

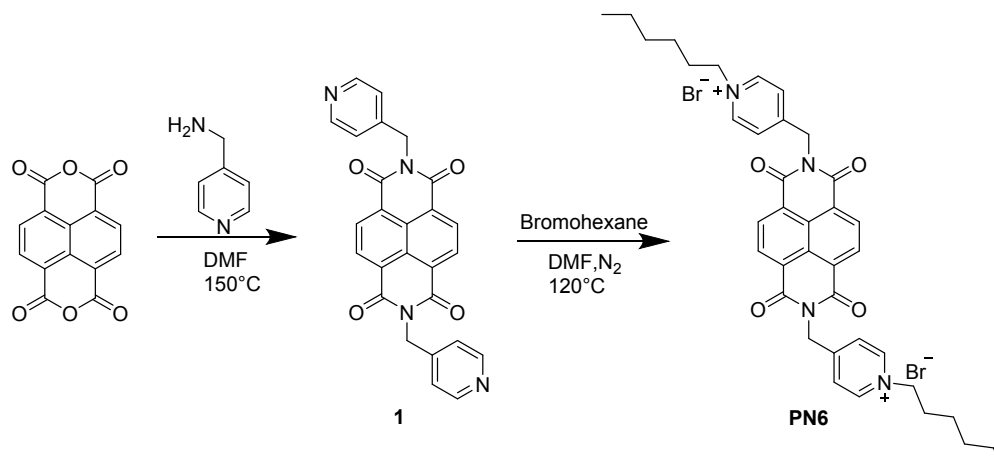
**An Efficient and Thickness Insensitive Cathode Interface Material
for High Performance Inverted Perovskite Solar Cells with 17.27 %
Efficiency**

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Experiment Details:

1. Synthesis of PN6



Scheme 1. Synthetic route of PN6

The representative synthesis process of PN6 is shown in Scheme 1. 1,4,5,8-Naphthalenetetracarboxylic dianhydride, 4-(aminomethyl)pyridine and bromohexane were obtained from commercial suppliers without further purification.

Synthesis of N,N'-Bis(4-pyridylmethyl)-1,4,5,8-naphthalenetetracarboxydiimide (1)

1,4,5,8-Naphthalenetetracarboxylic dianhydride (5.36 g, 20 mmol) and 4-(aminomethyl)pyridine (10.8 g, 100 mmol) were refluxed in anhydrous DMF (150 mL) for 12 h. The mixture was cooled down to room temperature and then filtrated. The solid was washed with ethanol and acetone prior to drying in vacuo. The crude product was further purified by sublimation and orange-brown crystalline solid was obtained with a yield of 56% (0.5 g). ¹H NMR (400 MHz, CDCl₃, δ): 8.81 (s, 4H; ArH), 8.58 (d, J = 8.0 Hz, 4H; PyH), 7.40 (d, J = 8.0 Hz, 4H; PyH), 5.39 (s, 4H; CH₂Py); Anal. calcd for C₂₆H₁₆N₄O₄: C 69.64; H 3.60; N 12.49. Found: C 69.67; H 3.65; N 12.48.

Synthesis of N,N'-Bis(1-n-hexylpyridin-1-ium-4-ylmethyl)-1,4,5,8-naphthalenetetracarboxydiimide (PN6)

A mixture of bromohexane (4.95 g, 30 mmol) and **1** (1.35 g, 3 mmol) in DMF (50 mL) was heated for 12 h at 120 °C under N₂. The mixture was cooled down to room temperature and then poured in ether. A golden powder was isolated by filtration and washed with acetone. The crude product was further purified by recrystallization with ethanol. Yield: 1.87 g, 80.2%. ¹H NMR (300 MHz, DMSO-d₆, δ): 9.05 (d, J = 6 Hz, 4H; PyH), 8.72 (s, 4H; ArH), 8.26 (d, J = 6 Hz, 4H; PyH), 5.53 (s, 4H; CH₂Py), 4.56 (t, J = 6 Hz, 4H; CH₂-C₅H₁₁), 1.89 (m, 4H; CH₂-C₄H₉), 1.26 (m, 12H; (CH₂)₃),

0.84 (t, $J=6$ Hz, 6H;CH₃). ¹³C (100 MHz, CD₃OD, δ): 163.19, 157.56, 144.40, 130.86, 127.27, 126.92, 126.72, 61.32, 42.96, 31.06, 30.95, 25.54, 22.14, 12.93; HRMS (ESI) m/z : [M + H]⁺ calcd for C₃₈H₄₂Br₂N₄O₄:779.5860; found, 779.5657.

2. Measurements

Nuclear magnetic resonance (NMR) was taken on Bruker AVANCE III 300MHz and 400MHz Spectrometer. All chemical shifts were reported relative to tetramethylsilane (TMS) at 0.0ppm, unless otherwise stated.

The absorption spectra were recorded with UV-visible spectrophotometer (Shimadzu 2450). Cyclic voltammetry was determined by electrochemical workstation (Chenhua CHI600E).

