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Chiho Katagiri, Tsukasa Yoshida, Matthew Schuette White, Cigdem Yumusak, Niyazi Serdar Sariciftci, and Ken-ichi Nakayama

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Application of MIS-CELIV technique to measure hole mobility of hole-transport material for organic light-emitting diodes

Chiho Katagiri,1,2 Tsukasa Yoshida,1 Matthew Schuette White,3 Cigdem Yumusak,4 Niyazi Serdar Sariciftci,4 and Ken-ichi Nakayama1,2,a

1Department of Organic Materials Engineering, Graduate School of Science and Engineering, Yamagata University, 4-3-16 Jonan, Yonezawa, Yamagata 992-8510, Japan
2Department of Material and Life Science, Graduate School of Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan
3Department of Physics, University of Vermont, Burlington, Vermont 05405, USA
4Linz Institute for Organic Solar Cells (LIOS), Physical Chemistry, Johannes Kepler University Linz, Altenbergerstraße 69, Linz 4040, Austria

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Injection-charge extraction by linearly increasing voltage in metal-insulator-semiconductor structures (MIS-CELIV) is applied for the hole mobility measurement of N,N'-Bis(naphthalen-1-yl)-N,N'-bis(phenyl)-benzidine (NPB), which is a standard hole-transporting material for organic light-emitting diodes. Ideal transient currents in agreement with the theory are observed in the NPB film due to its amorphous and homogenous structure, which is regarded as a continuous dielectric. This ideal response enables us to discuss the validity of the MIS-CELIV mobility by comparing its absolute value with that of the conventional space-charge-limited current method. In addition, to establish an experimental guideline for precise measurements, the effect of the voltage drop on the insulator is investigated. © 2018 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).

Charge-carrier mobility is one of the key parameters for organic light-emitting diodes (OLEDs) and organic photovoltaics (OPVs). Electron and hole mobilities have been investigated using various techniques, such as the time-of-flight (TOF),1,2 current-voltage measurement in the space-charge-limited current region (SCLC),3,4 dark-injection (DI) transient measurement,5,6 impedance spectroscopy (IS),7,8 and charge extraction by linearly increasing voltage (CELIV).9–11 As the device structure and charge transport mode for each measurement are different, the optimum method for OLED and OPV devices must be selected.

The TOF method has been conventionally used for measuring the charge-carrier mobility in organic semiconductors; the electron and hole mobilities are directly estimated from the transport time of the photogenerated charge carriers. However, this method requires a film, with a thickness in the order of 1–10 µm, to ensure sheet-like photogeneration in the proximity of an illuminated electrode. Such films are several times thicker than the organic layers of OLED or OPV devices, and may affect their morphology and carrier mobility.12 In contrast, the SCLC method, which uses the static current density-voltage (J–V) curves, is useful for measuring the carrier mobility of less than 1-µm thick films. However, this method requires perfect ohmic contact and the estimated mobility is considerably affected by the injection barrier. In addition, it is difficult to judge the correctness of the analysis because the J–V curves at times indicate a J ∝ V² relationship, even for suppressed current with the injection barrier.13

aE-mail: nakayama@mls.eng.osaka-u.ac.jp
The CELIV method is applicable for thin organic films with 100–300 nm thicknesses. The mobility is estimated from the current transients by the extraction of the equilibrium or photogenerated charge carriers, for the dark CELIV and photo-CELIV measurements, respectively. The photo-CELIV method is a powerful technique for analyzing the carrier lifetime and bimolecular recombination in OPV devices; however, it is difficult to distinguish between the electron and hole mobilities because both carriers are involved in the measurement. On the other hand, the dark CELIV method is useful for measuring unipolar mobility and the concentration of the thermal-equilibrium charge carriers; however, this method requires sufficient intrinsic carriers, and consequently, is limited to materials such as poly(3-hexylthiophene) in which the equilibrium holes are derived from oxygen and/or water doping.

