Electronic Supplementary Information

Solution-processed organic thermoelectric material exhibiting doping-concentration-dependent polarity

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Table of Contents

S1. Synthesis .................................................................................................................................2
S2. Measurement of Seebeck coefficient of single OTEGs ............................................................5
S3. Comparison between DMBI and NaNap as electron donors ....................................................9
S4. XPS ...........................................................................................................................................10
S5. Thermally stimulated doping of P(PymPh) ..............................................................................11
S6. TG-DTA measurement ............................................................................................................12
S7. XRD .........................................................................................................................................13
S8. Hall measurement ..................................................................................................................14
S9. Reference ................................................................................................................................16
S1. Synthesis

2,2'-(2,5-dibromo-1,4-phenylene)diethanol (1): With ice bath cooling and under nitrogen flow, sulfuric acid (90 g) was cautiously added to water (45 g) in a 300 mL flask. Once the mixture had cooled back down to room temperature, 1,4-bis(2-hydroxyethyl)benzene (30 g, 166.22 g/mol, 180 mmol, TCI) was added with stirring, followed by N-bromosuccinimide (77 g, 178.0 g/mol, 433 mol, Wako), and the resulting suspension was stirred at room temperature for 24 h. The suspension was added with stirring to an ice/water mixture (200 mL). After neutralizing with KOH aqueous solution, the mixture was extracted with EtOAc. The combined organic extracts were washed with brine and dried (Na$_2$SO$_4$), and the solvent was evaporated in vacuo to give compound 1 as a white solid (15.1 g, 24%). M.p. 135-136 °C. 1H NMR (500 MHz, DMSO-d$_6$): δ 2.80 (t, J = 6.8 Hz, 4H), 3.58 (q, J = 6.8 Hz, 4H), 4.77 (t, J = 5.3 Hz, 2H), 7.55 (s, 2H). 13C NMR (75 MHz, DMSO-d$_6$): δ 38.2, 60.0, 122.8, 134.6, 138.5. HR-MS (ESI): calcd for C$_{10}$H$_{12}$Br$_2$O$_2$ 344.9096 [M+Na]+; found 344.9094.

(2,2'-(2,5-dibromo-1,4-phenylene)bis(ethane-2,1-diyl))bis(oxy)bis(tert-butyldimethylsilane) (2): Compound 1 (16.8 g, 48.7 mmol) was dissolved in 200 mL of DMF and treated with imidazole (14 g, 205.6 mmol, 4.2 equiv., Wako) and TBDMS-Cl (18.9 g, 125.4 mmol, 2.6 equiv., TCI) at 0 °C. The reaction solution was warmed to room temperature and stirred for 24 h and then was extracted with hexane. The combined organic extracts were washed with brine and dried (Na$_2$SO$_4$), and the solvent was evaporated in vacuo to give compound 2 as a white solid (27.1 g, 95%). M.p. 50-51 °C. 1H NMR (500 MHz, CDCl$_3$): δ -0.03 (s, 12H), 0.87 (s, 18H), 2.89 (t, J = 6.8 Hz, 4H), 3.79 (t, J = 6.8 Hz, 4H), 7.43 (s, 2H). 13C NMR (75 MHz, CDCl$_3$): δ -5.3, 18.4, 26.0, 38.9, 62.2, 123.1, 135.3, 138.3. HR-MS (ESI): calcd for C$_{22}$H$_{40}$Br$_2$O$_2$Si$_2$ 573.0826 [M+Na]+; found 573.0826.
(2,2'-((2,5-bis(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)-1,4-phenylene)bis(ethane-2,1-diyl))bis(oxy)bis(tert-butyldimethylsilane) (3): Compound 2 (5.32 g, 9.6 mmol), pinacoldiboron ester (7.34 g, 28.9 mmol), KOAc (5.69 g, 57.9 mmol), and PdCl2(dppf) (472 mg, 0.58 mmol) were dissolved in DMF (20 mL) under nitrogen. The mixture was heated at 80 °C for 24 h, cooled, and then diluted with EtOAc. The organic layer was washed with water and brine and then dried (Na2SO4), and the solvent was evaporated. The crude product was purified through column chromatography (silica gel, hexane/EtOAc=8/1). Recrystallization from EtOAc gave compound 3 as a white solid (1.8 g, 28%). M.p. 133-134 °C. 1H NMR (500 MHz, CDCl3): δ -0.02 (s, 12H), 0.87 (s, 18H), 1.33 (s, 24H), 3.08 (t, J = 7.5 Hz, 4H), 3.73 (t, J = 7.5 Hz, 4H), 7.62 (s, 2H). 13C NMR (75 MHz, CDCl3): δ -5.1, 18.5, 25.0, 26.2, 38.9, 65.7, 83.6, 137.9, 138.1, 142.1. HR-MS (ESI): calcd for C34H64B2O6Si2 669.4346 [M+Na]+; found 669.4351.

