

Supporting Information

Methyl Functionalization on Polyimide Side Chains as Gate Dielectrics for Organic Transistors

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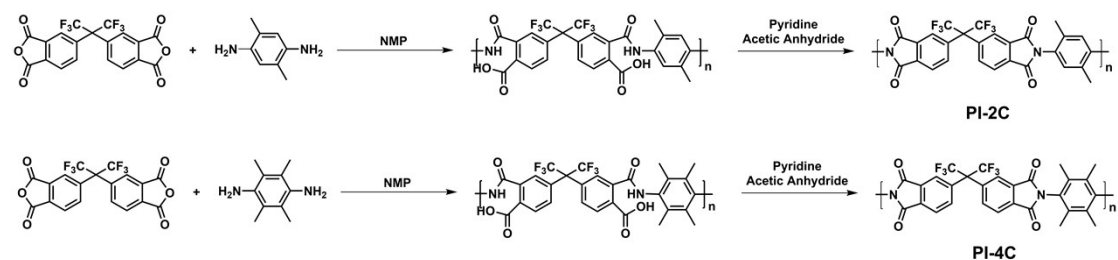
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Materials.

4,4'-(Hexafluoroisopropylidene) diphthalic anhydride (6FDA), 2,5-dimethyl-1,4-phenylenediamine and 2,3,5,6-tetramethyl-1,4-phenylenediamine, methanol, *n*-methylpyrrolidone (NMP), pyridine and acetic anhydride were both purchased from Energy Chemical Co., Ltd. and used without further purification.



Scheme S1. Synthetic routes of PI-2C and PI-4C.

Synthesis of the PI-2C and PI-4C polymer.

The synthetic routes of PI-2C and PI-4C are shown in **Scheme S1**. The specific experimental procedures are as follows:

PI-2C and PI-4C: 2,5-dimethyl-1,4-phenylenediamine (2.00 g, 14.68 mmol) and 6FDA (6.52 g, 14.68 mmol) in 1:1 molar ratio were dissolved in anhydrous NMP (30 mL) at room temperature under Ar atmosphere. After 5 h, a small excess of acetic anhydride and pyridine was added to the poly(amic acid) solutions at room temperature and allowed to stir for 12 h. The resulting polymer was purified by precipitation into excess methanol 5 times, and then the polymer was collected by filtration and dried under a 100 °C vacuum to obtain the PI-2C polymer. The synthesis of the PI-4C polymer follows similar experimental procedures and molar ratios, except that 2,5-dimethyl-1,4-phenylenediamine was replaced by 2,3,5,6-tetramethyl-1,4-phenylenediamine.

The ¹H-NMR spectrum of **PI-2C** polymer in DMSO-*d*₆. ¹H-NMR (500 MHz, DMSO-*d*₆, ppm): δ 8.17-8.25 (d, 2H), 7.94-8.03 (s, 2H), 7.78-7.87 (s, 2H), 7.41-7.49 (s, 2H), 2.05-2.24 (s, 6H).

The $^1\text{H-NMR}$ spectrum of **PI-4C** polymer in $\text{DMSO-}d_6$. $^1\text{H-NMR}$ (500 MHz, $\text{DMSO-}d_6$, ppm): δ 8.17-8.26 (d, 2H), 87.91-8.03 (d, 4H), 1.96-2.17 (s, 12H).

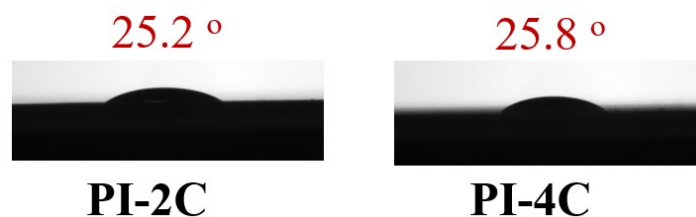


Figure S1. Images of diiodomethane contact angle of PI-2C and PI-4C.

Table S1. The summary and comparison of device performance.

Dielectric layer	Organic semiconductor	Mobility ($\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$)	Threshold voltage (V)	
PI-2C		0.31	~ 0	
PI-4C	<i>p</i> -6P/VOPc	0.05	~ 2.0	
PI- C_0		0.029	-13.4	Ref. S1

Reference

(S1) Wang, B.; Xu, T.; Yu, B.; Zou, J.; Luan, S. Optimization of Alkyl Side Chain Length in Polyimide for Gate Dielectrics to Achieve High Mobility and Outstanding Operational Stability in Organic Transistors. *ACS Appl. Mater. Interfaces* **2023**, *15*, 7204-7216.