

## Supplementary Information

### Efficient and selective capture of thorium ions by a covalent organic framework

Xiaojuan Liu <sup>1</sup>, Feng Gao <sup>1</sup>, Tiantian Jin <sup>1</sup>, Ke Ma <sup>1</sup>, Haijiang Shi <sup>1</sup>, Ming Wang <sup>2</sup>,  
Yanan Gao <sup>2</sup>, Wenjuan Xue <sup>3</sup>, Jing Zhao <sup>1,\*</sup>, Songtao Xiao <sup>1,\*</sup>, Yinggen Ouyang <sup>1,\*</sup>,  
Guoan Ye <sup>1,\*</sup>

<sup>1</sup> Department of Radiochemistry, China Institute of Atomic Energy, Beijing 102413, China.

<sup>2</sup> Key Laboratory of Ministry of Education for Advanced Materials in Tropical Island Resources, Hainan University, Haikou 570228, China.

<sup>3</sup> State Key Laboratory of Separation Membranes and Membrane Processes, Tiangong University, Tianjin 300387, China

\* Corresponding authors: Department of Radiochemistry, China Institute of Atomic Energy, Beijing 102413, China (J Zhao, S. Xiao, Y. Ouyang, G. Ye).

E-mail addresses: [Jing\\_zhao@tom.com](mailto:Jing_zhao@tom.com) (Jing Zhao) [Songtao\\_Xiao@tom.com](mailto:Songtao_Xiao@tom.com) (Songtao Xiao); [ouyang\\_yinggen@163.com](mailto:ouyang_yinggen@163.com) (Yinggen Ouyang); [guoan\\_ye@tom.com](mailto:guoan_ye@tom.com) (Guoan Ye).

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## 1. Experimental section

### 1.1 Materials and characterizations

All starting reagents/reactants, except 4,4',4'',4'''-(Pyrene-1,3,6,8-tetrayl) tetraaniline (PyTTA), 2'3'5'6'-tetrafluoro-[1,1':4',1''-terphenyl]-4,4'-dicarbaldehyde (TFTDA), 5,6-Bis(4-formylbenzyl)-1-methyl-1*H*-benzimidazole (BFIm), and 5,6-Bis(4-formylbenzyl)-1,3-dimethyl-benzimidazolinium iodide (BFIm), were purchased commercially. 1,3,6,8-tetrabromopyrene was procured from Yanshen Technology Co., Ltd. 4-formylbenzeneboronic acid pinacol ester, camphorsulfonic, and 4,7-dibromo-2,1,3-benzothiadiazole were procured from Shanghai Adamas Reagent Co., Ltd. Tetrakis (triphenylphosphine) palladium (0), triethyl orthoformate, and 4-aminophenylboronic acid pinacol ester were procured from Meryer (Shanghai) Chemical Technology Co., Ltd. Methyl iodide was procured from Xiya Reagent. 1,4-dibromotetrafluorobenzene was procured from J&K Scientific. Sodium borohydride, anhydrous MgSO<sub>4</sub>, anhydrous K<sub>2</sub>CO<sub>3</sub>, dioxane, ethyl acetate, tetrahydrofuran, ethanol, dichloromethane, and methanol were procured from Greagent of Shanghai Titan Scientific Co., Ltd. All reagents are of analytical grade and used directly as received without further purification.

Powder X-ray diffraction (PXRD) patterns analyses of the adsorbents in the form of powder to characterize the crystal structure were carried out by Bruker AXS X-ray diffractometer equipped with Nifiltered Cu KR radiation (40 kV, 40 mA). The patterns were recorded over the range of 2 $\theta$  angles from 2-30 ° in steps of 0.02 °/s. PXRD patterns of Th(IV)-loaded samples were carried out on Rigaku Ultima IV. Fourier

transform infrared spectroscopy (FTIR) spectra were obtained on Nicolet iS50 spectrometer (Thermo Fisher Scientific Company) by scans of 32 with a resolution of 4 cm<sup>-1</sup> by the KBr pellet method. The range of wavenumbers is 4000-400 cm<sup>-1</sup>. X-ray photoelectron spectroscopy (XPS) spectra were acquired using Perkin-Elmer PHI 5000CESCA system equipped with Al K $\alpha$  radiation (1486.6 eV). The assignment of the peak was based on the Handbook of X-ray Photoelectron Spectroscopy. <sup>1</sup>H and <sup>13</sup>C nuclear magnetic resonance (NMR) spectra were recorded on a Bruker Advance III 400 MHz NMR spectrometer. High-resolution mass spectral (HRMS) data was recorded by SHIMADZU LCMS-IT-TOF. Gas adsorption isotherms were measured at 77 K using a liquid N<sub>2</sub> bath on a surface area analyzer, Autosorb iQ. The fresh sample was activated in the degassing station of the instrument at 30 °C for 2 h, and then at 120 °C for 13 h to make the pores guest-free. Thermogravimetric analysis (TGA) was recorded using a Mettler Toledo DSC3+ thermogravimetric analyzer over the temperature range of 25-900 °C in a nitrogen atmosphere with a heating rate of 10 °C min<sup>-1</sup>. Scanning electron microscopy (SEM) and energy dispersive spectrometer (EDS) were collected using a Hitachi S-4800. The concentrations of all metal ions used in this work were analyzed by ICP-MS (Optima 8000, PerkinElmer) using the standard curve method.

## **1.2 Preparation of the Py-TFImI-25 COF and Py-TFIm-25 COF**

### **1.2.1 Synthesis of 4,4',4'',4'''-(Pyrene-1,3,6,8-tetrayl) tetraaniline (PyTTA)**

The synthesis process of the organic building block PyTTA was referred to the literature<sup>1,2</sup> and improved, and the specific synthetic pathway is shown in Supplementary Fig. 1, which was mainly prepared from 1,3,6,8-tetrabromopyrene with

4-aminophenylboronic acid pinacol ester by Suzuki coupling reaction.

K<sub>2</sub>CO<sub>3</sub> (2.18 g, 15.7 mmol), 1,3,6,8-tetrabromopyrene (1.48 g, 2.86 mmol), 4-aminophenylboronic acid pinacol ester (3.01 g, 13.7 mmol), and tetrakis(triphenylphosphine) palladium (0) (0.33 g, 0.29 mmol) were placed into a three-necked round-bottomed flask. 8 mL of deoxygenated ultrapure water and 32 mL of dry dioxane were added under N<sub>2</sub> atmosphere, respectively, and the reaction was refluxed at 85 °C for 72 h. After the reaction, the product was cooled to room temperature, and the solid on the filter paper was repeatedly washed with water until the filtrate was colorless. The solid was then dispersed in methanol and sonicated, suction filtered, the solid on the filter paper was washed several times with methanol, and the solid PyTTA was collected, and vacuum-dried at room temperature overnight. The yield was 74%. <sup>1</sup>H NMR (400 MHz, DMSO) δ 8.14 (s, 4H), 7.80 (s, 2H), 7.35 (d, *J* = 8.3 Hz, 8H), 6.78 (d, *J* = 8.4 Hz, 8H), 5.32 (s, 8H).

#### 1.2.2 Synthesis of 2'3'5'6'-tetrafluoro-[1,1':4',1''-terphenyl]-4,4'-dicarbaldehyde (TFTDA)

TFTDA was synthesized<sup>2</sup> by a Suzuki coupling reaction similar to PyTTA (see Supplementary Fig. 2). K<sub>2</sub>CO<sub>3</sub> (2.63 g, 19.0 mmol), 1,4-dibromotetrafluorobenzene (1.65 g, 5.36 mmol), 4-formylbenzeneboronic acid pinacol ester (3.36 g, 14.48 mmol) and tetrakis(triphenylphosphine) palladium(0) (0.35 g, 0.30 mmol) were added into the three-necked round-bottomed flask. 12 mL of deoxygenated ultrapure water and 50 mL of dry dioxane were added under N<sub>2</sub> atmosphere, respectively, and the reaction was refluxed at 100 °C for 24 h. Then the reaction product was cooled to room temperature,

the obtained solid was suction filtered, washed with a small amount of water, ethyl acetate, and ethanol in turn, and the product was dried at 80°C overnight to obtain TFTDA as a white light solid powder. The yield was 80%. <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ 10.12 (s, 2H), 8.12 (d, *J* = 8.4 Hz, 4H), 7.85 (d, *J* = 8.0 Hz, 4H).

### 1.2.3 Synthesis of 5,6-Bis(4-formylbenzyl)-1-methyl-1*H*-benzimidazole (BFIm)

The synthesis process of the organic building unit BFIm was referred to the literature<sup>2-4</sup> and improved, and the specific synthesis pathway is shown in Supplementary Fig. 3.

4,7-dibromo-2,1,3-benzothiadiazole (3.50 g, 11.91 mmol) was added to the two-necked round-bottomed flask, and under N<sub>2</sub> atmosphere, 400 mL of absolute ethanol and 40 mL of dry tetrahydrofuran (THF) were added for stirring. After the reaction system was cooled to 0 °C, sodium borohydride (8.11 g, 211.31 mmol) was added, and then transferred to room temperature for 3 h. After the reaction, it was placed in an ice-water bath, quenched by slowly adding 100 mL of deionized water, and rotary-evaporated under reduced pressure. The crude product was dissolved in 150 mL of ether, poured into a separatory funnel, and washed with saturated brine three times, the organic phase was dried over anhydrous MgSO<sub>4</sub>, filtered, and rotary-evaporated under reduced pressure to obtain an orange flaky solid 4,5-Dibromo-1,2-benzenediamine (P1). To prevent the decomposition of P1, it needs to be protected with N<sub>2</sub> or used directly in the next reaction step. The yield was 86%. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 6.85 (s, 2H), 3.74 (s, 4H).

P1 (2.16 g, 8.11 mmol) was dissolved in 90 mL of CH<sub>2</sub>Cl<sub>2</sub>, then triethyl orthoformate

(1.35 mL, 12.39 mmol) was added, and after stirring at room temperature for 10 min, 27.5 mg of camphorsulfonic acid was added to the reaction system and the stirring was continued for 3 h. A large amount of white solid was produced, which was filtered and washed several times with a small amount of diethyl ether to obtain a white layered solid 5,6-Dibromobenzimidazole (P2). The yield was 67%.  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ )  $\delta$  8.41 (s, 1H), 7.38 (s, 2H).

P2 (1.97 g, 7.14 mmol) and  $\text{K}_2\text{CO}_3$  (2.96 g, 21.4 mmol) were added to the three-necked flask, then 50 mL of absolute ethanol was added, the temperature was raised to 50 °C, and then 1.0 mL of methyl iodide was added dropwise. The reaction system was sealed and refluxed for 3 h. After the reaction was completed, the reaction product was cooled to room temperature, 20 mL of deionized water was added, and rotary evaporated under reduced pressure to remove ethanol, filtered and washed with water and then with n-hexane/ethyl ether (1:1), and vacuum-dried at 60 °C overnight to obtain beige solid powder 5,6-Dibromo-1-methyl-1*H*-Benzimidazole (P3). The yield was 86%.  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ )  $\delta$  8.36 (s, 1H), 7.37 (s, 2H), 4.09 (s, 3H).

$\text{K}_2\text{CO}_3$  (2.76 g, 20 mmol), P3 (1.93 g, 6.67 mmol), 4-formylbenzeneboronic acid pinacol ester (4.67 g, 15.35 mmol), and 385 mg tetrakis(triphenylphosphine) palladium(0) (385 mg, 0.33 mmol) were added to the three-necked round-bottomed flask, respectively, and deoxygenated deionization was added under  $\text{N}_2$  atmosphere. 12 mL of water and 50 mL of dry dioxane. The reaction was refluxed at 100 °C for 15 h. After the reaction system was cooled to room temperature, deionized water was added, and the mixture was poured into a separatory funnel. After the aqueous phase was

extracted with ethyl acetate, it was combined with the organic phase and washed with saturated NaCl solution. The product was rotary evaporated under reduced pressure, and the crude product was washed with n-hexane/diethyl ether (2/1) to obtain a brown powdery solid 5,6-Bis(4-formylbenzyl)-1-methyl-1*H*-benzimidazole (P4). The yield was 65%. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 10.14 (s, 1H), 10.09 (s, 1H), 8.21 (d, *J* = 8.1 Hz, 2H), 8.03 (d, *J* = 11.6 Hz, 4H), 7.93 (s, 1H), 7.67 (d, *J* = 8.0 Hz, 2H), 7.54 (d, *J* = 7.7 Hz, 1H), 7.27 (s, 1H), 3.46 (s, 3H). <sup>13</sup>C NMR (101 MHz, CDCl<sub>3</sub>) δ 192.28 (s), 191.83 (s), 145.89 (s), 144.70 (s), 144.44 (s), 142.52 (s), 135.89 (s), 135.37 (s), 132.44 (s), 131.55 (s), 130.70 (s), 130.07 (s), 129.99 (s), 129.54 (s), 126.06 (s), 125.26 (s), 121.93 (s), 34.71 (s). HRMS-ESI (*m/z*) [*M*+*H*]<sup>+</sup> Calcd for C<sub>22</sub>H<sub>17</sub>N<sub>2</sub>O<sub>2</sub><sup>+</sup>, 341.1212, Found 341.1262.

#### 1.2.4 Synthesis of 5,6-Bis(4-formylbenzyl)-1,3-dimethyl-benzimidazolinium iodide (BFIIIm)

The synthesis process of the organic building unit BFIIIm was referred to the literature<sup>2,5</sup> and improved. As shown in Supplementary Fig. 4, P4 (0.40 g, 1.17 mmol), 12 mL of acetonitrile, and methyl iodide (0.73 mL, 11.7 mmol) were added to a 100 mL single-neck flask and refluxed at 85 °C for 24 h. It was cooled to room temperature, evaporated under reduced pressure, and the solid was washed with n-hexane/ethyl acetate (2/1) and dried under vacuum at 100 °C overnight. The brown product P5 was obtained. The yield was 65%. <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ 10.17 (s, 2H), 9.72 (s, 1H), 8.12 (d, *J* = 8.1 Hz, 4H), 7.82 (d, *J* = 8.0 Hz, 4H), 7.63 (s, 2H), 3.56 (s, 6H).

#### 1.2.5 Synthesis of Py-TFImI-25 COF and Py-TFIm-25 COF

PyTTA (22.7 mg, 0.04 mmol), TFTDA (21.5 mg, 0.06 mmol), and BFIm (9.6 mg, 0.02 mmol) (or BFIm with 6.9 mg, 0.02 mmol) were added to a 10 mL Pyrex tube. Add 2.0 mL of o-dichlorobenzene and 0.2 mL of 6 M AcOH, respectively, and mix well by sonication. The solvent-containing part at the lower end of the Pyrex tube was refrigerated and degassed in 77 K liquid nitrogen, circulated about three times, and the tube was sealed at a high temperature. After the system was completely restored to room temperature, it was transferred to a constant temperature oven at 120 °C and left to react for 72 h. After the reaction, the product was fully washed with dry THF 5 times, then soaked in fresh 30 mL THF solution for about 8 h. Then, the products were filtered and washed three times with dry acetone. Vacuum dry at 20 °C overnight to obtain Py-TFImI-25 COF (or Py-TFIm-25 COF). The yield was 82% for Py-TFImI-25 COF, and 80% for Py-TFIm-25 COF.