The injection-CELIV measurement for metal-insulator-semiconductor structures (MIS-CELIV), which can estimate the unipolar charge mobility from the current transients by the extraction of the accumulated charges at the insulator/semiconductor interface, has been recently proposed by Juška et al. Its advantages over the conventional methods are as follows: (a) The abundance of accumulated charges allows clear current transients to be observed in a wide range of materials with high reproducibility, (b) the electron and hole mobilities are measured selectively in thin films with 100–300 nm thicknesses, and (c) the time-domain measurement includes considerable information on the charge transport states, unlike steady-state measurement such as the SCLC method.

The MIS-CELIV method is applicable to a wide range of organic semiconductors because clear current transients can be observed due to the injection and accumulation of sufficient charge carriers by the forward bias voltage. However, the MIS-CELIV transients should be analyzed carefully because the analysis of the transient time is indirect. In the past few years, the MIS-CELIV method has been mainly used for evaluating the mobility balance between holes and electrons in blend films consisting of conjugated polymer and fullerene, for OPVs. However, the validity of the absolute value of the estimated mobility has not been verified. The objective of this paper is to indicate the potential of the MIS-CELIV method as a standard technique for measuring the characteristic mobilities of pure thin films. A hole-transporting material, N, N'-Bis(naphthalen-1-yl)-N, N'-bis(phenyl)-benzidine (NPB), is selected as the organic material because the amorphous, homogeneous film is similar to a continuous dielectric, and suitable for simple and precise analysis. Such mobility measurement in a thin film is effective for accelerating the development of new OLED materials. The validity of the estimated hole mobility is discussed experimentally, in comparison with the mobility evaluated by the SCLC method. To clarify the appropriate measurement conditions for precise measurement, the effect of the insulator thickness is investigated.

Figure 1(a) shows the measurement setup and the MIS device structure, for MIS-CELIV measurement. A heavily n-doped silicon wafer with a thermal-silicon-dioxide (SiO$_2$) layer was used as the substrate. An NPB thin film was deposited by vacuum evaporation on the Si/SiO$_2$ substrate, followed by the deposition of a hole-injection layer of molybdenum trioxide (MoO$_3$) and an Al electrode on the organic layer. For SCLC measurement, an indium-tin oxide (ITO)-coated substrate was used. The structures of sample-1, for the MIS-CELIV measurement and sample-2, for the SCLC, were SiO$_2$(30 nm)/NPB(290 nm)/MoO$_3$(5 nm)/Al and ITO/MoO$_3$(5 nm)/NPB(290 nm)/MoO$_3$(5 nm)/Al,
respectively. The NPB and MoO\textsubscript{3}/Al layers were deposited, under the same conditions. Samples 3–5 for the MIS-CELIV had different SiO\textsubscript{2} thicknesses, and the device structures were SiO\textsubscript{2} (30, 50, and 100 nm)/NPB (300–310 nm)/MoO\textsubscript{3} (5 nm)/Al, respectively. The film thickness was measured by a stylus profiler (Bruker, Dektak XT).

Figure 1(b) displays an illustration of the MIS-CELIV measurement results. When a negative forward voltage, \( V_{FB} \), is applied to the Si substrate, holes are injected from the MoO\textsubscript{3}/Al to the NPB film and accumulated near the SiO\textsubscript{2}/NPB interface. These accumulated holes are extracted by applying a linearly increasing reverse voltage with a voltage rise speed, \( A = dV/dt \). The MIS-CELIV transient consists of the displacement current, \( j_0 \), due to the total geometric capacitance of the insulator and semiconductor, and the current peak, \( \Delta j \), by the saturation current, \( j_{sat} \), due to the extraction of the accumulated carriers. The mobility is determined by
due to a finite insulator layer, is defined as

\[
\mu = \frac{2d_i^2}{Atr_s^2 \left( 1 + \frac{\varepsilon_s d_i}{\varepsilon_i d_s} \right)},
\]