6,6'-(2,5-bis(2-(tert-butyldimethylsilyloxy)ethyl)-1,4-phenylene)bis(3-bromopyridine) (4): Compound 3 (1.35 g, 2.1 mmol), 2,5-dibromopyridine (1.6 g, 6.8 mmol), CsCO3 (2.7 g, 8.3 mmol), and Pd(PPh3)4 (200 mg, 0.17 mmol) were dissolved in THF/H2O (16/12 mL) under nitrogen. The mixture was heated at 100 °C for 24 h, cooled, and then diluted with EtOAc. The organic layer was washed with water and brine and then dried (Na2SO4), and the solvent was evaporated. The crude product was purified through column chromatography (silica gel, hexane/EtOAc=20/1) and further purified using vacuum thermal gradient sublimation (under 10^-3 Pa). Finally, recrystallization from EtOAc gave compound 4 as a white crystal (540 mg, 36.5%). M.p. 140-142 °C. 1H NMR (500 MHz, CDCl3): δ -0.08 (s, 12H), 0.80 (s, 18H), 2.94 (t, J = 7.5 Hz, 4H), 3.71 (t, J
= 7.5 Hz, 4H), 7.31 (s, 2H), 7.38 (d, J = 8.3 Hz, 2H), 7.89 (dd, J = 8.3 Hz, J = 2.5 Hz, 2H),
8.75 (d, J = 1.8 Hz, 2H). 13C NMR (75 MHz, CDCl3): δ -5.3, 18.4, 26.0, 26.1, 36.2, 64.4,
119.4, 125.6, 132.5, 135.1, 139.0, 139.8, 150.4, 158.1. HR-MS (ESI): calcd for
C32H46Br2N2O2Si2 705.1537 [M+H]+; found 705.1505.

_Siloxyethyl-substituted poly(pyridine phenylene) (5)_14: A solution of 4 (1 g, 1.42
mmol), bis(1,5-cyclooctadiene)nickel (0) (1 g, 3.65 mmol), 1,5-cyclooctadiene (200 μL,
1.6 mmol), and 2,2'-bipyridine (250 mg, 1.6 mmol) in 200 mL of anhydrous DMSO was
stirred for 24 h at 80 °C under nitrogen. The reaction mixture was cooled to room
temperature and then re-precipitated into ice-cooled methanol and filtered by vacuo
twice. After washing with EDTA 2Na dehydrate aqueous solution and pure water, the
precipitate was collected, dried, dissolved in chloroform, and then re-precipitated from
methanol twice. Finally, the precipitate was purified by the Soxhlet extraction with
methanol, hexane, THF and EtOAc. The precipitate was re-dissolved in chloroform, and
the solvent was evaporated in vacuo to give compound 5 as a pale yellowish white film
(428mg). 1H NMR (500 MHz, CDCl3): δ -0.04 (s, 12H), 0.83 (s, 18H), 3.10 (br, 4H), 3.81
(br, 4H), 7.47 (br, 2H), 7.66 (br, 2H), 8.07 (br, 2H), 9.03 (br, 2H).

_Ethylene-bridged poly(pyridinium phenylene) (6)_14: To a solution of 5 (428 mg) in 100
mL of anhydrous acetonitrile, SOCl₂ (18 mL) was added, and the mixture was stirred at
room temperature for 24 h under nitrogen flow. The solution was evaporated, and
then the resulting solid was washed with DMF, THF and methanol to give polymer 6 as
a yellow film (265.5mg). 1H NMR (500 MHz, D2O): δ 3.57 (br, 4H), 5.04 (br, 4H), 8.37
(br, 2H), 8.84 (br, 2H), 9.06 (br, 2H), 9.50 (br, 2H).
S2. Measurement of Seebeck coefficient of single OTEGs

Figure S2. Schematic of the experimental setup for in-plane Seebeck coefficient (S) measurement: (a) temperature measurement mode and (b) thermoelectric voltage (ΔV_{TE}) measurement mode. (c) Typical ΔV_{TE} curves and (d) S obtained from Eqs. 3 and 4. (e) Calculated temperature gradient (ΔT_{Cal}) obtained from Eq. 6 versus temperature gradient (ΔT) for alumel as a reference material.