### 1.3 Thorium ions capture

#### 1.3.1 Effect of pH on the Th(IV) adsorption

Five sets of  $\sim 25 \text{ mg L}^{-1}$  Th(IV) solutions were prepared at 50 mL each. The pH value was adjusted to 1.0, 2.0, 3.0, 4.0, and 5.0 by adding  $0.1 \text{ mol L}^{-1}$  NaOH and  $0.1 \text{ mol L}^{-1}$  HNO<sub>3</sub> by dropwise, respectively. 4.5 mL of the Th(IV) solution was mixed with 1.5 mg COF material at 25 °C and shaking with 200 rpm for 1 h. The adsorbed solution was filtered through a 0.22  $\mu\text{m}$  aqueous nylon filter, and the Th(IV) concentration before and after adsorption was measured. The following equations were used to obtain the equilibrium adsorption capacity  $q_e$  ( $\text{mg g}^{-1}$ ), the distribution coefficient  $K_d$  ( $\text{mL g}^{-1}$ ), and separation factor  $SF$  (a. u.).

$$q_e = \frac{C_0 - C_e}{m} \times V \quad (1)$$

$$K_d = \frac{C_0 - C_e}{C_e m} \times V \quad (2)$$

$$SF_{a/b} = \frac{K_d^a}{K_d^b} \quad (3)$$

Where,  $C_0$ ,  $C_e$ ,  $V$ ,  $m$  refer to the initial concentration, equilibrium concentration, solution volume, and adsorbent mass of an element, respectively.  $a$ , and  $b$  refer to two different elements, respectively.

### 1.3.2 Adsorption kinetics

The kinetic studies were carried out under the following conditions. The initial concentration of Th(IV) solution was around 40 mg L<sup>-1</sup> with a pH value of 4, and the solid-liquid ratio was maintained at 1:3000 g mL<sup>-1</sup> by adding 1.5 mg COF materials into 4.5 mL Th(IV) solution. At each predetermined time, a small amount of supernatant was taken and filtered through a 0.22 µm aqueous nylon filter to test the Th(IV) solution concentration before and after adsorption. The adsorption amount ( $q_t$ , mg g<sup>-1</sup>) at a contact time of  $t$  was calculated by Equation (4).

$$q_t = \frac{C_0 - C_t}{m} \times V \quad (4)$$

The adsorption kinetic data was analyzed using two models, the pseudo-first-order model and the pseudo-second-order model, and the linearized forms of the two models are shown in Equation (5) and Equation (6), respectively.

$$\ln(q_e - q_t) = \ln q_e - k_1 t \quad (5)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (6)$$

Where  $q_e$  and  $q_t$  (mg g<sup>-1</sup>) are the amounts of adsorbates adsorbed at equilibrium and

the predetermined time  $t$  (min),  $q_e$  (mg g<sup>-1</sup>) is fitting equilibrium capacity.  $k_1$  (min<sup>-1</sup>) and  $k_2$  (g.mg<sup>-1</sup>.min<sup>-1</sup>) are the pseudo-first-order and pseudo-second-order adsorption rate constant, respectively.

### 1.3.3 Effect of different Th(IV) concentration on the capture capacity

The initial concentration of Th(IV) solution was ranging from 15 to 300 mg L<sup>-1</sup> with a pH value of 4, and the solid-liquid ratio was maintained at 1:3000 g mL<sup>-1</sup> by adding 1.5 mg COF materials into 4.5 mL Th(IV) solution. At various metal ion concentrations, the saturation adsorption capabilities of the adsorbents toward Th(IV) were measured.

### 1.3.4 Adsorption selectivity

A multi-ion solution of Th(IV), U(VI), Sr(II), Cs(I), La(III), Pr(III), Nd(III), Sm(III), Eu(III), and Gd(III) was prepared with each metal ion maintaining at a concentration of around 25 mg L<sup>-1</sup> and the pH was adjusted to 4. The solid-liquid ratio was 1:3000 g mL<sup>-1</sup> by adding 1.5 mg COF materials into 4.5 mL multi-ion solution. The batch adsorption experiments were carried out at a temperature of 25 °C and a speed of 200 rpm, and then the multi-ion solution after adsorption was separated with a 0.22 μm aqueous nylon filter to measure the concentration of Th(IV) and the competing ions.

## 1.4 Computational Details

In this work, the crystal models of Py-TFImI-25 and Py-TFIm-25 COF were constructed with the Crystal Building module of the Material Studio software. Then, a series of geometry optimizations were performed with UFF force field at the ultra-fine quality in the Forcite module, allowing optimization of all lattice parameters and atomic coordinate. All the optimized COF materials were used for subsequent simulation of

the powder diffraction patterns with the Reflex module in the Material Studio software. Both the eclipsed AA and staggered AB stacking modes were tried for the Py-TFImI-25 and Py-TFIm-25 synthesized here. Compared the experimental XRD results with the simulated XRD, finally, the eclipsed AA stacking modes were settled due to the good match of the experimental XRD results with the simulated XRD on the eclipsed AA stacking mode. Based on the obtained results, periodic density functional theory (DFT) calculations were performed with the cell parameters of  $42.7443 \times 15.8124 \times 49.7048 \text{ \AA}^3$ ,  $\alpha=87.9005^\circ$ ,  $\beta=89.7684^\circ$ ,  $\gamma=81.1063^\circ$  for Py-TFImI-25 COF and  $45.3198 \times 15.8606 \times 48.2641 \text{ \AA}^3$ ,  $\alpha=89.3474^\circ$ ,  $\beta=90.1480^\circ$ ,  $\gamma=85.4182^\circ$  Py-TFIm-25 COF, respectively. During the structural DFT optimization, all the atoms in the periodic Py-TFImI-25 COF, Py-TFIm-25 COF, and the adsorbed heavy metal ions were allowed to be fully relaxed. The Pawley refinement was performed to iteratively optimize the lattice parameters until the  $R_{WP}$  value converges and the observed profile fitted well with the refined one. Detailed results are in Supplementary Table 1 and 2.

Structure optimization and energy calculations were performed using density functional theory, as implemented in the CP2K code of the QUICKSTEP program by employing a mixed Gaussian and plane-wave basis sets<sup>6-8</sup>. Core electrons were represented with norm-conserving Goedecker-Teter-Hutter pseudopotentials<sup>6,9,10</sup>, and the valence electron wave function was expanded in a double-zeta basis set with polarization functions<sup>11</sup> along with an auxiliary plane wave basis set with an energy cutoff of 400 Ry. The generalized gradient approximation exchange-correlation functional of Perdew, Burke, and Enzerhof (PBE)<sup>12</sup> was used. Each configuration was

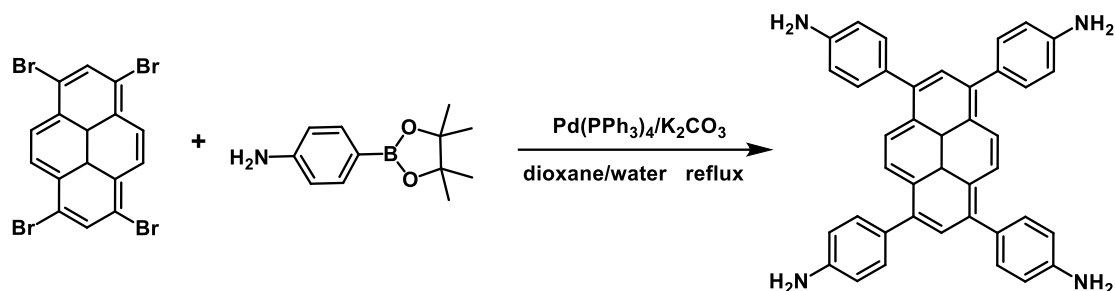
optimized with the Broyden-Fletcher-Goldfarb-Shanno (BGFS) algorithm with SCF convergence criteria of  $1.0 \times 10^{-8}$  au. To compensate the long-range van der Waals dispersion interaction between the adsorbate and the framework, the DFT-D3 scheme<sup>13</sup> with an empirical damped potential term was added into the energies obtained from exchange-correlation functional in all calculations. The self-consistent continuum solvation (SCCS) model with a dielectric constant of 78.54 was used to simulate the solvent environment<sup>14-16</sup>.

The adsorption potential energy between the heavy metal cations  $M$  ( $M = \text{Th}^{4+}$ ,  $\text{La}^{3+}$ ,  $\text{Nd}^{3+}$ ,  $\text{UO}_2^{2+}$ ,  $\text{Sr}^{2+}$ , and  $\text{Cs}^{+}$ ) and the Py-TFImI-25 COF and Py-TFIm-25 COF substrates can be calculated using the following equation:

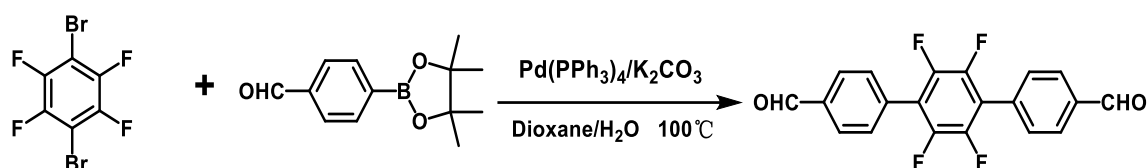
$$\Delta E_{\text{ads}} = E_{M@ \text{substrate}} - E_{\text{substrate}} - E_M \quad (7)$$

In Equation (7),  $E_{M@ \text{substrate}}$  and  $E_{\text{substrate}}$  represent the total energies of the substrate with and without adsorbate, respectively.  $E_M$  is the total energy of the metal cation  $M$ . According to this equation, negative adsorption energy corresponds a stable adsorption structure.

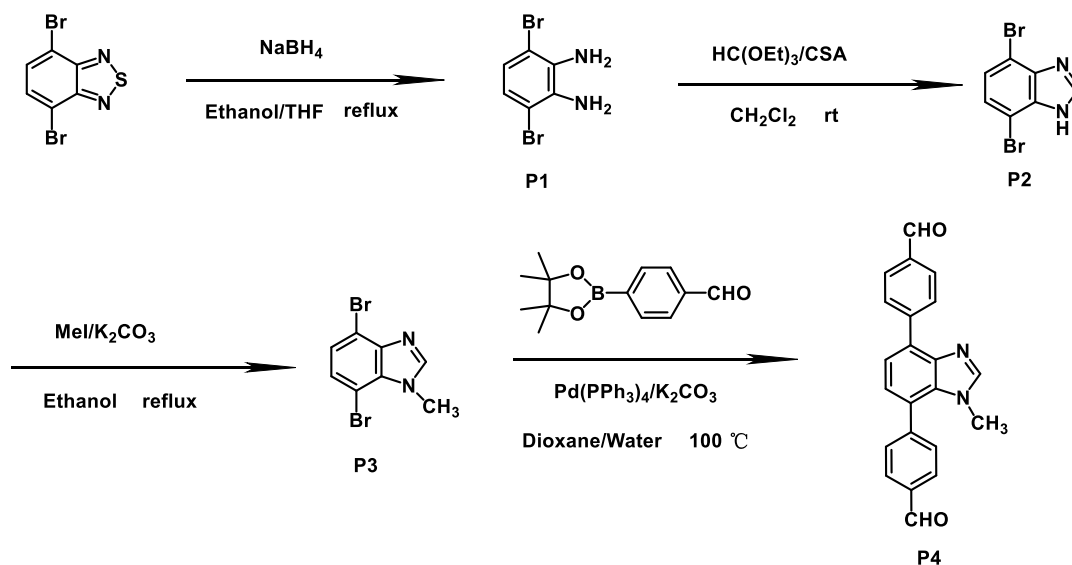
## 2. Supplementary Figures 1-25



**Supplementary Fig. 1. Synthesis of starting material.** Synthetic route to 4,4',4'',4'''-(Pyrene-1,3,6,8-tetrayl) tetraaniline (PyTTA).

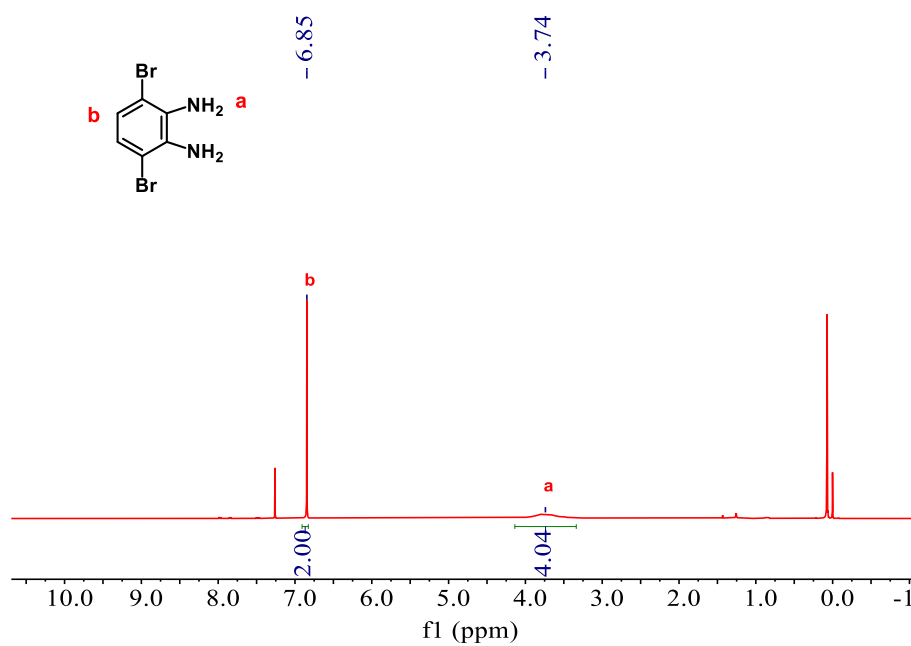


**Supplementary Fig. 2. Synthesis of starting material.** Synthetic route to 2,3,5,6-tetrafluoro-[1,1':4',1''-terphenyl]-4,4'-dicarbaldehyde (TFTDA).