where \( \varepsilon_0 \) is the dielectric permittivity of vacuum, \( \varepsilon_s (\varepsilon_i) \) is the dielectric permittivity of the semiconductor (insulator), and \( d_s (d_i) \) is the thickness of the semiconductor (insulator) layer. The carrier transport time, \( t_{tr} \), is related to the characteristic time, \( t_{2j0} \), for attaining twice the value of \( j_0 \). Juška et al. have shown that the \( t_{tr} \) is defined as

\[
t_{tr} = \frac{4}{\pi} t_{2j0},
\]

when the capacitance of the insulator, \( C_i \), is considerably greater than the capacitance of the semiconductor, \( C_s \) (\( C_i/C_s \gg 1 \)). Sandberg et al. proposed that \( t_{tr} \), considering the applied voltage drop due to a finite insulator layer, is defined as

\[
t_{tr} = \frac{4}{\pi} t_{2j0} \sqrt{\frac{1}{1 + \varepsilon_s d_i/\varepsilon_i d_s}},
\]

which can be applied under the condition where \( C_i \) is greater than or comparable with \( C_s \) (\( C_i/C_s \geq 1 \)). The MIS-CELIV transients were measured using a waveform generator (Agilent, 33511B) and an oscilloscope (Agilent, DSO-X 2004A), under medium vacuum (<10 Pa). The current-voltage characteristics of the SCLC measurement were measured using a semiconductor parameter analyzer (Agilent, 4155C), under nitrogen, using a glove-box system.

Figure 2(a) depicts the MIS-CELIV current transients in sample-1 at \( A = 150 \) kV/s at various forward voltages, \( V_{FB} \). At \( V_{FB} = 0 \) V, the current transient consists of the displacement current, \( j_0 \), alone, derived from the total capacitance of the SiO\textsubscript{2} and NPB layers. The flat response indicates that equilibrium charge carriers do not exist in the NPB film. The observed \( j_0 \) of 12.8 A/m\textsuperscript{2} is in good agreement with the value, 12.7 A/m\textsuperscript{2}, estimated using Eq. (S1) (see supplementary material), which corresponds to the geometric capacity, indicating that the SiO\textsubscript{2} and NPB layers function as an ideal capacitor with assumed permittivity and thickness. When \( V_{FB} = -2 \) to -10 V is applied, holes are injected from the MoO\textsubscript{3}/Al electrode and accumulated at the SiO\textsubscript{2}/NPB interface, resulting in a current peak, \( \Delta j \), due to hole extraction. \( \Delta j \) was increased by increasing \( V_{FB} \), and was further saturated at \( j_{sat} = 172 \) A/m\textsuperscript{2}, at \( V_{FB} = -10 \) V. The transient current in the MIS-CELIV is finally limited by the displacement current of the insulator layer (SiO\textsubscript{2}), as shown in Eq. (S2) (see supplementary material); the saturation of \( \Delta j \) against \( V_{FB} \) is observed, when sufficient holes are supplied from the insulator interface such that the displacement current is not limited for the linearly increasing voltage. Therefore, the result indicates that sufficient holes have been accumulated at the SiO\textsubscript{2}/NPB interface at \( V_{FB} = -10 \) V. The observed \( j_{sat} \) is in agreement with the theoretically predicted \( j_{sat-cal} \) of 173 A/m\textsuperscript{2}, estimated using Eq. (S2) (see supplementary material). This coincidence between \( j_0 \) and \( j_{0-cal} \), and \( j_{sat} \) and \( j_{sat-cal} \) suggests that the NPB film exhibits ideal transient response, according to the MIS-CELIV theory that assumes charge accumulation and extraction in a continuous dielectric medium.\textsuperscript{23} When there is a problem of carrier injection at the electrode, \( j_{sat} \) decreases, compared to \( j_{sat-cal} \). Therefore, we suggest that this coincidence is an indicator of the reliability of the MIS-CELIV analysis. Thus, the MIS-CELIV mobility of the NPB film was estimated from the ideal current transients for \( V_{FB} = -10 \) V and \( A = 150 \) kV/s, using Eqs. (1) and (3), and was found to be \( 2.03 \times 10^{-4} \) cm\textsuperscript{2}/V/s.
FIG. 2. (a) Observed MIS-CELIV current transients for a voltage rise speed, $A = 150$ kV/s, at various forward voltages, $V_{FB}$ and (b) Dependence of the experimentally observed $t_{2j0}$ on $A$ in an Si/SiO$_2$/NPB/MoO$_3$/Al device.

