香港浸會大學
HONG KONG BAPTIST UNIVERSITY

HKBU Scholars

# Using thin film transistors to quantify carrier transport properties of amorphous organic semiconductors

Cheung, C. H.; Tsung, K. K.; Kwok, K. C.; So, Shu Kong

*Published in:*
Applied Physics Letters

*DOI:*
10.1063/1.2972125

Published: 01/08/2008

*Document Version:*
Publisher's PDF, also known as Version of record

Link to publication

*Citation for published version (APA):*
Cheung, C. H., Tsung, K. K., Kwok, K. C., & So, S. K. (2008). Using thin film transistors to quantify carrier transport properties of amorphous organic semiconductors. *Applied Physics Letters*, *93*(8), Article 083307. https://doi.org/10.1063/1.2972125

**General rights**
Copyright and intellectual property rights for the publications made accessible in HKBU Scholars are retained by the authors and/or other copyright owners. In addition to the restrictions prescribed by the Copyright Ordinance of Hong Kong, all users and readers must also observe the following terms of use:

- Users may download and print one copy of any publication from HKBU Scholars for the purpose of private study or research
- Users cannot further distribute the material or use it for any profit-making activity or commercial gain
- To share publications in HKBU Scholars with others, users are welcome to freely distribute the permanent publication URLs

Downloaded on: 11 Oct, 2024

# Using thin film transistors to quantify carrier transport properties of amorphous organic semiconductors

C. H. Cheung, K. K. Tsung, K. C. Kwok, and S. K. So[a)]
*Department of Physics and Centre for Advanced Luminescence Materials, Hong Kong Baptist University, Kowloon Tong, Hong Kong, People's Republic of China*

(Received 11 April 2008; accepted 19 July 2008; published online 27 August 2008)

The hole transport properties of two phenylamine-based compounds were evaluated by thin film transistor (TFT) measurement and time-of-flight (TOF) technique. The compounds were $N,N'$-diphenyl-$N,N'$-bis(1-naphthyl) (1,1′biphenyl)-4,4′diamine (NPB) and 4,4′,4″-tris[n-(2-naphthyl)-n-phenyl-amino] triphenylamine (2TNATA). With tungsten oxide/gold as the charge injecting electrode, the field effect mobility of NPB was found to be $2.4\times10^{-5}$ cm$^2$/V s at room temperature, which was about one order of magnitude smaller than that obtained from independent TOF experiments ($3\times10^{-4}$ cm$^2$/V s). Similar observations were found for 2TNATA. Temperature dependent measurements were carried out to study the energetic disorder of the materials. It was found that the energetic disorder was increased in the neighborhood of a gate dielectric layer. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2972125]

The measurement of charge carrier mobility is an important step towards understanding the conduction mechanism of organic semiconductors.[1,2] Various techniques for evaluating carrier mobility of organic semiconductors have been reported. Examples are time-of-flight (TOF),[3] dark-injection space-charge-limited current,[4] admittance spectroscopy,[5] and thin film transistor (TFT).[6] Among them, TOF is, perhaps, the most popular.[3] In TOF, an UV laser is used to excite a material and generate free charge carriers for drift mobility measurements. Due to the finite penetration depth of the laser, TOF generally requires a thick film ($\sim\mu$m). In contrast, TFT technique requires much thinner film of thickness $\sim$0.1 $\mu$m or less. Carrier mobility, generally called field effect (FE) mobility ($\mu_{FE}$), can be extracted from the output characteristics of a TFT. The mobility in the linear and the saturation regions can be calculated from Eqs. (1) and (2), respectively,[7]

$$I_D = \frac{W}{L}\mu_{FE}C_i\left[V_D(V_G - V_T) - \frac{V_D^2}{2}\right], \quad (1)$$

$$I_D = \frac{W}{2L}\mu_{FE}C_i(V_G - V_T)^2. \quad (2)$$

$I_D$ is the current from the source (S) to drain (D). $W$ and $L$ are the channel width and length, respectively; $C_i$ is the capacitance of gate dielectric per unit area; $V_T$ is the threshold voltage; $V_G$ and $V_D$ are the gate bias and the potential difference across S and D, respectively; $\mu_{FE}$ is the FE mobility.