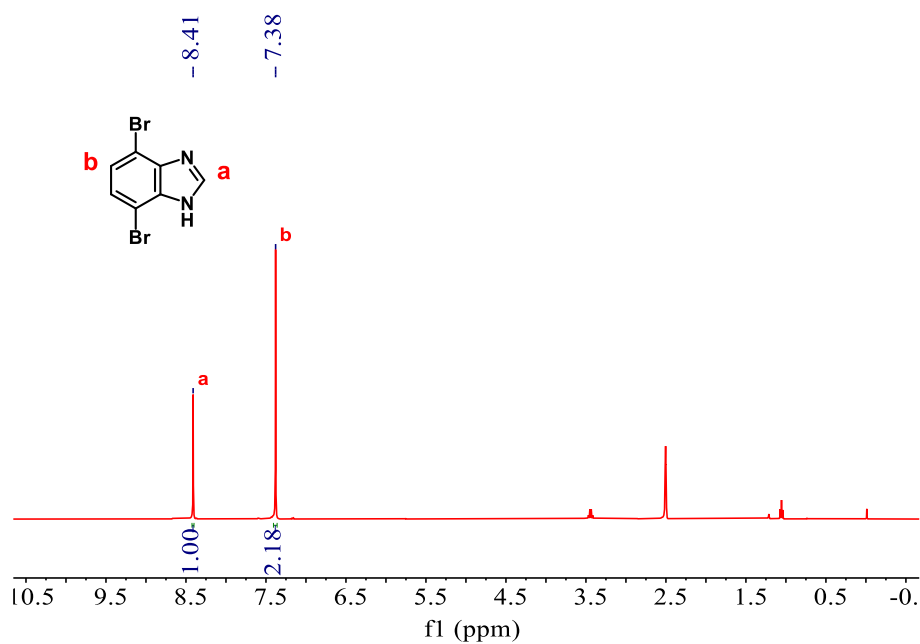


**Supplementary Fig. 3. Synthesis of starting material.** Synthetic route to 5,6-bis(4-formylbenzyl)-1-methyl-1H-benzimidazole (BFIm, P4).

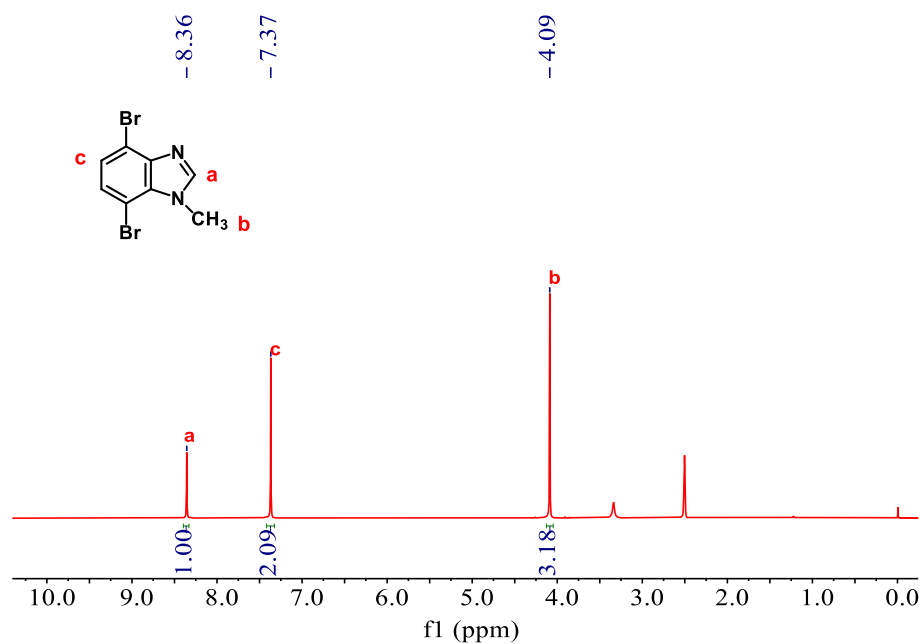




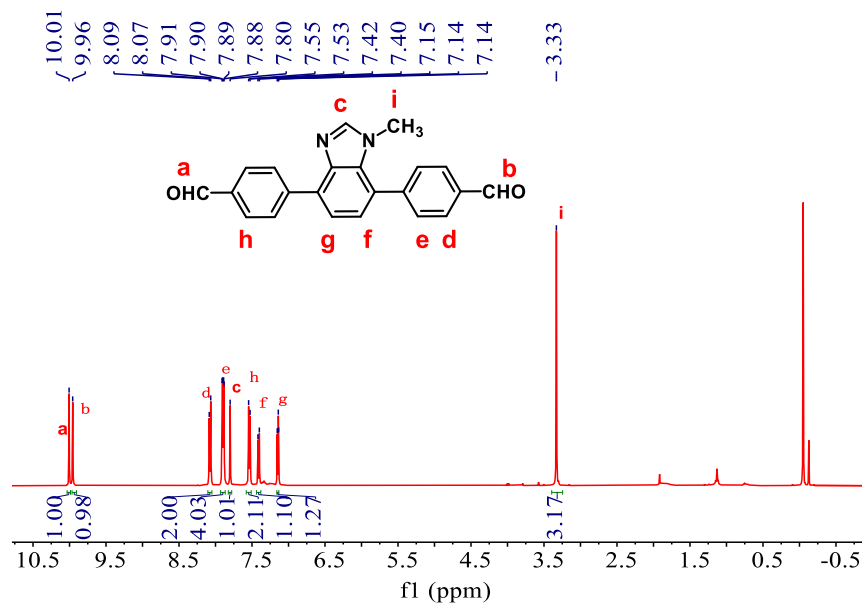
**Supplementary Fig. 7. Characterization of starting material.** <sup>1</sup>H NMR spectrum of 4,5-Dibromo-1,2-benzenediamine (P1).



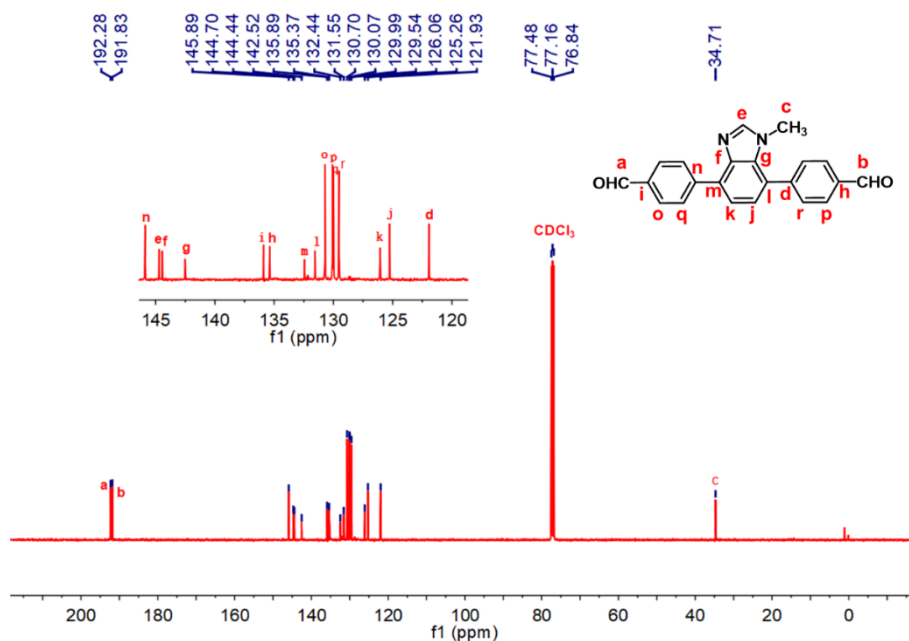
**Supplementary Fig. 8. Characterization of starting material.** <sup>1</sup>H NMR spectrum of 5,6-Dibromobenzimidazole (P2).



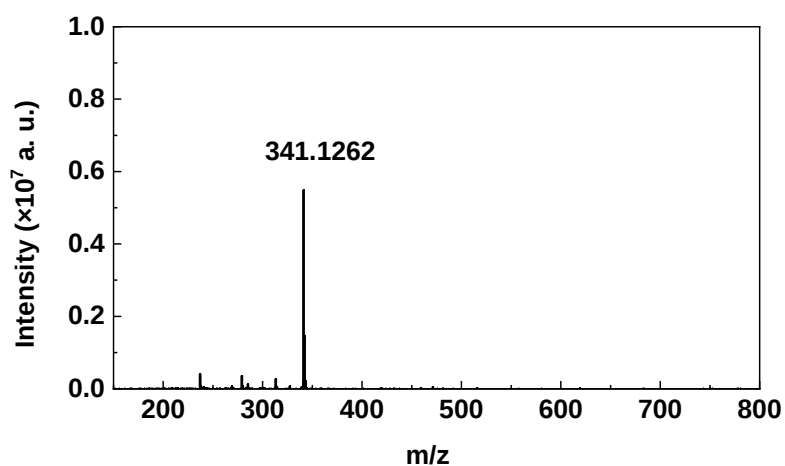
**Supplementary Fig. 9. Characterization of starting material.** <sup>1</sup>H NMR spectrum of 5,6-Dibromo-1-methyl-1H-Benzimidazole (P3).



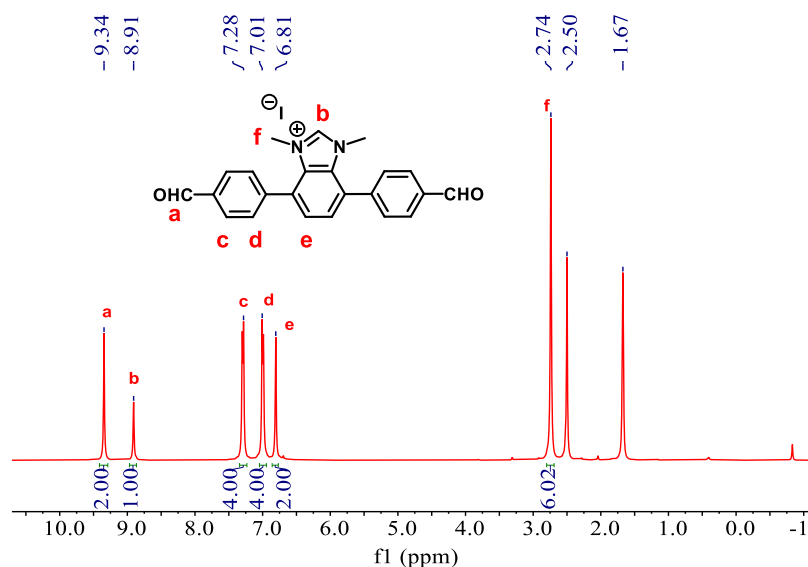
**Supplementary Fig. 10. Characterization of starting material.** <sup>1</sup>H NMR spectrum of 5,6-Bis(4-formylbenzyl)-1-methyl-1H-benzimidazole (BFIm, P4).



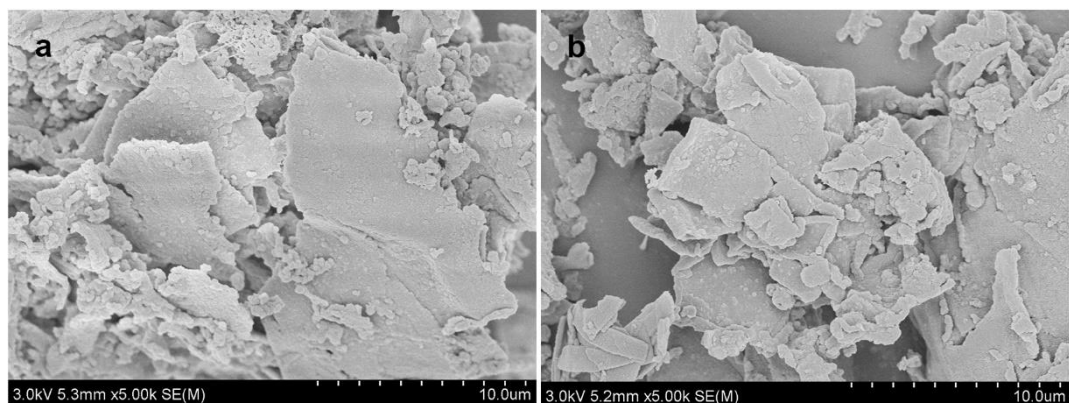
**Supplementary Fig. 11. Characterization of starting material.** <sup>13</sup>C NMR spectrum of 5,6-Bis(4-formylbenzyl)-1-methyl-1H-benzimidazole (BFIm, P4).



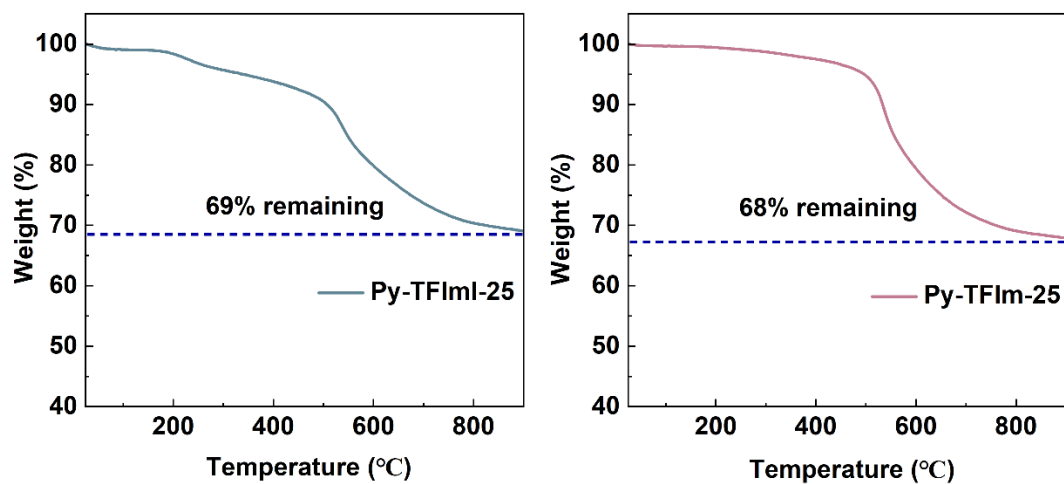
**Supplementary Fig. 12. Characterization of starting material.** HRMS data of 5,6-Bis(4-formylbenzyl)-1-methyl-1H-benzimidazole (BFIm, P4).



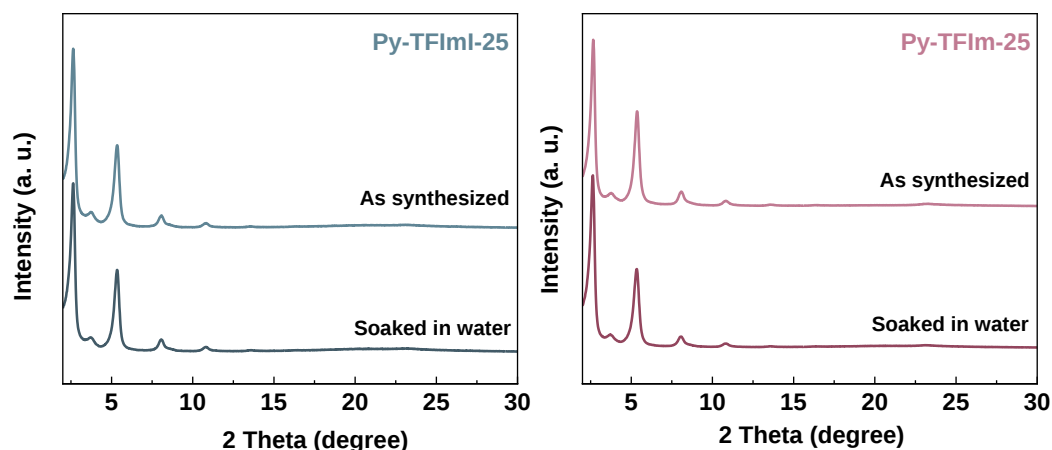
**Supplementary Fig. 13. Characterization of starting material.**  $^1\text{H}$  NMR spectrum of 5,6-Bis(4-formylbenzyl)-1,3-dimethyl-benzimidazolinium iodide (BFIm, P5).