Figure 2(b) shows the $t_{2j0}$ dependence on the voltage rise speed, $A$, from 100 to 350 kV/s. The extraction current transients for various values of $A$ are shown in Fig. S1. The $A$ dependence corresponds to the electric field dependence. Although the electric field ($E$) changes during the voltage sweep in the MIS-CELIV measurement, it has been proposed that $E$ is approximately determined by

$$E(d_s) = \frac{At_v}{d_s(1 + \varepsilon_s d_i/\varepsilon_i d_s)}. \quad (4)$$

When the carrier mobility is independent of the electric field, the relationship between $t_{2j0}$ and $A$ should be $t_{2j0} \propto A^{-0.5}$, as observed from Eq. (1). Juška et al. recently reported that the relationship of equal to or less than $t_{2j0} \propto A^{-0.5}$ corresponds to a positive field-dependent mobility, based on the Poole-Frenkel effect. In the NPB film, the relationship, $t_{2j0} \propto A^{-0.55}$, was observed, as depicted in Fig. 2(b), suggesting that the hole mobility has a positive dependence on the electric field. The mobility was increased from $1.89 \times 10^{-4}$ cm$^2$/Vs at $3.13 \times 10^4$ V/cm ($A = 100$ kV/s) to $2.16 \times 10^{-4}$ cm$^2$/Vs at a higher electric field limit of $5.48 \times 10^4$ V/cm ($A = 350$ kV/s). The estimated $E$, $t_{2j0}$, and $\mu$ are summarized in Table S1. The positively dependent mobilities of NPB have been reported in other measurement techniques, as well.

The hole mobility was estimated using SCLC measurement. Figure 3(a) shows the logarithmic current density-voltage ($J-V$) characteristics of sample-2. When the number of charge carrier traps is negligible, and the mobility is independent of the electric field, the SCL current density is described by the Mott-Gurney equation:

$$J = \frac{9}{8} \varepsilon_0 \varepsilon \mu \frac{V^2}{d^3}. \quad (5)$$

where $\varepsilon$ is the dielectric permittivity of the semiconductor and $d$ is the thickness of the organic layer. Since the work function of electrodes with MoO$_3$ is almost identical, the built-in voltage is not considered. The current density is proportional to square of the voltage ($J \propto V^2$), according to Eq. (5); however, a current density obeying $J \propto V^{2.3}$ was observed for the NPB film in the high-voltage region above 2 V. This result suggests that the hole mobility is dependent on the electric field, similar to the results of the MIS-CELIV measurement. The field dependence of the mobility, explained by the
FIG. 3. (a) Logarithmic current density versus the applied voltage and (b) Analysis curve of the space-charge-limited current in an ITO/MoO$_3$/NPB/MoO$_3$/Al device. The red line indicates the fitting line, based on the Murgatroyd equation (Eq. (7)).

Poole-Frenkel effect, is expressed as\(^\text{27}\)

$$\mu(E) = \mu_0 \exp(\gamma \sqrt{E}) .$$

(6)

By combining Eq. (5) with Eq. (6), the SCL current density, with the field dependent mobility, is given by the Murgatroyd equation:\(^\text{28}\)

$$J = \frac{9}{8} \varepsilon_0 \varepsilon \mu_0 V^3 \exp(0.89 \gamma \sqrt{V/d}) ,$$