In this contribution, we compare the two carrier mobility evaluation techniques, namely, TOF and TFT. The materials under investigation are $N,N'$-diphenyl-$N,N'$-bis(1-naphthyl)(1,1′biphenyl)-4,4′diamine (NPB) and 4,4′,4″-tris[n-(2-naphthyl)-n-phenyl-amino] triphenylamine (2TNATA). Both compounds are phenylamines (PA), which are amorphous hole transporting (HT) materials widely used in organic light-emitting diodes.[8] The PA compounds possess trap-free HT properties, so they are ideal candidates for examining charge transporting mechanism in amorphous organic semiconductors.[2,9]

Details of TOF experiments have been reported elsewhere.[2,9] For TFT, a heavily doped $p$-Si wafer with an

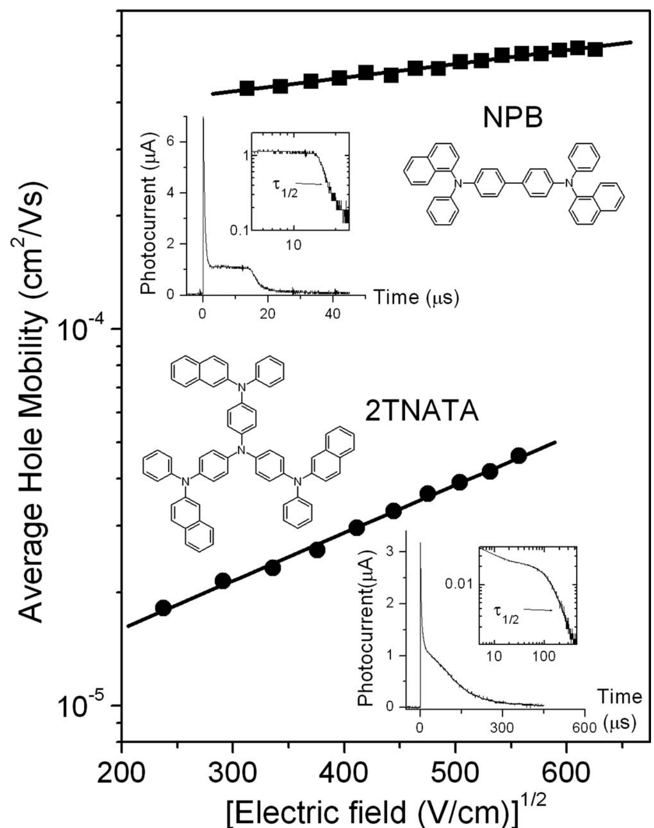


FIG. 1. The average hole mobilities of NPB (squares) and 2TNATA (circles) against the square root of electric field at 290 K derived from TOF. The insets are the TOF time transients and their log-log plots under an applied electric field strength of 140 kV/cm. The average hole transit time can be extracted from $\tau_{1/2}$, the instant when the TOF signal is half of its value at the turning point of the TOF transient. The chemical structures of NPB and 2TNATA are also shown here. The thicknesses of the organic films were 7 $\mu$m.

a)Author to whom correspondence should be addressed. Electronic mail: skso@hkbu.edu.hk.

10 October 2023 02:04:24

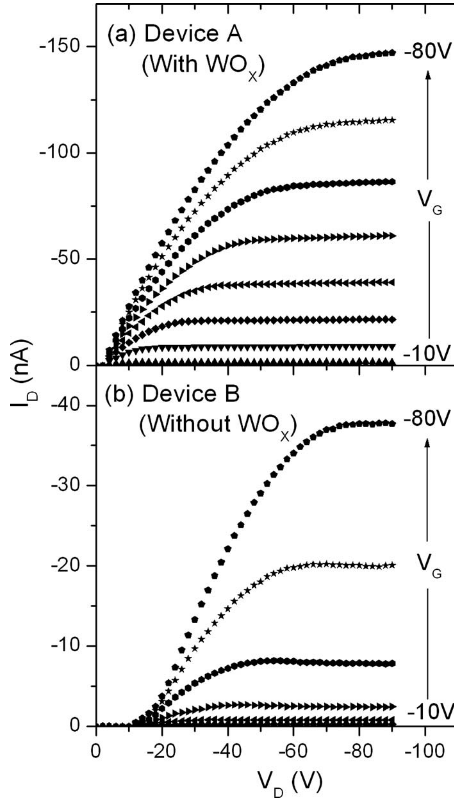


FIG. 2. Typical output characteristics of top contact OTFTs with 100 nm of NPB at 290 K. The gate voltages varied in steps of −10 V, starting from −10 V. In (a), 20 nm of tungsten oxide was inserted between NPB and the top (source and drain) gold contacts. In (b), gold (source and drain) electrodes were directly in contact with NPB. The channel length and width are 50 $\mu$m and 9 mm, respectively.