With every experiment, accurate measurement is important. Measurement of Seebeck coefficient (S) is particularly difficult because precise measurement of temperature gradients (ΔT) and thermoelectric (Seebeck) voltages are required. These difficulties arise from the low electrical conductivity of organic materials and the regular use of small-scale devices, i.e., thin films and narrow channel lengths.3–5

Low electrical conductivity results in a high resistance between the two-electrodes (output impedance) that are used to measure the thermoelectric voltage
(ΔV_{TE}). This high output impedance disturbs accurate measurement of ΔV_{TE} due to floating electromotive force. To suppress the floating electromotive force and accurately measure ΔV_{TE}, the ratio between input impedance of the voltage meter and output impedance of the device should be over 10^3.\textsuperscript{6}

In our measurement system, we use a digital multi meter (Keithley DMM2000 6\textsuperscript{1/2}, input impedance > 10^{10} Ω, resolution 0.1 μV); thus, the output impedance of the sample must be < 1MΩ. Considering our sample configuration (W = 23 mm, L = 3 mm, t = 100-300 nm), σ of the sample must be > 10^{-2} S cm\textsuperscript{-1}. This is why S of intrinsic organic semiconductors is so difficult to measure. For these reasons, people using thin-film materials prefer a narrow channel length, e.g., several tens of micrometers, in order to decrease output impedance. However, an extremely narrow channel length makes applying a controlled and steady ΔT difficult.

In this study, S of single OTEGs were measured with a homemade setup as described in \textbf{Fig. 3a}. To determine S, it is necessary to apply a ΔT across the sample and measure ΔV_{TE} and ΔT. Fine chromel and alumel wires (φ 50 μm, Nilaco) were used in order to resolve these problems. Chromel and alumel are alloys with known S and well known as materials for forming K-type thermocouples.

Even though chromel and alumel have high thermal conductivity compared with organic materials, a thermocouple of the two wires has high thermal resistance due to their fine and long geometry. Thus, heat loss through these thermocouple while measuring thermoelectric voltage can be prevented, and this ultrafine thermocouple can provide fast, precise, and accurate monitoring of local temperature changes of the sample.\textsuperscript{7}

In addition, thermoelectric voltages can be measured through the inner two
electrode strips using the chromel-chromel or alumel-alumel wire pairs after the temperature stabilized (steady-state). These $\Delta V_{\text{fe}}$ can be expressed as

$$\Delta V_{\text{Chr-Chr}} = \Delta T (S_{\chi} - S_{\text{Chr}}), \text{ Equation S1}$$

$$\Delta V_{\text{Alu-Alu}} = \Delta T (S_{\text{Alu}} - S_{\chi}). \text{ Equation S2}$$

Here $\Delta V_{\text{Chr-Chr}}$ and $\Delta V_{\text{Alu-Alu}}$ are the thermoelectric voltages measured across the inner two chromel-chromel and alumel-alumel wire pairs, respectively. $\Delta T$ is the temperature gradient applied to the sample. $S_{\text{Chr}}$ and $S_{\text{Alu}}$ are the Seebeck coefficients of chromel (+22 $\mu$V K$^{-1}$) and alumel ($-19$ $\mu$V K$^{-1}$) respectively. Finally, $S_{\chi}$ is the Seebeck coefficient of the sample being measured and can be expressed using Eqs. S1 and S2 as follows:

$$S_{\chi, \text{Chr-Chr}} = \frac{\Delta V_{\text{Chr-Chr}}}{\Delta T} + S_{\text{Chr}}, \text{ Equation S3}$$

$$S_{\chi, \text{Alu-Alu}} = -\frac{\Delta V_{\text{Alu-Alu}}}{\Delta T} + S_{\text{Alu}}, \text{ Equation S4}$$

$$S_{\chi} = S_{\chi, \text{Chr-Chr}} = S_{\chi, \text{Alu-Alu}} = \frac{\Delta V_{\text{Chr-Chr}} - \Delta V_{\text{Alu-Alu}}}{2\Delta T} + \frac{S_{\text{Chr}} + S_{\text{Alu}}}{2}. \text{ Equation S5}$$

From Eqs. S3-S5, $S_{\chi}$ can be evaluated using either the chromel-chromel or alumel-alumel wire pair or both. Here, $S_{\chi, \text{Chr-Chr}}$ and $S_{\chi, \text{Alu-Alu}}$ are the $S_{\chi}$ estimated through the inner two chromel-chromel and alumel-alumel wire pairs respectively.