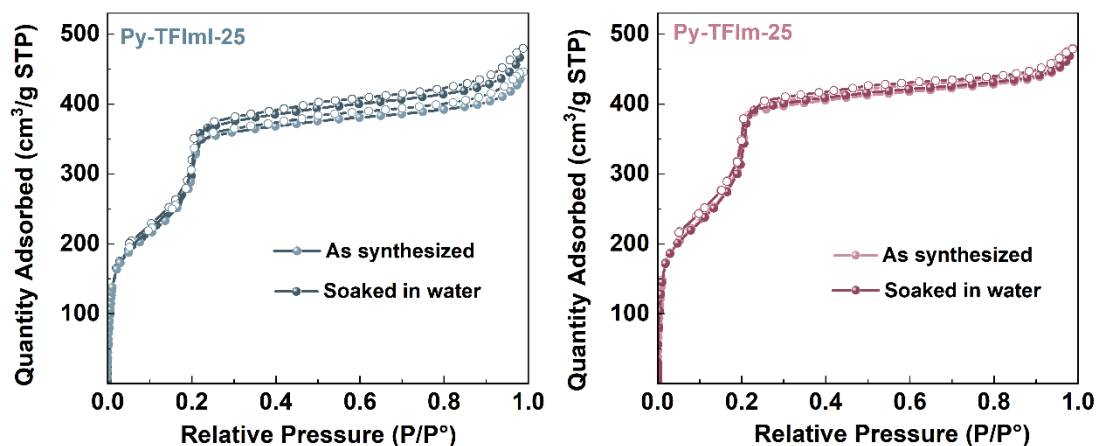
**Supplementary Fig. 14. COF characterization.** SEM images of **a** Py-TFImI-25 COF and **b** Py-TFIm-25 COF.



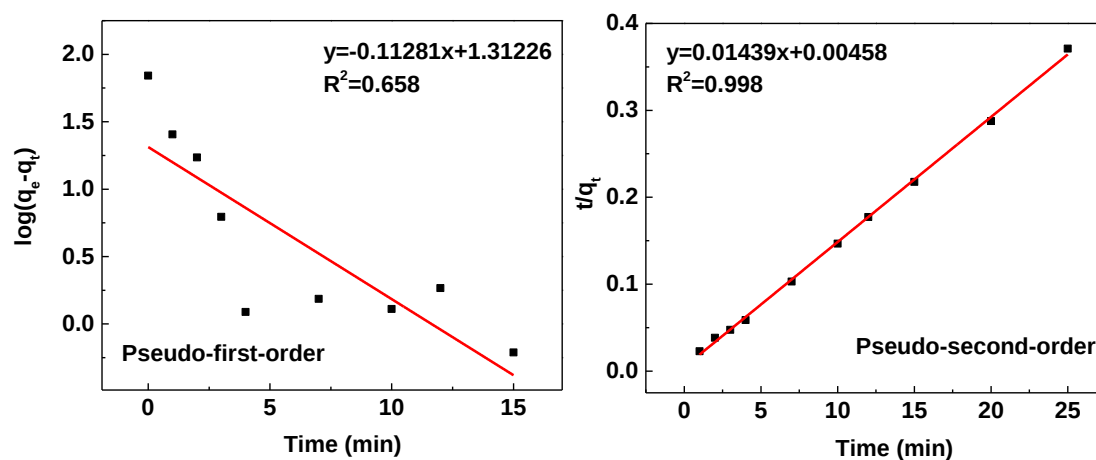
**Supplementary Fig. 15. COF characterization.** TGA measurements of Py-TFImI-25 COF and Py-TFIm-25 COF.



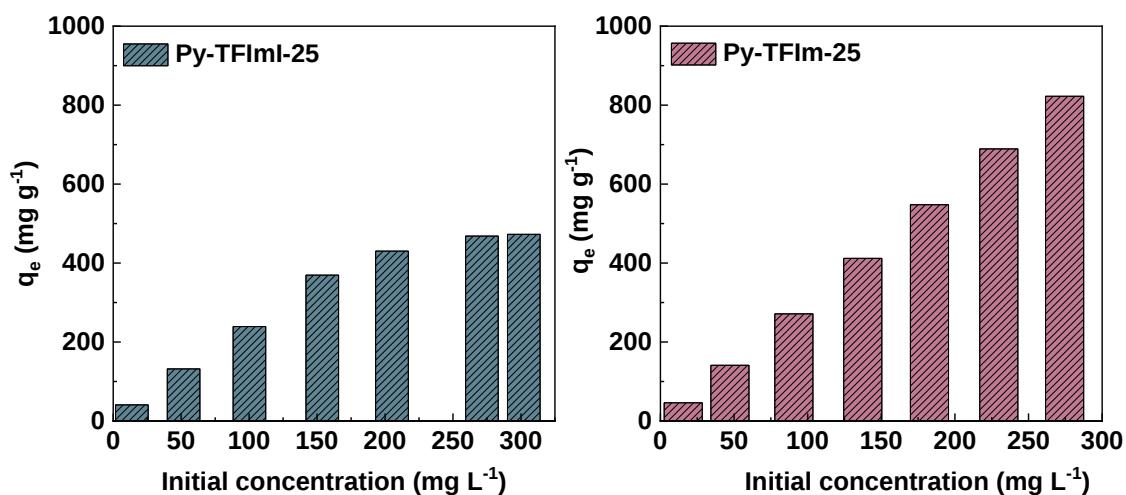
**Supplementary Fig. 16. COF characterization.** PXRD patterns of Py-TFImI-25 COF and Py-TFIm-25 COF after soaking in water. (Solid-liquid ratio = 1:3000 g mL<sup>-1</sup>, Shaking time = 1 h, T= 25°C).



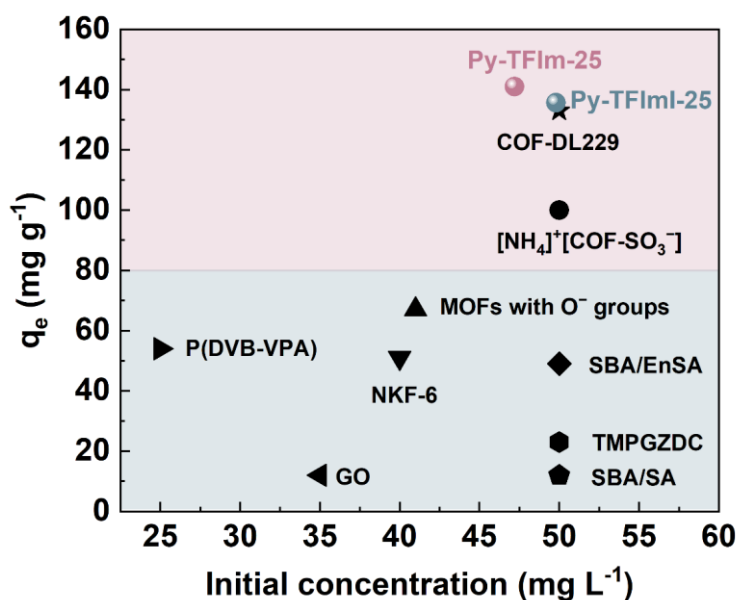
**Supplementary Fig. 17. COF characterization.** N<sub>2</sub> sorption isotherms of Py-TFImI-25 COF and Py-TFIm-25 COF after soaking in water. (Solid-liquid ratio = 1:3000 g mL<sup>-1</sup>, Shaking time = 1 h, T= 25°C).



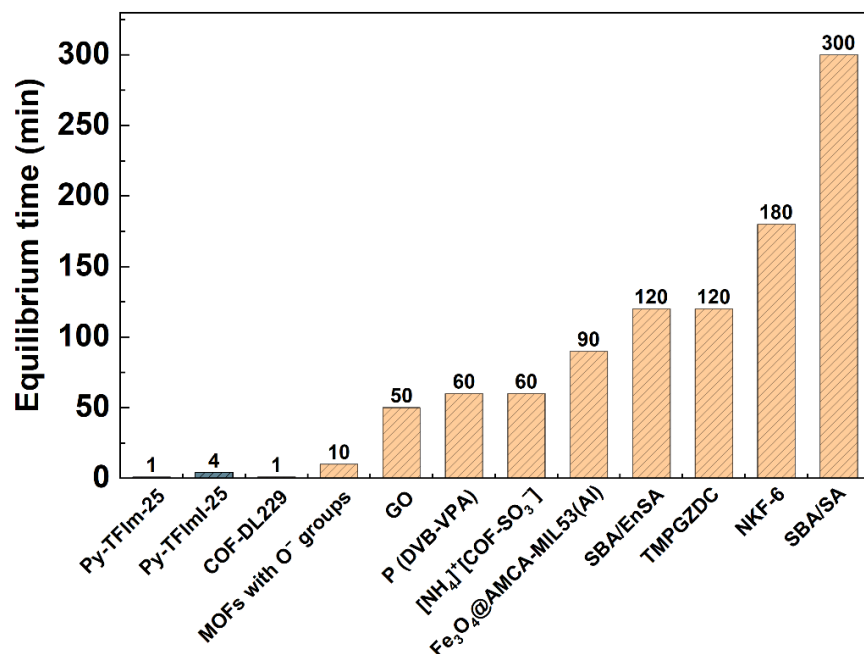
**Supplementary Fig. 18. Adsorption performance.** Linear fitting of Py-TFImI-25 COF to Th(IV) adsorption kinetics by two kinetic models.



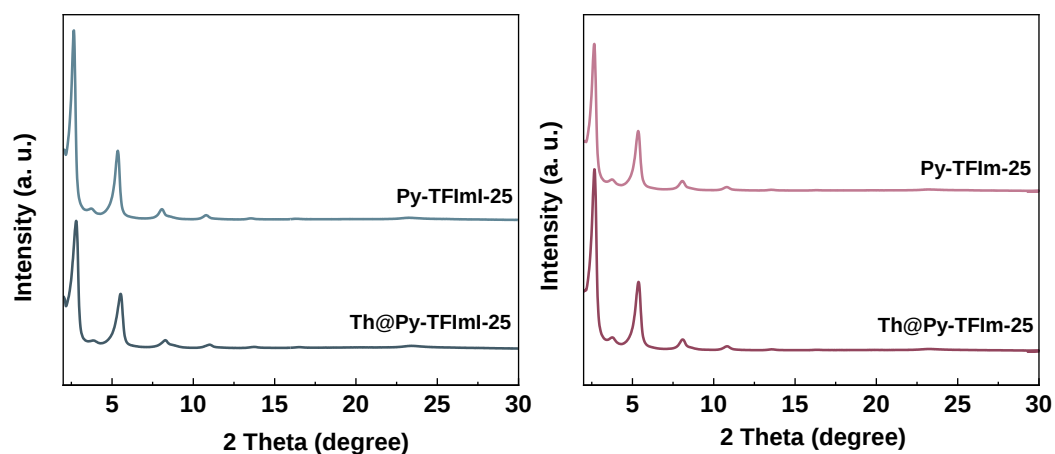
**Supplementary Fig. 19. Adsorption performance.** Effect of different Th(IV) concentration on the capture capacity of Py-TFImI-25 COF and Py-TFIm-25 COF.



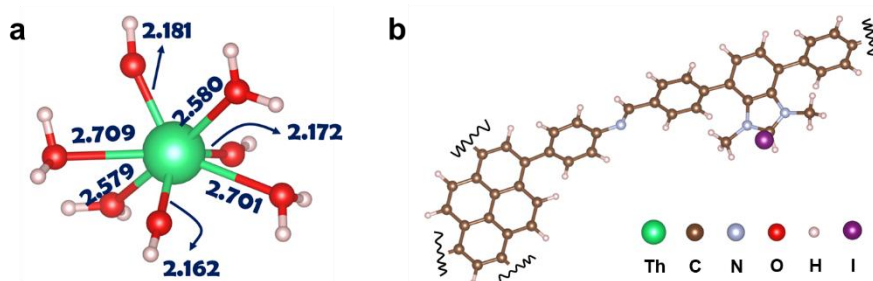
**Supplementary Fig. 20. Adsorption performance.** Comparison of Th(IV) uptake capacity of prepared COF materials with the various materials for Th(IV) capture.



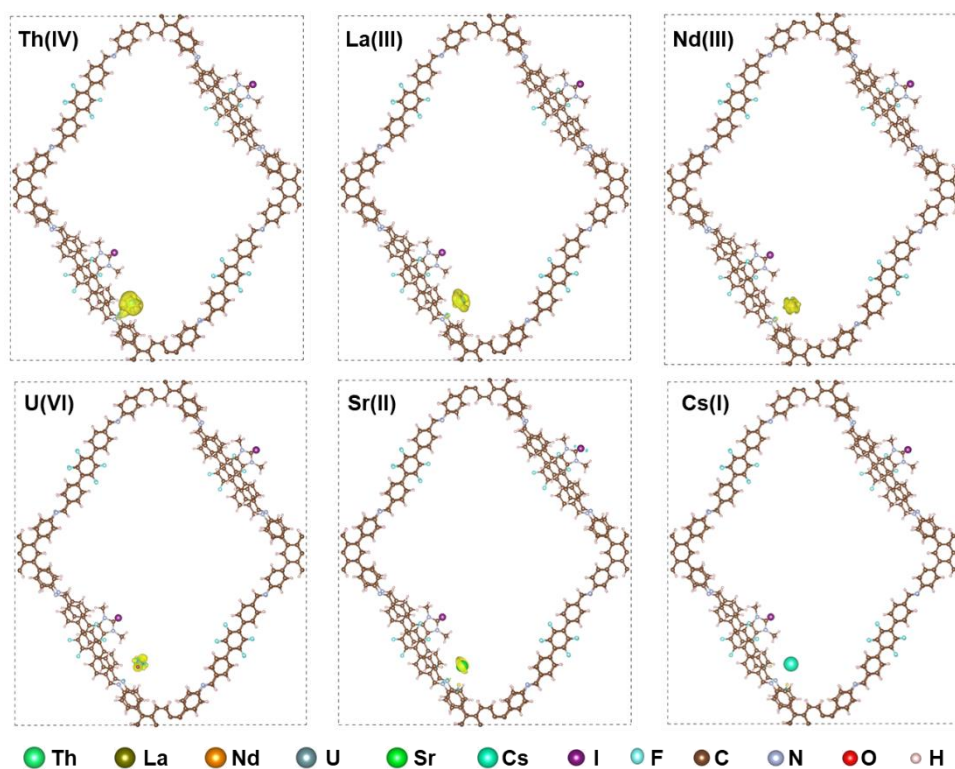
**Supplementary Fig. 21. Adsorption performance.** Comparison of the equilibrium time of prepared COF materials with the various materials for Th(IV) capture.