overlayer of $SiO_2$ (300 nm, $C_i = 11$ nF cm$^{-1}$) was used as the substrate. It was held at 60 °C during organic film deposition. The thickness of the organic film was 100 nm. For some NPB-based organic TFTs (OTFTs), 20 nm of tungsten oxide ($WO_X$, $x \sim 2-3$) was thermally evaporated through a shadow mask on NPB before the deposition of gold S/D electrodes.[10–12] Measurements were performed in vacuum ($\sim 10^{-3}$ Torr). The sample temperature was regulated between 235 and 360 K.

Figure 1 (insets) show the TOF transients of NPB and 2TNATA at room temperature. The average hole transit times ($\tau_{1/2}$) can be determined from log-log plots of the transients. Figure 1 also shows the field dependent hole mobilities ($\mu_h$) of NPB and 2TNATA.

Next, we use TFT samples to evaluate carrier mobility. For the case of NPB-based OTFTs, the output characteristics of device A (with $WO_X$) and device B (without $WO_X$) are shown in Fig. 2. Device A shows well-defined linear and saturation regions when compared to device B, especially when the $V_G$ is small. From Eqs. (1) and (2), the FE mobilities for device A are $1.7 \times 10^{-5}$ and $2.4 \times 10^{-5}$ cm$^2$/V s in linear and saturation regions, respectively. On the other hand, without a well-defined linear region, FE mobility can only be extracted from saturation region for device B, and is found to be $2.7 \times 10^{-5}$ cm$^2$/V s. The large differences in the shapes of output characteristics suggest that the introduction of $WO_X$ can effectively reduce the contact resistance at the Au/NPB interface.[10–12]

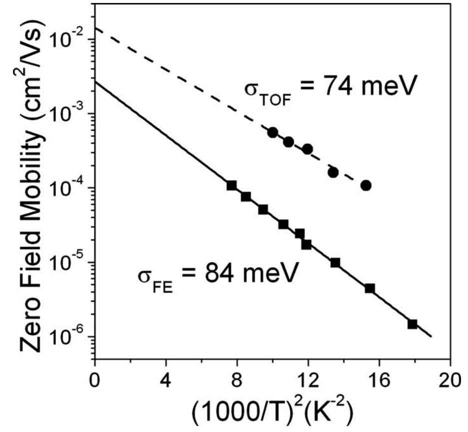


FIG. 3. Low field FE mobilities from TFT, and zero-field mobilities (obtained from $\tau_{1/2}$) from TOF against $1/T^2$ for NPB. The solid and dashed lines are the best fits to TFT and TOF data, respectively.

Our results indicate that the FE mobility is about one order of magnitude smaller than the TOF mobility. Similar results were reported for other amorphous organic materials recently.[1,6,13] In the TFT configuration, injected holes from the source electrode are accumulated and transport in the organic layer adjacent to the gate dielectric under the influence of $V_G$ and $V_D$. Therefore, morphological or other changes in the organic layer induced by the gate dielectric should affect the carrier mobility. Veres *et al.* proposed that there is an increase in the energetic disorder of an organic material in the neighborhood of a polar gate dielecitic.[13] The randomly oriented polar groups of the dielectric result in an additional fluctuation of local electrostatic field. This broadens the density of states (DOS) of the organic material, resulting in an increase in the energetic disorder and a decrease in the FE mobility.