The reason why two equations and wire pairs were used when estimating $S_{\chi}$ is
that the temperature gradient (\(\Delta T_{\text{cal}}\)) also can be calculated from Eqs. S3 and S4 as

\[
\Delta T_{\text{cal}} = \left( \frac{\Delta V_{\text{Chr-Chr}} + \Delta V_{\text{Alu-Alu}}}{S_{\text{Chr}} - S_{\text{Alu}}} \right) \text{.} \tag{Equation S6}
\]

Thus, the accuracy of \(S_x\) and \(\Delta T\) can be confirmed by comparing \(S_{x, \text{Chr-Chr}}\) with \(S_{x, \text{Alu-Alu}}\) and comparing \(\Delta T\) measured by the two K-type thermocouples at the hot and cold sides with \(\Delta T_{\text{cal}}\)\(^9\).

The in-plane \(S\) of alumel was measured as a reference using the homemade setup described in the experimental section to ensure the accuracy of the measurement system (see Figs. S2c-d). While varying the temperature gradient through control of the current of the Peltier devices, \(\Delta V_{\text{TE}}\) versus \(\Delta T\) was automatically recorded by computer. The applied \(\Delta T\) increased from zero up to +2.5 K (forward), decreased from +2.5 K down to -2.5 K (reverse), and then increased back to zero. If the measured material was pyroelectric, exhibited ion conductivity, or did not achieve thermal steady state, this cycle would show hysteresis because these phenomena are related to the temperature rise rate\(^10\). A typical \(\Delta V_{\text{TE}}\) versus \(\Delta T\) curve when measuring alumel as a reference material is shown in Figure S2c. The \(\Delta V_{\text{Alu-Alu}}\) is practically zero over the entire region because the measured alumel sample was measured with alumel wires, so \(\Delta V_{\text{TE}}\) should cancel each other regardless of the applied temperature gradient. In contrast, \(\Delta V_{\text{Chr-Chr}}\) should change at a rate of \(S_{\text{Alu}} - S_{\text{Chr}}\). Thus, the slopes measured through the inner alumel-alumel and chromel-chromel wire pairs were 0 \(\mu V\) K\(^{-1}\) and -41 \(\mu V\) K\(^{-1}\) respectively. A Seebeck coefficient of \(-19\pm0.1\ \mu V\ \text{K}^{-1}\) was obtained from the linear slopes of lines fit to each \(\Delta V_{\text{TE}}\)-\(\Delta T\) curve over all the data and Eqs. S3-S5, which matches well with literature.
Figure S2d shows instantaneous $S_{\chi, \text{Chr-Chr}}$ and $S_{\chi, \text{Alu-Alu}}$ at each $\Delta T$ obtained using Eqs. S3 and S4 and the individual data points in Fig. S2c. $S_{\chi, \text{Chr-Chr}}$ and $S_{\chi, \text{Alu-Alu}}$ obtained from Eqs. S3 and 4 converge to $S_{\text{Alu}}$ at about $|\Delta T| > 1$ K. For $|\Delta T| < 1$ K, the error is large because of high output impedance and floating electromotive force. Furthermore Fig. S2e shows that the calculated temperature gradient $\Delta T_{\text{cal}}$ obtained from Eq. S6 matches well with the measured temperature gradient $\Delta T$.

The $S$ values of several other reference samples were also measured to further confirm the accuracy of measurement system. The measured $S$ of chromel, gold, nickel, and platinum were 21.9±0.2 µV K$^{-1}$, 2.0±0.4 µV K$^{-1}$, −19.3±0.4 µV K$^{-1}$, and −0.9±0.65 µV K$^{-1}$ respectively.

**S3. Comparison between DMBI and NaNap as electron donors**

![Absorption Spectra](image)

**Figure S3.** Optical absorption spectra of a pristine P(PymPh) film, a film annealed at 150 °C for 30 min, and films doped with DMBI or NaNap. Inset: The molecular structures of P(PymPh), DMBI, and NaNap.
Another n-type dopant that has been used in OTEGs is 4-(1,3-dimethyl-2,3-dihydro-1H- benzoimidazol-2-yl)phenyl)dimethylamine (DMBI). To compare the effectiveness of doping with DMBI, the optical absorption spectra of a pristine P(PymPh) film, a film annealed at 150 °C for 30 min, and films doped with DMBI or NaNap are shown in Fig. S3. The NaNap doping was done the same way as described in the experimental section. For DMBI doping, DMF was used as solvent and heated to 150 °C to activate DMBI.\textsuperscript{11,12} All processing was done in nitrogen. The higher ratio of the absorption peaks attributed to charged polymer species (592 nm, 1100-2500 nm) to those of the neutral polymer (428 nm) for the film doped with NaNap than the annealed film and film doped with DMBI suggests that NaNap is the stronger reductant.