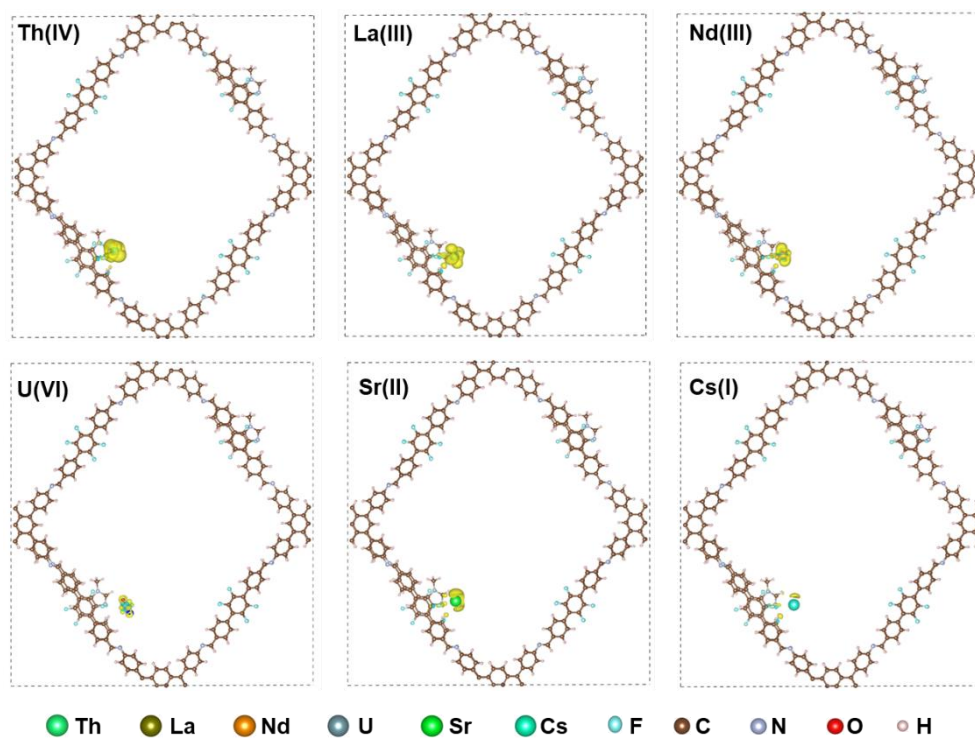
**Supplementary Fig. 22. COF characterization.** PXRD patterns of Py-TFImI-25 COF and Py-TFIm-25 COF before and after Th(IV) adsorption.



**Supplementary Fig. 23. DFT calculations.** The optimized structures of the most energetically stable of **a**  $[\text{Th}(\text{OH})_3(\text{H}_2\text{O})_4]^+$  and **b** Py-TFImI-25 COF framework.



**Supplementary Fig. 24. DFT calculations.** Adsorption configuration of Py-TFImI-25 COF towards different ions.



**Supplementary Fig. 25. DFT calculations.** Adsorption configuration of Py-TFImI-25 COF towards different ions.

### 3. Supplementary Tables 1-6

**Supplementary Table 1.** Fractional atomic coordinates in the refined unit cell of Py-TFImI-25 COF

| Py-TFImI-25 COF: Space group: P1                                            |         |          |         |      |         |         |         |
|-----------------------------------------------------------------------------|---------|----------|---------|------|---------|---------|---------|
| a = 43.30 Å, b = 7.94 Å, c = 50.22 Å                                        |         |          |         |      |         |         |         |
| $\alpha = 87.41^\circ$ , $\beta = 90.59^\circ$ , and $\gamma = 81.35^\circ$ |         |          |         |      |         |         |         |
| Atom                                                                        | x       | y        | z       | Atom | x       | y       | z       |
| H1                                                                          | 0.27703 | 0.37074  | 0.65689 | H280 | 0.74545 | 0.97106 | 0.17251 |
| H2                                                                          | 0.30721 | 0.20104  | 0.64569 | H281 | 0.77154 | 0.79408 | 0.1582  |
| C3                                                                          | 0.72402 | 0.10942  | 0.85292 | C282 | 0.72112 | 0.62406 | 0.85121 |
| C4                                                                          | 0.69329 | 0.19825  | 0.85617 | C283 | 0.69033 | 0.71358 | 0.85388 |
| C5                                                                          | 0.67818 | 0.29346  | 0.83415 | C284 | 0.67551 | 0.80462 | 0.83139 |
| C6                                                                          | 0.56748 | 0.27749  | 0.97846 | C285 | 0.56438 | 0.786   | 0.97603 |
| C7                                                                          | 0.53517 | 0.25531  | 0.97692 | C286 | 0.53252 | 0.75427 | 0.97556 |
| C8                                                                          | 0.52008 | 0.24464  | 0.95215 | C287 | 0.51697 | 0.73334 | 0.95138 |
| C9                                                                          | 0.58884 | 0.27637  | 0.95458 | C288 | 0.58516 | 0.78704 | 0.95178 |
| C10                                                                         | 0.43361 | 0.06352  | 0.92539 | C289 | 0.42423 | 0.58016 | 0.92767 |
| C11                                                                         | 0.41688 | 0.06287  | 0.90131 | C290 | 0.40451 | 0.59857 | 0.90508 |
| C12                                                                         | 0.38676 | 0.15956  | 0.89807 | C291 | 0.37584 | 0.71128 | 0.9049  |
| C13                                                                         | 0.37324 | 0.25445  | 0.91983 | C292 | 0.36731 | 0.80814 | 0.92771 |
| C14                                                                         | 0.39029 | 0.2574   | 0.94373 | C293 | 0.38746 | 0.79312 | 0.94995 |
| N15                                                                         | 0.64879 | 0.26945  | 0.88576 | N294 | 0.64518 | 0.78571 | 0.88295 |
| C16                                                                         | 0.67728 | 0.18828  | 0.88243 | C295 | 0.67404 | 0.70783 | 0.88006 |
| H17                                                                         | 0.53207 | 0.25246  | 0.9332  | H296 | 0.52838 | 0.73872 | 0.93211 |
| H18                                                                         | 0.45663 | -0.01347 | 0.92714 | H297 | 0.44549 | 0.48711 | 0.92756 |
| H19                                                                         | 0.42742 | -0.01403 | 0.88507 | H298 | 0.41128 | 0.52185 | 0.88778 |
| H20                                                                         | 0.34975 | 0.32763  | 0.91871 | H299 | 0.34568 | 0.89859 | 0.92845 |
| H21                                                                         | 0.37963 | 0.33569  | 0.95979 | H300 | 0.38064 | 0.87181 | 0.96704 |
| C22                                                                         | 0.30597 | 0.23569  | 0.78538 | C301 | 0.27974 | 0.80093 | 0.80252 |
| C23                                                                         | 0.29032 | 0.23185  | 0.75868 | C302 | 0.77027 | 0.79557 | 0.27198 |
| C24                                                                         | 0.25733 | 0.23073  | 0.75877 | C303 | 0.73746 | 0.79506 | 0.27504 |
| C25                                                                         | 0.33172 | 0.113    | 0.79388 | C304 | 0.31134 | 0.72114 | 0.80549 |
| C26                                                                         | 0.2917  | 0.35099  | 0.80432 | C305 | 0.26472 | 0.88946 | 0.82419 |
| C27                                                                         | 0.30522 | 0.23437  | 0.73303 | C306 | 0.78187 | 0.80531 | 0.24517 |
| H28                                                                         | 0.36554 | 0.02572  | 0.82515 | H307 | 0.35141 | 0.65932 | 0.83148 |
| H29                                                                         | 0.29231 | 0.44241  | 0.84428 | H308 | 0.26871 | 0.96205 | 0.86508 |
| H30                                                                         | 0.34123 | 0.01048  | 0.78109 | H309 | 0.32348 | 0.6505  | 0.78925 |
| H31                                                                         | 0.27075 | 0.44104  | 0.79904 | H310 | 0.24068 | 0.95594 | 0.8222  |
| H32                                                                         | 0.24469 | 0.22513  | 0.77764 | H311 | 0.72732 | 0.78331 | 0.29491 |
| N33                                                                         | 0.33774 | 0.23673  | 0.72556 | N312 | 0.81345 | 0.80361 | 0.23485 |
| H34                                                                         | 0.6898  | 0.11204  | 0.89865 | H313 | 0.68668 | 0.63559 | 0.89657 |
| C35                                                                         | 0.16878 | 0.33588  | 0.32553 | C314 | 0.17109 | 0.82662 | 0.32863 |
| C36                                                                         | 0.1842  | 0.24542  | 0.34782 | C315 | 0.18666 | 0.73092 | 0.35041 |
| C37                                                                         | 0.21514 | 0.15883  | 0.34476 | C316 | 0.21744 | 0.64373 | 0.34679 |
| C38                                                                         | 0.06288 | 0.30024  | 0.47331 | C317 | 0.06522 | 0.79207 | 0.47565 |
| C39                                                                         | 0.03109 | 0.26727  | 0.47402 | C318 | 0.03302 | 0.7669  | 0.47557 |

|     |         |         |         |      |         |         |         |
|-----|---------|---------|---------|------|---------|---------|---------|
| C40 | 0.01415 | 0.24994 | 0.45041 | C319 | 0.01636 | 0.7587  | 0.45152 |
| C41 | 0.08236 | 0.30316 | 0.44828 | C320 | 0.08519 | 0.793   | 0.45094 |
| C42 | 0.92023 | 0.09517 | 0.42972 | C321 | 0.92798 | 0.5802  | 0.42853 |
| C43 | 0.89915 | 0.11775 | 0.40796 | C322 | 0.90943 | 0.58685 | 0.40535 |
| C44 | 0.87054 | 0.23064 | 0.40926 | C323 | 0.87947 | 0.68697 | 0.40409 |
| C45 | 0.86345 | 0.3224  | 0.43283 | C324 | 0.86796 | 0.77793 | 0.42686 |
| C46 | 0.88487 | 0.30278 | 0.45425 | C325 | 0.88685 | 0.77412 | 0.4498  |
| N47 | 0.13938 | 0.30975 | 0.37762 | N326 | 0.14244 | 0.79524 | 0.38055 |
| C48 | 0.16852 | 0.23749 | 0.37425 | C327 | 0.17119 | 0.71871 | 0.37683 |
| H49 | 0.02441 | 0.25939 | 0.43071 | H328 | 0.02701 | 0.77049 | 0.43205 |
| H50 | 0.94149 | 0.00272 | 0.4284  | H329 | 0.95082 | 0.50066 | 0.42885 |
| H51 | 0.90488 | 0.04458 | 0.39012 | H330 | 0.91839 | 0.51288 | 0.38828 |
| H52 | 0.84199 | 0.41282 | 0.43482 | H331 | 0.84462 | 0.85284 | 0.42729 |
| H53 | 0.87906 | 0.3779  | 0.47189 | H332 | 0.87775 | 0.84945 | 0.46669 |
| C54 | 0.76585 | 0.35065 | 0.31232 | C333 | 0.78899 | 0.79481 | 0.29715 |
| C55 | 0.7461  | 0.34808 | 0.28781 | C334 | 0.26287 | 0.79083 | 0.77673 |
| C56 | 0.71536 | 0.30543 | 0.28939 | C335 | 0.23209 | 0.74842 | 0.77608 |
| C57 | 0.75311 | 0.43103 | 0.33552 | C336 | 0.77713 | 0.90799 | 0.31723 |
| C58 | 0.79771 | 0.2719  | 0.31264 | C337 | 0.8152  | 0.66953 | 0.30343 |
| C59 | 0.75816 | 0.38697 | 0.26246 | C338 | 0.27819 | 0.81799 | 0.75234 |
| H60 | 0.76104 | 0.48957 | 0.3765  | H339 | 0.7822  | 0.99277 | 0.35711 |
| H61 | 0.84031 | 0.20359 | 0.33564 | H340 | 0.85189 | 0.57484 | 0.33197 |
| H62 | 0.72893 | 0.4968  | 0.33562 | H341 | 0.75591 | 0.9998  | 0.3137  |
| H63 | 0.80816 | 0.20765 | 0.29515 | H342 | 0.82297 | 0.56872 | 0.28981 |
| F64 | 0.70333 | 0.25914 | 0.31339 | F343 | 0.217   | 0.71381 | 0.79924 |
| F65 | 0.78694 | 0.43703 | 0.26035 | F344 | 0.30714 | 0.86683 | 0.75221 |
| H66 | 0.18203 | 0.16899 | 0.39066 | H345 | 0.18442 | 0.6448  | 0.39291 |
| C67 | 0.27825 | 0.34244 | 0.1467  | C346 | 0.27869 | 0.83199 | 0.14865 |
| C68 | 0.30887 | 0.25151 | 0.14367 | C347 | 0.3095  | 0.74298 | 0.14594 |
| C69 | 0.32363 | 0.1565  | 0.16586 | C348 | 0.3245  | 0.65271 | 0.16842 |
| C70 | 0.43548 | 0.16944 | 0.02182 | C349 | 0.43544 | 0.65929 | 0.02399 |
| C71 | 0.46759 | 0.1957  | 0.02337 | C350 | 0.46735 | 0.68953 | 0.02442 |
| C72 | 0.48243 | 0.21065 | 0.04811 | C351 | 0.4829  | 0.70946 | 0.04859 |
| C73 | 0.41398 | 0.16953 | 0.04563 | C352 | 0.41463 | 0.66182 | 0.04823 |
| C74 | 0.61277 | 0.19219 | 0.05673 | C353 | 0.61271 | 0.67892 | 0.05162 |
| C75 | 0.63016 | 0.20191 | 0.08029 | C354 | 0.63185 | 0.67879 | 0.07433 |
| C76 | 0.61739 | 0.30381 | 0.10132 | C355 | 0.62131 | 0.77787 | 0.09627 |
| C77 | 0.58687 | 0.3991  | 0.09806 | C356 | 0.59126 | 0.88009 | 0.09492 |
| C78 | 0.56981 | 0.39126 | 0.07435 | C357 | 0.57261 | 0.88349 | 0.07192 |
| N79 | 0.35355 | 0.17717 | 0.11425 | N358 | 0.35453 | 0.67002 | 0.11696 |
| C80 | 0.32511 | 0.25952 | 0.11746 | C359 | 0.3257  | 0.74804 | 0.11974 |
| H81 | 0.47037 | 0.20307 | 0.06703 | H360 | 0.47154 | 0.70326 | 0.06787 |
| H82 | 0.62316 | 0.10921 | 0.04114 | H361 | 0.62129 | 0.59812 | 0.03518 |
| H83 | 0.65362 | 0.12793 | 0.08231 | H362 | 0.65489 | 0.59934 | 0.07498 |
| H84 | 0.57609 | 0.48345 | 0.11322 | H363 | 0.58206 | 0.96132 | 0.111   |
| H85 | 0.54692 | 0.46866 | 0.07238 | H364 | 0.55027 | 0.96718 | 0.07118 |
| C86 | 0.68867 | 0.36353 | 0.21623 | C365 | 0.70545 | 0.82763 | 0.20402 |
| C87 | 0.70893 | 0.34995 | 0.24073 | C366 | 0.23259 | 0.76364 | 0.72698 |