(7)

where \(E\) is the electric field, \(\mu_0\) is the carrier mobility at zero electric field, and \(\gamma\) is the electric-field activation factor. Figure 3(b) shows the natural logarithm \(J/E^2\) versus the square root of \(E\). The experimental data indicates good linear relationship for a wide electric-field region, indicating that the SCLC in the NPB film has a positive field-dependence, based on the Poole-Frenkel effect. The fitting (red) line calculated from Eq. (7) gives the values, \(\mu_0 = 1.46 \times 10^{-4} \text{ cm}^2/\text{Vs}\) and \(\gamma = 1.79 \times 10^{-3} \text{ (cm/V)}^{1/2}\), which are close to the reported values of \(\mu_0 = 2.73 \times 10^{-4} \text{ cm}^2/\text{Vs}\) and \(\gamma = 1.32 \times 10^{-3} \text{ (cm/V)}^{1/2}\), measured by the TOF method.\(^\text{29}\)

Figure 4 shows the electric field dependence of the hole mobility, estimated from the MIS-CELIV (sample-1) and SCLC (sample-2) measurements. The electric-field dependence of the SCLC mobility was plotted using Eq. (6), with the measured values of \(\mu_0\) and \(\gamma\). The MIS-CELIV mobility gradually increased on increasing the electric field, which is interpreted as the Poole-Frenkel effect obeying the relationship, \(\log \mu \propto E^{1/2}\). From the fitted line in Fig. 4, \(\mu_0\) and \(\gamma\) were estimated to be \(1.29 \times 10^{-4} \text{ cm}^2/\text{Vs}\) and \(2.28 \times 10^{-3} \text{ (cm/V)}^{1/2}\), respectively, for the MIS-CELIV measurement. It should be noted that the absolute value and electric-field dependence of the MIS-CELIV method showed good agreement with those of the SCLC method. This agreement is explained by two aspects: One is the charge transport model for the MIS-CELIV and SCLC measurements. As suggested by the

FIG. 4. Electric-field dependence of the hole mobility, estimated using MIS-CELIV and SCLC measurements.
equation defining the extraction current transients for the MIS-CELIV method (Eq. (8)), the MIS-
CELIV theory is originally based on the current transient determined by the electric-field distribution
cause by drifting charges, \( i \), the space charges:

\[
j(t) = e \mu p(x, t) E(x, t) + \varepsilon_s \varepsilon_0 \frac{dE(x, t)}{dt},
\]

where \( p \) is the hole density. When sufficient injected charge carriers are accumulated, the boundary
condition, \( E = 0 \), is satisfied at the insulator-semiconductor interface; the insulator-semiconductor
interface is regarded as a reservoir-type ohmic contact electrode. Therefore, this transport mode is
identical to that of the SCLC measurement. This situation results in a large current density, as observed
in the SCLC, which reduces the effect of charge trapping due to the state distribution density. The other
is due to the film morphology and molecular orientation of the NPB film. The hole-transport-material
organic film in OLEDs is amorphous and homogeneous; therefore, the charge transport is hardly
affected by the grain-boundaries or the anisotropy of microcrystals. This film structure approximates
a uniform and continuous dielectric, as assumed by the MIS-CELIV and SCLC theories. The films
are deposited on the different substrates (Si/SiO\(_2\) for MIS-CELIV and ITO for SCLC); however,
it has been reported that, for amorphous small-molecules like NPB, the molecular orientation is
hardly affected by the substrates or underlying layers. Thus, we conclude that the charge transport
properties for both samples can be directly compared, and that the MIS-CELIV mobility is of the
same value as the SCLC mobility, in the case of the NPB.