To verify if, indeed, the energetic disorder is modified, we performed temperature dependent measurements on the OTFTs. The temperature dependent FE mobilities were then analyzed by the gaussian disorder model (GDM).[14] The GDM can be described as follows:[14]

$$\mu(F,T) = \mu_\infty \exp\left[-\left(\frac{2\sigma}{3kT}\right)^2\right] \times \exp\left\{CF^{1/2}\left[\left(\frac{\sigma}{kT}\right)^2 - \Sigma^2\right]\right\}, \quad (3)$$

In Eq. (3), $F$, $T$, and $k$ are the applied electric field, the absolute temperature and the Boltzmann constant; $\mu_\infty$ is the high temperature limit of mobility, and $C$ is a constant. The energetic disorder ($\sigma$) and positional disorder ($\Sigma$) can be interpreted as the width of Gaussian distribution of the potential energy and position for the transport sites, respectively. At low field ($F \to 0$), the second exponential term in Eq. (3) approaches 1, and thus, $\mu(0,T) \sim \exp[-4/9(\sigma/kT)^2]$. So, a semilog plot of low field mobilities versus $1/T^2$ should yield a straight line. The slope of this line can be used to extract $\sigma$. Figure 3 shows the FE mobilities extracted in low field region [linear region in Fig. 2(a)] plotted against $1/T^2$. The results are shown as solid squares. The extracted $\sigma$ for NPB in an OTFT is 84 meV. On the other hand, $\sigma$ can also be extracted solely from the TOF data by the same method. In this case, we extracted the zero-field mobilities at different temperatures and plotted them against $1/T^2$. The results are

10 October 2023 02:04:24

TABLE I. Transport parameters of NPB and 2TNATA evaluated by TFT and TOF techniques.

| Material | Technique | Hole mobility ($cm^2 V^{-1} s^{-1}$) | TFT threshold voltage (V) | Energetic disorder (meV) | $\mu_\infty$ ($cm^2 V^{-1} s^{-1}$) |
|---|---|---|---|---|---|
| NPB | TFT with $WO_x$ | $2.4\times10^{-5}$ (saturation) $1.7\times10^{-5}$ (linear) | −0.7 | 84 | $2.6\times10^{-3}$ |
| NPB | TFT without $WO_x$ | $2.7\times10^{-5}$ (saturation) | −40.7 | ⋯ | ⋯ |
| NPB | TOF | $3.0\times10^{-4}$ ($F=12$ kV/cm) | ⋯ | 74 | $1.4\times10^{-2}$ |
| 2TNATA | TFT[a] | $9.8\times10^{-7}$ (saturation) | 25.2 | 79 | $7.3\times10^{-5}$ |
| 2TNATA | TOF | $1.2\times10^{-5}$ ($F=12$ kV/cm) | ⋯ | 70 | $1.2\times10^{-3}$ |

[a]Bottom contact configuration was used in 2TNATA OTFTs. Only FE mobility in saturation region can be extracted as the linear region was not well defined.

shown as solid circles in Fig. 3. The extracted $\sigma$ for NPB from TOF is 74 meV, which is in good agreement with previous results.[2,6] The higher energetic disorder from TFT concurs with Ref. 13 In the present work, the polar Si–O moieties of the $SiO_2$ gate dielectric are responsible for the dipolar contribution to the energetic disorder. Due to the amorphous nature of thermally grown $SiO_2$, the polar Si–O moieties are randomly oriented, which results in an additional fluctuation of local electrostatic field and an increase in the energetic disorder of the organic material.

Besides NPB, analogous experiments were performed on 2TNATA. The results are summarized in Table I. An order of magnitude difference in mobility is again observed from 2TNATA. In this case, the extracted energetic disorder from TFT is larger than that from TOF by about ~10 meV. The results from 2TNATA are consistent to those from NPB.

The amount of energetic disorder introduced by the gate dielectric can be extracted from the experimental results. From TFT technique, the energetic disorder ($\sigma_{TFT}$) is contributed by: (i) the intrinsic energetic disorder of the material ($\sigma_{TOF}$); and (ii) the broadening of the DOS due to the polar gate dielectric ($\sigma_{dielectric}$). The relation between the measured energetic disorder and the contributions can be expressed as[15]

$$\sigma_{TFT}^2 = \sigma_{TOF}^2 + \sigma_{dielectric}^2. \tag{4}$$

From Eq. (4), $\sigma_{dielectric}$ are 40 and 37 meV for NPB and 2TNATA, respectively.