|      |         |         |         |      |         |         |         |
|------|---------|---------|---------|------|---------|---------|---------|
| C88  | 0.74015 | 0.38502 | 0.23918 | C367 | 0.26349 | 0.80289 | 0.72784 |
| C89  | 0.65741 | 0.45023 | 0.21684 | C368 | 0.67591 | 0.93299 | 0.20587 |
| C90  | 0.70036 | 0.29156 | 0.19205 | C369 | 0.7114  | 0.7229  | 0.18178 |
| C91  | 0.69694 | 0.30674 | 0.26611 | C370 | 0.21706 | 0.73518 | 0.7514  |
| H92  | 0.61388 | 0.52549 | 0.1953  | H371 | 0.63105 | 1.02332 | 0.18732 |
| H93  | 0.69042 | 0.24051 | 0.15113 | H372 | 0.69463 | 0.65169 | 0.14391 |
| H94  | 0.64804 | 0.512   | 0.23489 | H373 | 0.67034 | 1.01327 | 0.22297 |
| H95  | 0.72435 | 0.22497 | 0.19099 | H374 | 0.73338 | 0.63594 | 0.18016 |
| F96  | 0.75301 | 0.42479 | 0.21526 | F375 | 0.27935 | 0.83273 | 0.70495 |
| F97  | 0.66761 | 0.26185 | 0.26833 | F376 | 0.18779 | 0.68875 | 0.75141 |
| H98  | 0.3128  | 0.3362  | 0.10119 | H377 | 0.31303 | 0.81907 | 0.10312 |
| C99  | 0.83168 | 0.12441 | 0.67366 | C378 | 0.83025 | 0.62918 | 0.67181 |
| C100 | 0.81614 | 0.21591 | 0.65151 | C379 | 0.81488 | 0.7243  | 0.6499  |
| C101 | 0.78521 | 0.30223 | 0.65473 | C380 | 0.78407 | 0.81168 | 0.65331 |
| C102 | 0.93726 | 0.15836 | 0.52599 | C381 | 0.93705 | 0.66525 | 0.52498 |
| C103 | 0.9691  | 0.19011 | 0.52526 | C382 | 0.96913 | 0.69292 | 0.5251  |
| C104 | 0.98606 | 0.20673 | 0.54886 | C383 | 0.98565 | 0.70367 | 0.54915 |
| C105 | 0.9178  | 0.15734 | 0.55103 | C384 | 0.91698 | 0.66312 | 0.54965 |
| C106 | 0.07844 | 0.37341 | 0.56941 | C385 | 0.0743  | 0.88587 | 0.57162 |
| C107 | 0.09946 | 0.36217 | 0.5911  | C386 | 0.09305 | 0.88532 | 0.59454 |
| C108 | 0.12908 | 0.25853 | 0.59014 | C387 | 0.1233  | 0.78857 | 0.59589 |
| C109 | 0.13717 | 0.16301 | 0.56708 | C388 | 0.13452 | 0.69009 | 0.57381 |
| C110 | 0.11577 | 0.17108 | 0.54572 | C389 | 0.11544 | 0.68831 | 0.55108 |
| N111 | 0.86084 | 0.15262 | 0.62167 | N390 | 0.85935 | 0.65936 | 0.61989 |
| C112 | 0.83172 | 0.22499 | 0.62507 | C391 | 0.83055 | 0.73588 | 0.62352 |
| H113 | 0.97585 | 0.19673 | 0.56856 | H392 | 0.97495 | 0.69212 | 0.5686  |
| H114 | 0.05642 | 0.45967 | 0.57046 | H393 | 0.0514  | 0.9646  | 0.57113 |
| H115 | 0.09298 | 0.43845 | 0.6085  | H394 | 0.08413 | 0.96298 | 0.61123 |
| H116 | 0.15966 | 0.08112 | 0.56521 | H395 | 0.15749 | 0.61097 | 0.57403 |
| H117 | 0.12246 | 0.09438 | 0.52839 | H396 | 0.12435 | 0.60839 | 0.53463 |
| C118 | 0.2355  | 0.21826 | 0.68514 | C397 | 0.21661 | 0.75538 | 0.70074 |
| C119 | 0.25485 | 0.23332 | 0.70957 | C398 | 0.72852 | 0.82044 | 0.22644 |
| C120 | 0.28773 | 0.24114 | 0.70942 | C399 | 0.7614  | 0.82238 | 0.2233  |
| C121 | 0.24649 | 0.10462 | 0.66487 | C400 | 0.23274 | 0.67372 | 0.67906 |
| C122 | 0.20464 | 0.31032 | 0.68288 | C401 | 0.18344 | 0.83785 | 0.69649 |
| C123 | 0.24012 | 0.22993 | 0.7349  | C402 | 0.71718 | 0.80607 | 0.25289 |
| H124 | 0.23709 | 0.00323 | 0.62683 | H403 | 0.2308  | 0.60225 | 0.63808 |
| H125 | 0.16238 | 0.36948 | 0.65922 | H404 | 0.14471 | 0.88874 | 0.66886 |
| H126 | 0.2697  | 0.0281  | 0.66623 | H405 | 0.25694 | 0.61102 | 0.6814  |
| H127 | 0.19521 | 0.3975  | 0.69827 | H406 | 0.17039 | 0.90786 | 0.71234 |
| N128 | 0.30886 | 0.2495  | 0.68713 | N407 | 0.77974 | 0.83232 | 0.19925 |
| H129 | 0.21515 | 0.22081 | 0.73645 | H408 | 0.69238 | 0.79986 | 0.25658 |
| H130 | 0.81822 | 0.29397 | 0.6087  | H409 | 0.81741 | 0.80977 | 0.6074  |
| C131 | 0.51745 | 0.24105 | 0.0009  | C410 | 0.51586 | 0.73924 | 0.00014 |
| C132 | 0.58096 | 0.29205 | 0.00371 | C411 | 0.57851 | 0.80462 | 0.00084 |
| H133 | 0.60534 | 0.31416 | 0.00462 | H412 | 0.60261 | 0.83182 | 0.00117 |
| C134 | 0.01596 | 0.24696 | 0.49921 | C413 | 0.01707 | 0.74668 | 0.50034 |
| C135 | 0.07841 | 0.31566 | 0.49761 | C414 | 0.08022 | 0.80481 | 0.50027 |

|      |         |         |         |      |         |         |         |
|------|---------|---------|---------|------|---------|---------|---------|
| H136 | 0.1025  | 0.34316 | 0.49703 | H415 | 0.10452 | 0.8291  | 0.50016 |
| C137 | 0.33791 | 0.24008 | 0.69828 | C416 | 0.81015 | 0.8137  | 0.20771 |
| H138 | 0.35915 | 0.24454 | 0.68645 | H417 | 0.82991 | 0.81675 | 0.19406 |
| C139 | 0.30197 | 0.30984 | 0.65932 | C418 | 0.76984 | 0.90035 | 0.17236 |
| C140 | 0.30396 | 0.35142 | 0.83027 | C419 | 0.28077 | 0.89378 | 0.84857 |
| C141 | 0.33136 | 0.2395  | 0.83786 | C420 | 0.31201 | 0.80956 | 0.85167 |
| C142 | 0.34506 | 0.11857 | 0.81951 | C421 | 0.32727 | 0.72481 | 0.82979 |
| C143 | 0.43884 | 0.17043 | 0.97251 | C422 | 0.43652 | 0.66353 | 0.97476 |
| C144 | 0.47082 | 0.20005 | 0.97393 | C423 | 0.46869 | 0.68924 | 0.97494 |
| C145 | 0.48861 | 0.22312 | 0.95069 | C424 | 0.48583 | 0.70401 | 0.95107 |
| C146 | 0.42104 | 0.16473 | 0.94669 | C425 | 0.4163  | 0.67991 | 0.95016 |
| C147 | 0.58097 | 0.38641 | 0.93203 | C426 | 0.57606 | 0.88995 | 0.92866 |
| C148 | 0.6008  | 0.38043 | 0.90963 | C427 | 0.59608 | 0.8875  | 0.90633 |
| C149 | 0.62971 | 0.27032 | 0.90964 | C428 | 0.62605 | 0.78666 | 0.90684 |
| C150 | 0.63844 | 0.16383 | 0.93259 | C429 | 0.63579 | 0.68801 | 0.93034 |
| C151 | 0.61809 | 0.16677 | 0.95475 | C430 | 0.61552 | 0.68818 | 0.95243 |
| N152 | 0.37089 | 0.15549 | 0.87263 | N431 | 0.35663 | 0.72422 | 0.88108 |
| C153 | 0.34492 | 0.25276 | 0.86488 | C432 | 0.3284  | 0.81058 | 0.87768 |
| H154 | 0.47876 | 0.22606 | 0.93077 | H433 | 0.47562 | 0.69479 | 0.93154 |
| H155 | 0.55933 | 0.47641 | 0.9316  | H434 | 0.55338 | 0.97123 | 0.92769 |
| H156 | 0.59383 | 0.46437 | 0.89226 | H435 | 0.5883  | 0.9657  | 0.88855 |
| H157 | 0.66032 | 0.07497 | 0.93345 | H436 | 0.65849 | 0.60672 | 0.93179 |
| H158 | 0.62495 | 0.08075 | 0.9719  | H437 | 0.62333 | 0.60733 | 0.96995 |
| C159 | 0.72409 | 0.20737 | 0.80595 | C438 | 0.72183 | 0.71302 | 0.80388 |
| C160 | 0.7399  | 0.20839 | 0.77934 | C439 | 0.73808 | 0.70991 | 0.77742 |
| C161 | 0.77104 | 0.24438 | 0.77702 | C440 | 0.76934 | 0.74433 | 0.77528 |
| C162 | 0.69345 | 0.29786 | 0.80929 | C441 | 0.69113 | 0.80427 | 0.80665 |
| C163 | 0.73932 | 0.11423 | 0.82807 | C442 | 0.73674 | 0.62391 | 0.82647 |
| C164 | 0.72393 | 0.17291 | 0.75601 | C443 | 0.72234 | 0.67348 | 0.75402 |
| H165 | 0.65455 | 0.36449 | 0.83607 | H444 | 0.65185 | 0.87598 | 0.83288 |
| H166 | 0.7361  | 0.0347  | 0.86957 | H445 | 0.73298 | 0.55232 | 0.86823 |
| H167 | 0.6814  | 0.37309 | 0.7927  | H446 | 0.67927 | 0.87625 | 0.78968 |
| H168 | 0.76286 | 0.0427  | 0.82602 | H447 | 0.76027 | 0.55152 | 0.82487 |
| F169 | 0.78692 | 0.28284 | 0.79901 | F448 | 0.78512 | 0.78277 | 0.79732 |
| F170 | 0.69419 | 0.13314 | 0.75757 | F449 | 0.6926  | 0.63338 | 0.75545 |
| H171 | 0.33317 | 0.35075 | 0.87763 | H450 | 0.3162  | 0.88201 | 0.89374 |
| C172 | 0.81606 | 0.2682  | 0.33591 | C451 | 0.83117 | 0.66982 | 0.32799 |
| C173 | 0.80296 | 0.34413 | 0.35935 | C452 | 0.81975 | 0.78823 | 0.34749 |
| C174 | 0.7715  | 0.42779 | 0.3588  | C453 | 0.79207 | 0.90353 | 0.34214 |
| C175 | 0.93542 | 0.17039 | 0.4767  | C454 | 0.93704 | 0.67571 | 0.47563 |
| C176 | 0.96747 | 0.19839 | 0.47572 | C455 | 0.96892 | 0.70677 | 0.47557 |
| C177 | 0.98312 | 0.21856 | 0.45124 | C456 | 0.98498 | 0.73472 | 0.45153 |
| C178 | 0.9137  | 0.19031 | 0.45292 | C457 | 0.91738 | 0.67762 | 0.45078 |
| C179 | 0.07224 | 0.40854 | 0.42564 | C458 | 0.07611 | 0.90361 | 0.42868 |
| C180 | 0.09121 | 0.40815 | 0.40263 | C459 | 0.095   | 0.90035 | 0.40569 |
| C181 | 0.12114 | 0.30717 | 0.40201 | C460 | 0.12413 | 0.79246 | 0.40483 |
| C182 | 0.13179 | 0.20501 | 0.42495 | C461 | 0.13393 | 0.68471 | 0.42741 |
| C183 | 0.11257 | 0.20311 | 0.4477  | C462 | 0.11456 | 0.68492 | 0.45013 |

