8-BTBT: 2,7-dioctyl [1] benzothieno[3,2-b][1]benzothiophene; HOMO: Highest occupied molecular orbital; HV: High vacuum;  $I_D$ : Drain current; L: Channel length; MoO<sub>3</sub>: Molybdenum oxide; OTFTs/OTFT: Organic thin-film transistors; PMMA: Polymethyl methacrylate;  $R_C$ : Contact resistance; RMS: Root mean square;  $R_{\text{total}}$ : Total resistances; TFTs: Thin-film transistors;  $V_G$ : Gate voltage; W: Channel width

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## Authors' Contributions

JYM performed the experiments and drafted the manuscript. XBL directed the study and revised the manuscript. NWT and WEH helped with the data analysis and Raman measurements. ZMZ, CLL, AHZ, ZF, SJW, MZ, JWG, GFZ, and JML provided constructive advice during drafting. All authors read and approved the final manuscript.

## Competing Interests

The authors declare that they have no competing interests.

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