Theoretically, an order of magnitude estimation for $\sigma_{dielectric}$ can be computed as follows. According to Dieckmann *et al.*, randomly oriented dipoles increases the energetic disorder by an amount $\sigma_d$,[16]

$$\sigma_d = \frac{3.06c^{2/3}p}{a^2\varepsilon_r} \quad (\text{eV}). \tag{5}$$

Here, $c$ is the fractional concentration of dipoles and $p$ is the dipole moment (in Debye). Together $c$ and $p$ represent the strength of the dipole perturbation. $\varepsilon_r$ is the dielectric constant of the hopping medium, and $a$ is the charge hopping distance (in angstrom). For NPB, $\varepsilon_r \approx 3$. $a \sim 10$ Å is approximately the intersite distance between two NPB molecules.[17] In addition, $c=1$ as all sites are occupied by Si–O within the gate dielectric. To find $p$, we note that the Si–O bond has a bond length of 1.6 Å, and according to Pauling, the O atom has a fractional charge of −0.5e;[18] their product gives an effective dipole moment, $p=3.84$ D. Using Eq. (5), we obtain $\sigma_d \sim 39$ meV, which is in good agreement with the extracted $\sigma_{dielectric}$ from our data analysis.

Apart from $\sigma$, Eq. (3) can be used to extract $\mu_\infty$, the high temperature limit of the carrier mobility. The results are shown in Table I. Both NPB and 2TNATA show smaller $\mu_\infty$ from TFT when compared to TOF. The mismatch of $\mu_\infty$ suggests that the energetic disorder of the organic material is indeed modified by the gate dielectric, but it is unlikely to be the only interface mechanism affecting the FE mobility. As discussed before, any morphological or other changes of the active material induced by the gate dielectric surface can also be the factors.

In conclusion, TOF and TFT configuration were used to evaluate the transport parameters of two PA compounds. The FE mobilities were found to be one order of magnitude smaller than the TOF mobilities. We found that the presence of the $SiO_2$ gate dielectric increases $\sigma$ but reduces $\mu_\infty$. With small material consumption, TFT configuration can be used as an alternative for measuring charge transport parameters of amorphous organic materials.

Support of this research by the Research Grant Council of Hong Kong under Grant Nos. HKBU211006E and HKBU211107E are gratefully acknowledged.

[1]Y. Shirota and H. Kageyama, Chem. Rev. (Washington, D.C.) **107**, 953 (2007).
[2]K. K. Tsung and S. K. So, Appl. Phys. Lett. **92**, 103315 (2008).
[3]P. M. Borsenberger and D. S. Weiss, *Organic Photoreceptors for Imaging Systems* (New York, Marcel Dekker, 1993), Chap. 9.
[4]S. C. Tse, S. W. Tsang, and S. K. So, J. Appl. Phys. **100**, 063708 (2006).
[5]S. W. Tsang, S. K. So, and J. B. Xu, J. Appl. Phys. **99**, 013706 (2006).
[6]T. P. I. Saragi, T. F. Lieker, and J. Salbeck, Adv. Funct. Mater. **16**, 966 (2006).
[7]S. M. Sze, *Physics of Semiconductor Devices* (Wiley, New York, 1981).
[8]C. Adachi, K. Nagai, and N. Tamoto, Appl. Phys. Lett. **66**, 2679 (1995).
[9]C. H. Cheung, K. C. Kwok, S. C. Tse, and S. K. So, J. Appl. Phys. **103**, 093705 (2006).
[10]C. W. Chu, S. H. Li, C. W. Chen, V. Shrotriya, and Y. Yang, Appl. Phys. Lett. **87**, 193508 (2005).
[11]H. You, Y. Dai, Z. Zhang, and D. Ma, J. Appl. Phys. **101**, 026105 (2007).
[12]T. Matsushima, Y. Kinoshita, and H. Murata, Appl. Phys. Lett. **91**, 253504 (2007).
[13]J. Veres, S. D. Ogier, S. W. Leeming, and D. C. Cupertino, Adv. Funct. Mater. **13**, 199 (2003).
[14]H. Bässler, Phys. Status Solidi B **175**, 15 (1993).
[15]P. M. Borsenberger and H. Bässler, J. Chem. Phys. **95**, 5327 (1991).
[16]A. Dieckmann, H. Bässler, and P. M. Borsenberger, J. Chem. Phys. **99**, 8136 (1993).
[17]K. Imaizumi, K. Kaneko, T. Mori, and T. Mizutani, Jpn. J. Appl. Phys., Part 1 **41**, 366 (2002).
[18]L. Pauling, Am. Mineral. **65**, 321 (1980).

10 October 2023 02:04:24