|      |         |          |         |      |         |         |         |
|------|---------|----------|---------|------|---------|---------|---------|
| N184 | 0.8499  | 0.24946  | 0.38617 | N463 | 0.86156 | 0.69012 | 0.37959 |
| C185 | 0.82152 | 0.3369   | 0.38447 | C464 | 0.83589 | 0.79502 | 0.37349 |
| H186 | 0.97181 | 0.21149  | 0.4321  | H465 | 0.97381 | 0.73921 | 0.43212 |
| H187 | 0.04965 | 0.49072  | 0.42561 | H466 | 0.05434 | 0.99249 | 0.42896 |
| H188 | 0.0827  | 0.48873  | 0.38528 | H467 | 0.0872  | 0.98512 | 0.38861 |
| H189 | 0.15437 | 0.12231  | 0.42546 | H468 | 0.15593 | 0.5968  | 0.4276  |
| H190 | 0.1211  | 0.11989  | 0.46478 | H469 | 0.12231 | 0.59813 | 0.467   |
| C191 | 0.21493 | 0.24985  | 0.29747 | C470 | 0.21672 | 0.74436 | 0.29995 |
| C192 | 0.23096 | 0.2493   | 0.27089 | C471 | 0.23226 | 0.74763 | 0.27327 |
| C193 | 0.26223 | 0.28369  | 0.26871 | C472 | 0.26334 | 0.78429 | 0.27091 |
| C194 | 0.23036 | 0.16106  | 0.31987 | C473 | 0.23235 | 0.65056 | 0.32182 |
| C195 | 0.18404 | 0.33812  | 0.30061 | C474 | 0.18601 | 0.83324 | 0.30364 |
| C196 | 0.21517 | 0.21327  | 0.24747 | C475 | 0.21626 | 0.71186 | 0.24997 |
| H197 | 0.22746 | 0.08759  | 0.36161 | H476 | 0.22985 | 0.56866 | 0.36325 |
| H198 | 0.14499 | 0.40508  | 0.32733 | H477 | 0.14741 | 0.89646 | 0.33087 |
| H199 | 0.25404 | 0.09095  | 0.31805 | H478 | 0.25594 | 0.5802  | 0.3195  |
| H200 | 0.17177 | 0.40995  | 0.28381 | H479 | 0.17367 | 0.90896 | 0.28722 |
| F201 | 0.27806 | 0.32211  | 0.29072 | F480 | 0.27929 | 0.82253 | 0.29287 |
| F202 | 0.18545 | 0.17299  | 0.24891 | F481 | 0.18658 | 0.67127 | 0.25159 |
| H203 | 0.81054 | 0.40334  | 0.4015  | H482 | 0.82624 | 0.89406 | 0.38695 |
| C204 | 0.6809  | 0.30018  | 0.16929 | C483 | 0.68912 | 0.73138 | 0.16108 |
| C205 | 0.64932 | 0.38222  | 0.17029 | C484 | 0.66    | 0.84053 | 0.16275 |
| C206 | 0.63796 | 0.45909  | 0.19415 | C485 | 0.65343 | 0.93906 | 0.1855  |
| C207 | 0.56417 | 0.27843  | 0.02776 | C486 | 0.56312 | 0.7838  | 0.02531 |
| C208 | 0.53189 | 0.25322  | 0.02636 | C487 | 0.5311  | 0.75535 | 0.02507 |
| C209 | 0.51371 | 0.23584  | 0.04957 | C488 | 0.51396 | 0.7392  | 0.04892 |
| C210 | 0.58207 | 0.28547  | 0.05353 | C489 | 0.58264 | 0.78063 | 0.05021 |
| C211 | 0.42206 | 0.0622   | 0.06845 | C490 | 0.42332 | 0.55811 | 0.07133 |
| C212 | 0.40201 | 0.0678   | 0.09074 | C491 | 0.4033  | 0.56321 | 0.09365 |
| C213 | 0.37277 | 0.17532  | 0.09041 | C492 | 0.3737  | 0.66749 | 0.09312 |
| C214 | 0.36387 | 0.27859  | 0.06716 | C493 | 0.3644  | 0.76801 | 0.06971 |
| C215 | 0.38434 | 0.27568  | 0.04509 | C494 | 0.38472 | 0.76528 | 0.04762 |
| N216 | 0.63638 | 0.30897  | 0.12511 | N495 | 0.64204 | 0.77251 | 0.11908 |
| C217 | 0.62766 | 0.38835  | 0.14703 | C496 | 0.63621 | 0.85492 | 0.14112 |
| H218 | 0.52325 | 0.2379   | 0.06953 | H497 | 0.52406 | 0.74687 | 0.06851 |
| H219 | 0.44406 | -0.02487 | 0.06923 | H498 | 0.44567 | 0.47403 | 0.07228 |
| H220 | 0.40913 | -0.01387 | 0.10832 | H499 | 0.41075 | 0.48403 | 0.11141 |
| H221 | 0.34171 | 0.36502  | 0.06594 | H500 | 0.34204 | 0.85256 | 0.06837 |
| H222 | 0.37729 | 0.35949  | 0.02773 | H501 | 0.37729 | 0.84716 | 0.03012 |
| C223 | 0.27758 | 0.24668  | 0.19376 | C502 | 0.27831 | 0.74405 | 0.19603 |
| C224 | 0.26148 | 0.24749  | 0.22028 | C503 | 0.26226 | 0.7474  | 0.22254 |
| C225 | 0.23021 | 0.21324  | 0.22242 | C504 | 0.2311  | 0.71173 | 0.22486 |
| C226 | 0.30812 | 0.15424  | 0.19064 | C505 | 0.30902 | 0.65329 | 0.19321 |
| C227 | 0.26273 | 0.33985  | 0.17148 | C506 | 0.26321 | 0.83233 | 0.17345 |
| C228 | 0.2773  | 0.28278  | 0.2437  | C507 | 0.27813 | 0.78494 | 0.2458  |
| H229 | 0.34716 | 0.08399  | 0.16411 | H508 | 0.34818 | 0.58169 | 0.1669  |
| H230 | 0.26645 | 0.41721  | 0.12994 | H509 | 0.26671 | 0.90302 | 0.13162 |
| H231 | 0.3199  | 0.07915  | 0.20736 | H510 | 0.32101 | 0.58188 | 0.21017 |

|      |         |          |          |      |         |         |          |
|------|---------|----------|----------|------|---------|---------|----------|
| H232 | 0.23931 | 0.41323  | 0.17336  | H511 | 0.23963 | 0.90406 | 0.17509  |
| F233 | 0.21441 | 0.17547  | 0.20035  | F512 | 0.21529 | 0.67158 | 0.20295  |
| F234 | 0.30716 | 0.32101  | 0.2423   | F513 | 0.30779 | 0.82594 | 0.24418  |
| H235 | 0.60404 | 0.45807  | 0.14912  | H514 | 0.61405 | 0.93813 | 0.14407  |
| C236 | 0.18583 | 0.2947   | 0.66047  | C515 | 0.16904 | 0.82818 | 0.67161  |
| C237 | 0.19738 | 0.18535  | 0.63984  | C516 | 0.18593 | 0.74254 | 0.65046  |
| C238 | 0.2278  | 0.08999  | 0.64233  | C517 | 0.21766 | 0.66814 | 0.6543   |
| C239 | 0.06464 | 0.28874  | 0.52264  | C518 | 0.06522 | 0.784   | 0.52498  |
| C240 | 0.03268 | 0.25915  | 0.52358  | C519 | 0.03319 | 0.75502 | 0.5251   |
| C241 | 0.01705 | 0.23818  | 0.54805  | C520 | 0.01691 | 0.73036 | 0.54914  |
| C242 | 0.08605 | 0.27607  | 0.54659  | C521 | 0.08487 | 0.78474 | 0.54979  |
| C243 | 0.92774 | 0.05125  | 0.57363  | C522 | 0.92623 | 0.55403 | 0.57207  |
| C244 | 0.90883 | 0.05266  | 0.59665  | C523 | 0.90716 | 0.55636 | 0.59497  |
| C245 | 0.87909 | 0.15533  | 0.59732  | C524 | 0.87777 | 0.66218 | 0.59564  |
| C246 | 0.86864 | 0.259    | 0.57448  | C525 | 0.86783 | 0.76813 | 0.5729   |
| C247 | 0.88785 | 0.26     | 0.55171  | C526 | 0.88731 | 0.76857 | 0.55026  |
| N248 | 0.15016 | 0.25822  | 0.61265  | N527 | 0.14134 | 0.79344 | 0.6201   |
| C249 | 0.17823 | 0.16911  | 0.61568  | C528 | 0.17082 | 0.72843 | 0.62415  |
| H250 | 0.02831 | 0.24464  | 0.5672   | H529 | 0.02789 | 0.72933 | 0.5686   |
| H251 | 0.95014 | -0.03247 | 0.5736   | H530 | 0.94828 | 0.46735 | 0.572    |
| H252 | 0.91718 | -0.02879 | 0.61395  | H531 | 0.91509 | 0.47286 | 0.61216  |
| H253 | 0.84622 | 0.34326  | 0.57408  | H532 | 0.84562 | 0.85426 | 0.57251  |
| H254 | 0.8795  | 0.34402  | 0.53467  | H533 | 0.87941 | 0.85422 | 0.53329  |
| C255 | 0.78572 | 0.2093   | 0.70193  | C534 | 0.78434 | 0.71224 | 0.70021  |
| C256 | 0.76992 | 0.20901  | 0.72858  | C535 | 0.76852 | 0.70952 | 0.72682  |
| C257 | 0.73876 | 0.17332  | 0.73091  | C536 | 0.73733 | 0.67398 | 0.72898  |
| C258 | 0.77014 | 0.29889  | 0.67967  | C537 | 0.76893 | 0.80555 | 0.67819  |
| C259 | 0.81656 | 0.12106  | 0.69862  | C538 | 0.81509 | 0.62325 | 0.69673  |
| C260 | 0.78584 | 0.24533  | 0.7519   | C539 | 0.78437 | 0.74445 | 0.75025  |
| H261 | 0.77281 | 0.37413  | 0.63798  | H540 | 0.7718  | 0.88638 | 0.63675  |
| H262 | 0.85545 | 0.05529  | 0.67173  | H541 | 0.85395 | 0.55926 | 0.66974  |
| H263 | 0.74646 | 0.36875  | 0.68165  | H542 | 0.74532 | 0.87612 | 0.68033  |
| H264 | 0.82891 | 0.04844  | 0.71532  | H543 | 0.8273  | 0.54793 | 0.71324  |
| F265 | 0.72282 | 0.13476  | 0.70897  | F544 | 0.72149 | 0.63658 | 0.70691  |
| F266 | 0.81548 | 0.28665  | 0.75029  | F545 | 0.81411 | 0.78443 | 0.74882  |
| H267 | 0.18832 | 0.08185  | 0.60061  | H546 | 0.18495 | 0.66383 | 0.60842  |
| C268 | 0.48531 | 0.21092  | -0.00059 | C547 | 0.48403 | 0.70424 | -0.00016 |
| C269 | 0.42221 | 0.15282  | 0.99653  | C548 | 0.42121 | 0.6415  | 0.99923  |
| H270 | 0.39802 | 0.12752  | 0.99557  | H549 | 0.39691 | 0.61734 | 0.99892  |
| C271 | 0.98424 | 0.21008  | 0.50006  | C550 | 0.98508 | 0.71392 | 0.50034  |
| C272 | 0.92165 | 0.14348  | 0.50173  | C551 | 0.92216 | 0.65189 | 0.50031  |
| H273 | 0.89742 | 0.1181   | 0.50232  | H552 | 0.89798 | 0.62586 | 0.50035  |
| H274 | 0.31691 | 0.40814  | 0.65336  | H553 | 0.78546 | 0.9922  | 0.16502  |
| C275 | 0.36645 | 0.20571  | 0.74199  | C554 | 0.844   | 0.76384 | 0.24857  |
| H276 | 0.38696 | 0.23861  | 0.73021  | H555 | 0.86325 | 0.79519 | 0.23508  |
| H277 | 0.37207 | 0.06794  | 0.74787  | H556 | 0.84929 | 0.62481 | 0.25362  |
| H278 | 0.36374 | 0.28748  | 0.75976  | H557 | 0.84416 | 0.84172 | 0.26665  |
| I279 | 0.351   | 0.61622  | 0.71854  | I558 | 0.82789 | 1.17942 | 0.22687  |

**Supplementary Table 2.** Fractional atomic coordinates in the refined unit cell of Py-TFIm-25 COF.

| Py-TFIm-25 COF: Space group: P1                                             |         |         |         |      |         |         |         |
|-----------------------------------------------------------------------------|---------|---------|---------|------|---------|---------|---------|
| a = 44.98 Å, b = 7.93 Å, c = 47.37 Å                                        |         |         |         |      |         |         |         |
| $\alpha = 88.52^\circ$ , $\beta = 90.90^\circ$ , and $\gamma = 85.17^\circ$ |         |         |         |      |         |         |         |
| Atom                                                                        | x       | y       | z       | Atom | x       | y       | z       |
| H1                                                                          | 0.26392 | 0.22533 | 0.67173 | H275 | 0.76632 | 0.97148 | 0.16448 |
| H2                                                                          | 0.28491 | 0.02675 | 0.66627 | H276 | 0.79403 | 0.80429 | 0.15693 |
| C3                                                                          | 0.67078 | 0.14401 | 0.82605 | C277 | 0.66933 | 0.64382 | 0.8269  |
| C4                                                                          | 0.6816  | 0.21179 | 0.85058 | C278 | 0.68015 | 0.7125  | 0.85136 |
| C5                                                                          | 0.71073 | 0.26219 | 0.85044 | C279 | 0.70921 | 0.76394 | 0.85111 |
| C6                                                                          | 0.56201 | 0.28993 | 0.97242 | C280 | 0.561   | 0.79028 | 0.97351 |
| C7                                                                          | 0.53035 | 0.28767 | 0.97292 | C281 | 0.52935 | 0.78857 | 0.97426 |
| C8                                                                          | 0.51359 | 0.25878 | 0.94861 | C282 | 0.51239 | 0.76051 | 0.95005 |
| C9                                                                          | 0.5804  | 0.27013 | 0.94709 | C283 | 0.57923 | 0.77045 | 0.94808 |
| C10                                                                         | 0.42472 | 0.12382 | 0.92999 | C284 | 0.42315 | 0.63177 | 0.93162 |
| C11                                                                         | 0.4065  | 0.10376 | 0.90724 | C285 | 0.4048  | 0.61424 | 0.90886 |
| C12                                                                         | 0.37836 | 0.19369 | 0.90508 | C286 | 0.37679 | 0.70565 | 0.90697 |
| C13                                                                         | 0.36919 | 0.30464 | 0.92662 | C287 | 0.36789 | 0.81537 | 0.92875 |
| C14                                                                         | 0.38782 | 0.32664 | 0.949   | C288 | 0.38668 | 0.83494 | 0.95111 |
| N15                                                                         | 0.63512 | 0.21685 | 0.87544 | N289 | 0.63373 | 0.71714 | 0.87629 |
| C16                                                                         | 0.66335 | 0.2348  | 0.87566 | C290 | 0.66194 | 0.7355  | 0.87646 |
| H17                                                                         | 0.52518 | 0.23697 | 0.92885 | H291 | 0.52382 | 0.73849 | 0.93021 |
| H18                                                                         | 0.44562 | 0.04392 | 0.93177 | H292 | 0.44389 | 0.55032 | 0.9332  |
| H19                                                                         | 0.41361 | 0.0126  | 0.89114 | H293 | 0.41171 | 0.52381 | 0.8926  |
| H20                                                                         | 0.34753 | 0.37765 | 0.92529 | H294 | 0.3463  | 0.88907 | 0.92764 |
| H21                                                                         | 0.38046 | 0.41825 | 0.96498 | H295 | 0.37952 | 0.92582 | 0.96727 |
| C22                                                                         | 0.28351 | 0.16107 | 0.80472 | C296 | 0.27883 | 0.67638 | 0.80864 |
| C23                                                                         | 0.26713 | 0.15414 | 0.77838 | C297 | 0.77404 | 0.78838 | 0.26863 |
| C24                                                                         | 0.23589 | 0.17062 | 0.77688 | C298 | 0.74347 | 0.76718 | 0.2665  |
| C25                                                                         | 0.27049 | 0.12441 | 0.83046 | C299 | 0.26906 | 0.60583 | 0.83361 |
| C26                                                                         | 0.3131  | 0.20609 | 0.80496 | C300 | 0.3065  | 0.74619 | 0.80862 |
| C27                                                                         | 0.28162 | 0.13612 | 0.75238 | C301 | 0.78795 | 0.82731 | 0.24326 |
| H28                                                                         | 0.27602 | 0.09608 | 0.87473 | H302 | 0.27839 | 0.54772 | 0.87668 |
| H29                                                                         | 0.35209 | 0.24398 | 0.82925 | H303 | 0.34536 | 0.79849 | 0.83208 |
| H30                                                                         | 0.24759 | 0.08809 | 0.8312  | H304 | 0.24779 | 0.54876 | 0.83433 |
| H31                                                                         | 0.32356 | 0.23903 | 0.78535 | H305 | 0.3144  | 0.806   | 0.78973 |
| H32                                                                         | 0.22293 | 0.18634 | 0.79603 | H306 | 0.73109 | 0.73613 | 0.28519 |
| N33                                                                         | 0.31196 | 0.1175  | 0.74749 | N307 | 0.8169  | 0.8696  | 0.23951 |
| H34                                                                         | 0.67493 | 0.27756 | 0.8946  | H308 | 0.67353 | 0.77876 | 0.89536 |
| C35                                                                         | 0.16747 | 0.3807  | 0.32645 | C309 | 0.16704 | 0.87695 | 0.32881 |
| C36                                                                         | 0.17756 | 0.26848 | 0.34795 | C310 | 0.17695 | 0.76352 | 0.35021 |
| C37                                                                         | 0.20516 | 0.17563 | 0.34443 | C311 | 0.20448 | 0.67004 | 0.34665 |
| C38                                                                         | 0.06374 | 0.24715 | 0.4744  | C312 | 0.06195 | 0.74875 | 0.4759  |
| C39                                                                         | 0.03228 | 0.23552 | 0.47403 | C313 | 0.03052 | 0.73562 | 0.47518 |
| C40                                                                         | 0.01593 | 0.23332 | 0.44874 | C314 | 0.01452 | 0.73132 | 0.44972 |
| C41                                                                         | 0.08201 | 0.26867 | 0.44935 | C315 | 0.08054 | 0.76863 | 0.45101 |
| C42                                                                         | 0.92805 | 0.09319 | 0.42423 | C316 | 0.92691 | 0.5924  | 0.42462 |