\section*{S4. XPS}

\begin{figure}[h]
\centering
\begin{subfigure}{0.32\textwidth}
\includegraphics[width=\textwidth]{Na_1s}
\caption{Na 1s}
\end{subfigure}
\hfill
\begin{subfigure}{0.32\textwidth}
\includegraphics[width=\textwidth]{Cl_2p}
\caption{Cl 2p}
\end{subfigure}
\hfill
\begin{subfigure}{0.32\textwidth}
\includegraphics[width=\textwidth]{C_1s}
\caption{C 1s}
\end{subfigure}
\caption{XPS (a) Na 1s, (b) Cl 2p, and (c) C 1s spectra of pristine, annealed, and NaNap-doped films of P(PymPh). The concentration of NaNap in the doping solution ranged from 1.0-6.0 mM as indicated on the plots. As references, Ag and NaNap were also measured.}
\end{figure}
Comparing the XPS Na 1s and Cl 2p signals in Figs. S4a and b, peaks derived from the formation of sodium chloride, 1072.6 eV at Na 1s and 199.6 eV at Cl 2p, can be observed.\textsuperscript{13} Thus, we conclude that the sodium, which was introduced by NaNap, reduces the pyridinium ions and reacts with the chloride anion to form sodium chloride. In contrast, although peaks consisting of C-N were obtained in Fig. S4c, a large chemical shift following the doping was not obtained.

S5. Thermally stimulated doping of P(PymPh)

![Figure S5](image)

**Figure S5.** Conductivity ($\sigma$) of annealed P(PymPh) films at room temperature.

To investigate the effect of annealing treatment on electrical conductivities ($\sigma$), a device with bottom silver electrodes on a glass substrate was used. The bottom silver electrodes and a P(PymPh) thin film were prepared the same way as described in the experimental section and Fig. 3a. The film was measured, annealed for 30 min in nitrogen atmosphere, cooled to room temperature, and re-measured, and this process was repeated for consecutively high temperatures. The $\sigma$ measured by 4T-sensing method at room temperature are shown in Fig. S5 for the different annealing
temperatures. The film and metal electrode fabrication and annealing treatments were carried out under nitrogen without any air exposure.

From Fig. S5, \( \sigma \) can be seen to start to increase at an annealing temperature of about 125 °C. One reason for the increase could be the degassing of oxygen and water. Another one would be self-doping due to the cleaving of \( \text{N}^+-\text{Cl}^- \) chemical bonds in accordance with thermal stimulation.

S6. TG-DTA measurement

A commercial TG-DTA system (TG-DTA2400SA, Bruker) was used for thermogravimetric (TG)/differential thermal analysis measurements (DTA) measurements. After loading the sample of P(PymPh), nitrogen gas was flowed at a rate of 200 mL min\(^{-1}\) during the measurement. The resulting TG-DTA curves are shown in Fig. S6.

Below 100-125 °C, the TG continuously decreases with increasing temperature. Because P(PymPh) is very polar, water and oxygen are expected to be absorbed during the sample loading, so this may correspond to degassing of the sample. Thus, an
annealing temperature of 150 °C was chosen to degas water and oxygen without the decomposition of P(PymPh), which begins at about 200 °C.

S7. XRD

Figure S7: (a) Out-of-plane XRD patterns of pristine and annealed P(PymPh) on Si substrates. (b) Main crystallographic structures of P(PymPh).