PN6 electron only device fabrication: The J-V characteristics of PN6 electron-only device were measured with device structure of ITO/ZnO (30nm)/PN6 (90 nm)/Ag (90nm), The PN6 layer films were prepared by dissolving PN6 in methanol and spin-coated on top of the ZnO layer. Then, the 90 nm Ag was evaporated through a shadow mask in a vacuum chamber under a pressure of 1×10^{-4} Pa.

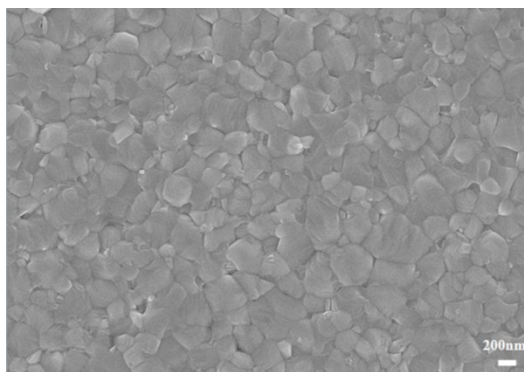


Figure S1. SEM image of the perovskite layer

Table S1 Photovoltaic Parameters of PHJ PSCs with 20 nm PFN

Cathode	V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	PCE_{max} (PCE_{ave}^a) (%)
Thickness of PFN	1.11	10.24	12.91	1.47(1.05)

^aThe average PCE was calculated from over 8 devices

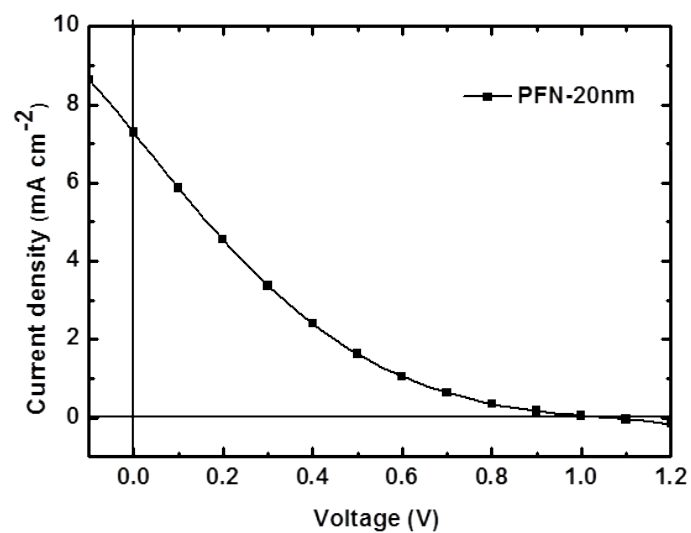


Figure S2. J-V curves of the devices with 20nm PFN interface layer, as measured under 100mW/cm² AM 1.5G irradiation

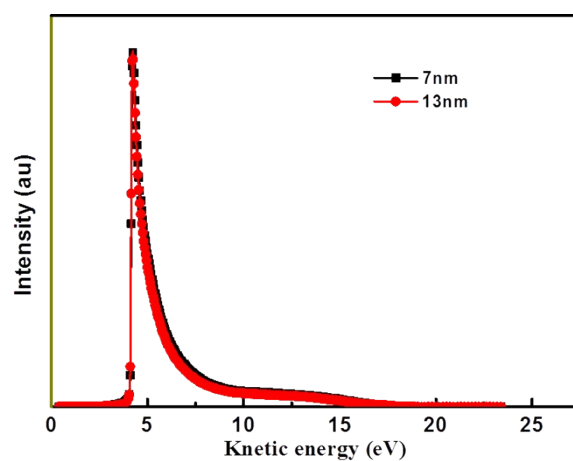


Figure S3. UPS spectra of Ag/PN6 thin films with the different thickness PN6 layers.

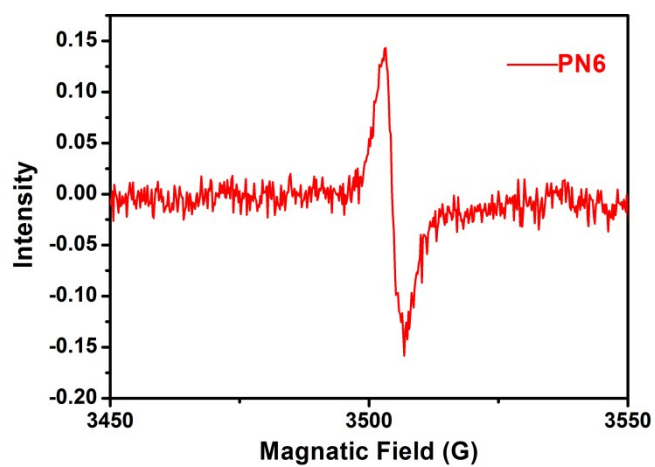


Figure S4. ESR spectra of PN6 in solid state.