The original MIS-CELIV theory assumes that the capacitance of the insulator is considerably
greater than that of the semiconductor (\( C_i/C_s \gg 1 \)). The insulator layer needs to be as thin as possible,
within a range that does not suffer from the leakage current. Here, we experimentally investigate
the influence of the capacitance ratio, \( C_i/C_s \), on the MIS-CELIV mobility, by varying the insulator
thicknesses. The respective SiO\(_2\) thicknesses of samples 3, 4, and 5 were 30, 50, and 100 nm, resulting
in a \( C_i/C_s \) of 13, 8.1, and 3.9, respectively. Figure 5 shows the current transients at \( A = 150 \) kV/s and
\( V_{FB} = -10 \) V for various SiO\(_2\) thicknesses. \( j_{sat} \) obviously decreased on increasing SiO\(_2\) the thickness,
according to Eq. (S2), and \( j_0 \) slightly decreased, according to Eq. (S1). The observed values, \( j_0 \) and \( j_{sat} \),
were in good agreement with the calculated values, \( j_{0-cal} \) and \( j_{sat-cal} \), indicating that these transients
reasonably reflect the decreasing SiO\(_2\) thickness.

Table I shows the MIS-CELIV hole mobilities for various SiO\(_2\) thicknesses, estimated using
Eqs. (2) or (3), which should be used for \( C_i/C_s \gg 1 \) or \( \geq 1 \), respectively. The mobility estimated using
Eq. (2) slightly decreased on increasing the SiO\(_2\) thickness from 30 to 100 nm, corresponding a
decrease in \( C_i/C_s \) from 13 to 3.9. The efficient voltage for the organic layer decreased on increasing
the SiO\(_2\) thickness; however, Eq. (2) does not consider the voltage drop on the insulator. Consequently,
\( t_n \) becomes longer and the mobility tends to be underestimated. In contrast, the mobility estimated
using Eq. (3), considering the voltage drop, indicated nearly constant values regardless of the SiO\(_2\)
thickness, up to 100 nm. Therefore, the MIS-CELIV mobility can be correctly estimated using
Eq. (3) for \( C_i/C_s \) in the range 3.9–13, suggesting that a 100-nm-thick insulator can be used for a

![FIG. 5. MIS-CELIV current transients with SiO\(_2\) thicknesses of 30, 50 and 100 nm, respectively, at \( A = 150 \) kV/s and \( V_{FB} = -10 \) V.](image-url)
300-nm-thick organic layer. On the other hand, the mobilities estimated using Eqs. (2) and (3) mostly agreed for a 30-nm-thick insulator, indicating that the simple model described by Eq. (2) can be used for $C_i/C_0 > 13$. These are important experimental guidelines for precise measurement using the MIS-CELIV method.

In conclusion, ideal current transients, in agreement with the theory, were observed in the NPB films, enabling the precise estimation of the hole mobility using the MIS-CELIV method. The value of the MIS-CELIV mobility was the same as that of the SCLC, due to the charge transport model and the film structure of the NPB film. In addition, the effect of the voltage drop on the insulator layer was analyzed, and an experimental guideline for precise measurement was obtained; a thick insulator even up to 100 nm can be used with an appropriate analytical model. The MIS-CELIV method has significant potential for measuring the unipolar charge mobility of organic material, and can be applied to a wide variety of OLED hole-transport materials, for rapid material screening.

See supplementary material for the theoretical equation of the current transient, and the MIS-CELIV current transients for various voltage rise speeds.

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### Table I. Hole-mobility dependence on the SiO$_2$ thickness and the estimated parameters by MIS-CELIV measurement.

| $d_i$ [nm] | $j_0$ [A/m$^2$] | $j_{sat}$ [A/m$^2$] | $\tau_{20}$ [µs] | $\mu$ (Eq. 3) [cm$^2$/Vs] | $\mu$ (Eq. 2) [cm$^2$/Vs] |
|------------|----------------|-------------------|----------------|--------------------------|--------------------------|
| 30         | 12.6           | 169               | 5.5            | $2.84 \times 10^4$       | $2.63 \times 10^4$       |
| 50         | 11.6           | 99                | 6.0            | $2.77 \times 10^4$       | $2.48 \times 10^4$       |
| 100        | 10.6           | 66                | 6.4            | $2.85 \times 10^4$       | $2.27 \times 10^4$       |
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