The effects of doping on the films were further studied using X-ray diffraction (XRD) analysis because the crystallinity of the polymer matrix can be changed by annealing treatment and dopants physically penetrating into or moving out of the polymer during doping. Thus, the polymer films can undergo swelling, shrinking, or other morphological changes that can also impact the electrical properties.14,15

The changes in the crystal structure of P(PymPh) films on silicon substrates were investigated using a Rigaku Ultima IV XRD system. The out-of-plane XRD patterns of pristine and annealed P(PymPh) films are shown in Fig. S7a. Results for doped films are not shown in Fig. S7a because the doped films were too unstable in ambient air to reliably measure in our XRD system.
In all cases, only one small peak was found at $2\Theta = 23\text{-}24^\circ$, which indicates that the matrix of P(PymPh) should be almost amorphous compared with P3HT, which is a general conducting polymer that can exhibit a large number of diffraction peaks such as (100) to (300) and (010) to (020). The peak for P(PymPh) corresponds to the (010) $\pi\text{-}\pi$ stacking distance, and lattice constants of $d = 3.5\text{-}3.7$ Å were estimated according to Bragg’s law ($2dsin\Theta = n\lambda$). Thus, the P(PymPh) polymer crystallites have preferential face-on orientation and are predominantly oriented horizontally to the substrate surface as shown in Fig. S7b.

Comparing the full widths at half maximum (FWHM) of the (010) peaks for the pristine and annealed films, the annealed film (0.94 Å) is much narrower than the pristine film (1.12 Å), which may be advantageous for charge transport (i.e., electrical conduction) due to an increase in crystallinity and also play a role in the results in Fig. S5.

S8. Hall measurement

When a current-carrying semiconductor is kept in a magnetic field, the charge carriers of the semiconductor experience a Lorentz force in a direction perpendicular to both the magnetic field and the current, which is called the Hall effect. Thus, Hall measurements make discrimination of carrier type and estimation of carrier density and mobility possible. Here, $R_H$ is the Hall coefficient, $V_H$ is the Hall voltage, $B$ is the magnetic field, $I$ is the current, $t$ is the thickness of the sample, and $q$ is the charge of an electron. The values $n_h$ and $n_e$ are the charge carrier densities of holes and electrons, respectively, and $\mu_h$ and $\mu_e$ are the hole and electron mobilities.
Finally, $\sigma$ is electrical conductivity, which can be broken into components from holes ($\sigma_h$) and electrons ($\sigma_e$).

$$R_H = \frac{V_H}{BI} = \frac{n_h \mu_h^2 - n_e \mu_e^2}{|q|(n_h \mu_h + n_e \mu_e)^2} \quad \text{Equation S7}$$

$$\sigma = \sigma_h + \sigma_e = |q|(n_h \mu_h + n_e \mu_e) \quad \text{Equation S8}$$

When a single carrier species, i.e., electrons or holes, dominates the current flow in the semiconductor, the equations can be simplified as Eqs. S9 and S10. Here, $n_H$ and $\mu_H$ correspond to the charge carrier density and carrier mobility of the dominate carrier species.

$$n_H = \frac{1}{|qR_H|} \quad \text{Equation S9}$$

$$\mu_H = |R_H|\sigma \quad \text{Equation S10}$$

Hall measurements of P(PymPh) doped with NaNap were performed to obtain further information on carrier type, carrier density, and mobility. Device fabrication was almost the same as described in the experimental section, but a different device configuration was adopted based on the Van der Pauw method (Fig. S8). All processing and measurements were carried out under nitrogen without any exposure to air.

For the Hall measurements, a commercial magnet system (ResiTest 8340DC/CSE, TOYO Corporation) was used. A constant magnetic field of $-0.5$ T or $+0.5$ T and a constant DC current (Source Meter 2400, Keithley) were applied to the sample. To
eliminate the drift of background voltage and thermoelectric voltage in accordance with the Peltier effect in the sample, delta-mode measurement was adopted for measuring the precise Hall voltage (Nanovolt Meter 2182A, Keithley).

![Diagram](image)

**Figure S8.** (a) Device configuration for Van der Pauw method, and (b) the result of the Hall measurements.

**Figure S8** shows the device configuration for the Van der Pauw method and the results of the Hall measurements. A film of P(PymPh) doped at a concentration of 2.0 mM was used as the active layer. The negative value of $V_H$ indicates that the main carriers are electrons, which agrees with the negative values for Seebeck coefficient. From Eqs. S9 and S10, $R_H$ should be independent of applied current, and $n_H$ and $\mu_H$ should be constant.\(^{17}\) From Fig. S8b, $V_H$ increases linearly as a function of $I$ with a slope of nearly unity, satisfying the above requirements. From Eqs. S7-S10 and the results in Fig. S8, $R_H$, $n_H$ and $\mu_H$ can be estimated as a $-1.13 \text{ cm}^3 \text{ C}^{-1}$, $5.5 \times 10^{18} \text{ cm}^{-3}$ and $5.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively.
S9. Reference