|     |         |         |         |      |         |         |         |
|-----|---------|---------|---------|------|---------|---------|---------|
| C43 | 0.90997 | 0.09288 | 0.40091 | C317 | 0.90934 | 0.59223 | 0.40096 |
| C44 | 0.88246 | 0.19048 | 0.39954 | C318 | 0.88206 | 0.6917  | 0.39912 |
| C45 | 0.87371 | 0.28762 | 0.42295 | C319 | 0.87303 | 0.79134 | 0.42216 |
| C46 | 0.89175 | 0.28596 | 0.44633 | C320 | 0.89061 | 0.78987 | 0.44584 |
| N47 | 0.13595 | 0.33847 | 0.37812 | N321 | 0.13531 | 0.83493 | 0.38024 |
| C48 | 0.16005 | 0.24262 | 0.37296 | C322 | 0.15923 | 0.73749 | 0.3751  |
| H49 | 0.02768 | 0.24024 | 0.42893 | H323 | 0.02653 | 0.73656 | 0.43003 |
| H50 | 0.94874 | 0.01066 | 0.4248  | H324 | 0.9474  | 0.50835 | 0.42571 |
| H51 | 0.91695 | 0.0134  | 0.38323 | H325 | 0.91646 | 0.51108 | 0.38353 |
| H52 | 0.85268 | 0.367   | 0.42291 | H326 | 0.85223 | 0.8727  | 0.42138 |
| H53 | 0.88448 | 0.36437 | 0.46409 | H327 | 0.88325 | 0.87022 | 0.46334 |
| C54 | 0.78855 | 0.27126 | 0.29682 | C328 | 0.79026 | 0.77244 | 0.29514 |
| C55 | 0.7709  | 0.28455 | 0.27068 | C329 | 0.26058 | 0.67685 | 0.78293 |
| C56 | 0.74051 | 0.25673 | 0.26971 | C330 | 0.22927 | 0.69151 | 0.78294 |
| C57 | 0.7761  | 0.33323 | 0.32181 | C331 | 0.77573 | 0.80367 | 0.32068 |
| C58 | 0.81795 | 0.19825 | 0.29728 | C332 | 0.82102 | 0.72521 | 0.2958  |
| C59 | 0.78301 | 0.32849 | 0.24489 | C333 | 0.27324 | 0.66254 | 0.75621 |
| H60 | 0.78253 | 0.37561 | 0.36537 | H334 | 0.7796  | 0.81917 | 0.3651  |
| H61 | 0.85712 | 0.12936 | 0.32188 | H335 | 0.86037 | 0.67273 | 0.32085 |
| H62 | 0.75333 | 0.39218 | 0.32201 | H336 | 0.75193 | 0.84319 | 0.32093 |
| H63 | 0.82791 | 0.14547 | 0.27823 | H337 | 0.83271 | 0.6977  | 0.27627 |
| F64 | 0.7265  | 0.2087  | 0.29324 | F338 | 0.21419 | 0.7094  | 0.80739 |
| F65 | 0.81209 | 0.3615  | 0.24329 | F339 | 0.30327 | 0.64225 | 0.7532  |
| H66 | 0.16849 | 0.13501 | 0.38678 | H340 | 0.16725 | 0.62837 | 0.38879 |
| C67 | 0.32742 | 0.42253 | 0.1729  | C341 | 0.32728 | 0.92035 | 0.17541 |
| C68 | 0.31734 | 0.35762 | 0.14784 | C342 | 0.31703 | 0.85809 | 0.15021 |
| C69 | 0.28889 | 0.29753 | 0.14747 | C343 | 0.28843 | 0.80001 | 0.14976 |
| C70 | 0.43578 | 0.33081 | 0.02553 | C344 | 0.43517 | 0.83274 | 0.02748 |
| C71 | 0.46728 | 0.33988 | 0.02463 | C345 | 0.46671 | 0.84037 | 0.02643 |
| C72 | 0.48359 | 0.39067 | 0.04802 | C346 | 0.48328 | 0.88898 | 0.0498  |
| C73 | 0.41772 | 0.34738 | 0.05103 | C347 | 0.41724 | 0.84876 | 0.05308 |
| C74 | 0.57369 | 0.27349 | 0.07358 | C348 | 0.57448 | 0.77584 | 0.07468 |
| C75 | 0.59199 | 0.28005 | 0.09662 | C349 | 0.59361 | 0.78181 | 0.09716 |
| C76 | 0.61879 | 0.359   | 0.0951  | C350 | 0.6206  | 0.85795 | 0.09464 |
| C77 | 0.62621 | 0.43543 | 0.06978 | C351 | 0.62725 | 0.93345 | 0.06904 |
| C78 | 0.60762 | 0.42974 | 0.04686 | C352 | 0.60779 | 0.92883 | 0.04675 |
| N79 | 0.36346 | 0.378   | 0.12318 | N353 | 0.36317 | 0.87686 | 0.12543 |
| C80 | 0.33578 | 0.34611 | 0.12279 | C354 | 0.33538 | 0.8473  | 0.12508 |
| H81 | 0.47152 | 0.43313 | 0.06674 | H355 | 0.4714  | 0.92996 | 0.0687  |
| H82 | 0.55379 | 0.20324 | 0.07506 | H356 | 0.5545  | 0.70691 | 0.07689 |
| H83 | 0.58612 | 0.21801 | 0.11608 | H357 | 0.58825 | 0.72082 | 0.11685 |
| H84 | 0.64653 | 0.50203 | 0.0682  | H358 | 0.64771 | 0.99797 | 0.06676 |
| H85 | 0.61367 | 0.49263 | 0.02748 | H359 | 0.61332 | 0.99108 | 0.02716 |
| C86 | 0.71761 | 0.33247 | 0.19346 | C360 | 0.7233  | 0.82131 | 0.18998 |
| C87 | 0.73567 | 0.31767 | 0.21937 | C361 | 0.22487 | 0.67858 | 0.73185 |
| C88 | 0.7661  | 0.34533 | 0.22041 | C362 | 0.25617 | 0.66306 | 0.73188 |
| C89 | 0.73025 | 0.29014 | 0.16756 | C363 | 0.73258 | 0.73504 | 0.16574 |
| C90 | 0.68721 | 0.38634 | 0.19409 | C364 | 0.69487 | 0.90789 | 0.18989 |

|      |         |          |          |      |         |         |          |
|------|---------|----------|----------|------|---------|---------|----------|
| C91  | 0.72372 | 0.2713   | 0.24516  | C365 | 0.21221 | 0.69291 | 0.75855  |
| H92  | 0.72345 | 0.25978  | 0.12369  | H366 | 0.72193 | 0.6664  | 0.12397  |
| H93  | 0.64635 | 0.4341   | 0.17105  | H367 | 0.65454 | 0.97797 | 0.16703  |
| H94  | 0.7539  | 0.24854  | 0.16617  | H368 | 0.75407 | 0.66021 | 0.16547  |
| H95  | 0.67674 | 0.42498  | 0.21375  | H369 | 0.68711 | 0.97702 | 0.20845  |
| F96  | 0.78025 | 0.39467  | 0.19689  | F370 | 0.27129 | 0.65259 | 0.70726  |
| F97  | 0.69481 | 0.23372  | 0.24676  | F371 | 0.1821  | 0.7043  | 0.76147  |
| H98  | 0.32504 | 0.29914  | 0.1037   | H372 | 0.32441 | 0.80291 | 0.10593  |
| C99  | 0.8321  | 0.09194  | 0.67251  | C373 | 0.82974 | 0.60142 | 0.67259  |
| C100 | 0.82313 | 0.20201  | 0.65036  | C374 | 0.82024 | 0.70898 | 0.65029  |
| C101 | 0.79616 | 0.30226  | 0.65313  | C375 | 0.79309 | 0.80743 | 0.65315  |
| C102 | 0.93769 | 0.18096  | 0.52431  | C376 | 0.93532 | 0.68346 | 0.52452  |
| C103 | 0.96918 | 0.19209  | 0.52472  | C377 | 0.96685 | 0.6922  | 0.52522  |
| C104 | 0.98552 | 0.19564  | 0.55     | C378 | 0.98294 | 0.69272 | 0.55067  |
| C105 | 0.91917 | 0.16458  | 0.54934  | C379 | 0.9166  | 0.6686  | 0.54945  |
| C106 | 0.07443 | 0.32232  | 0.5747   | C380 | 0.06945 | 0.83402 | 0.57626  |
| C107 | 0.09322 | 0.32275  | 0.59754  | C381 | 0.08719 | 0.83856 | 0.59974  |
| C108 | 0.12081 | 0.22603  | 0.59814  | C382 | 0.11533 | 0.74819 | 0.60123  |
| C109 | 0.12925 | 0.13277  | 0.57427  | C383 | 0.12545 | 0.65766 | 0.57758  |
| C110 | 0.11045 | 0.1338   | 0.55142  | C384 | 0.10764 | 0.65397 | 0.55412  |
| N111 | 0.86431 | 0.11459  | 0.62053  | N385 | 0.86158 | 0.62349 | 0.62064  |
| C112 | 0.84125 | 0.2193   | 0.62547  | C386 | 0.83793 | 0.72466 | 0.62511  |
| H113 | 0.97374 | 0.19182  | 0.56982  | H387 | 0.971   | 0.68528 | 0.57036  |
| H114 | 0.05375 | 0.40494  | 0.57471  | H388 | 0.04825 | 0.91185 | 0.57554  |
| H115 | 0.08659 | 0.40058  | 0.61551  | H389 | 0.07929 | 0.91458 | 0.61753  |
| H116 | 0.1508  | 0.05888  | 0.57339  | H390 | 0.1475  | 0.58903 | 0.57736  |
| H117 | 0.11745 | 0.05768  | 0.53332  | H391 | 0.1159  | 0.57954 | 0.53625  |
| C118 | 0.21585 | 0.14748  | 0.70021  | C392 | 0.2069  | 0.67769 | 0.70598  |
| C119 | 0.23426 | 0.15313  | 0.7256   | C393 | 0.7416  | 0.81441 | 0.21562  |
| C120 | 0.26566 | 0.13863  | 0.7268   | C394 | 0.77222 | 0.8382  | 0.21755  |
| C121 | 0.22319 | 0.0331   | 0.67863  | C395 | 0.217   | 0.57984 | 0.68318  |
| C122 | 0.1893  | 0.25229  | 0.69746  | C396 | 0.17933 | 0.77275 | 0.70348  |
| C123 | 0.22037 | 0.17103  | 0.75174  | C397 | 0.7281  | 0.78058 | 0.24126  |
| H124 | 0.21153 | -0.0641  | 0.63889  | H398 | 0.20825 | 0.49566 | 0.64186  |
| H125 | 0.15116 | 0.33226  | 0.67216  | H399 | 0.14129 | 0.84602 | 0.67766  |
| H126 | 0.24274 | -0.05759 | 0.68074  | H400 | 0.23814 | 0.50206 | 0.6845   |
| H127 | 0.1827  | 0.34149  | 0.71402  | H401 | 0.17098 | 0.85207 | 0.72069  |
| N128 | 0.2876  | 0.12837  | 0.70656  | N402 | 0.79288 | 0.88796 | 0.19839  |
| H129 | 0.19605 | 0.18305  | 0.75242  | H403 | 0.7044  | 0.75994 | 0.24126  |
| H130 | 0.83435 | 0.32941  | 0.61156  | H404 | 0.83004 | 0.82945 | 0.61051  |
| C131 | 0.51463 | 0.31374  | 0.99844  | C405 | 0.51388 | 0.8143  | 0.99992  |
| C132 | 0.57687 | 0.31061  | -0.00244 | C406 | 0.57608 | 0.81067 | -0.00147 |
| H133 | 0.60118 | 0.30727  | -0.00273 | H407 | 0.60039 | 0.80714 | -0.002   |
| C134 | 0.01653 | 0.22024  | 0.49955  | C408 | 0.01444 | 0.72105 | 0.50053  |
| C135 | 0.07841 | 0.23772  | 0.50012  | C409 | 0.0762  | 0.74369 | 0.50181  |
| H136 | 0.10245 | 0.24901  | 0.50039  | H410 | 0.10014 | 0.75779 | 0.50236  |
| C137 | 0.31452 | 0.11555  | 0.71997  | C411 | 0.81891 | 0.90424 | 0.21247  |
| H138 | 0.33558 | 0.10497  | 0.70867  | H412 | 0.83877 | 0.94418 | 0.20208  |

|      |         |         |         |      |         |         |         |
|------|---------|---------|---------|------|---------|---------|---------|
| C139 | 0.28468 | 0.14998 | 0.67683 | C413 | 0.78916 | 0.92102 | 0.16901 |
| C140 | 0.32909 | 0.20927 | 0.82948 | C414 | 0.32387 | 0.74336 | 0.83246 |
| C141 | 0.31632 | 0.16861 | 0.85504 | C415 | 0.31428 | 0.67164 | 0.85736 |
| C142 | 0.28655 | 0.12797 | 0.85503 | C416 | 0.28642 | 0.60441 | 0.85747 |
| C143 | 0.43527 | 0.27189 | 0.97525 | C417 | 0.43423 | 0.7764  | 0.97711 |
| C144 | 0.46689 | 0.27758 | 0.97433 | C418 | 0.46588 | 0.78055 | 0.97606 |
| C145 | 0.48326 | 0.25782 | 0.94919 | C419 | 0.48205 | 0.76074 | 0.95081 |
| C146 | 0.41633 | 0.24013 | 0.9511  | C420 | 0.41509 | 0.74703 | 0.95297 |
| C147 | 0.57269 | 0.35499 | 0.92187 | C421 | 0.57139 | 0.85482 | 0.92285 |
| C148 | 0.59095 | 0.33796 | 0.8988  | C422 | 0.58956 | 0.83769 | 0.89972 |
| C149 | 0.61825 | 0.23882 | 0.90007 | C423 | 0.61693 | 0.73917 | 0.90094 |
| C150 | 0.6263  | 0.15441 | 0.9252  | C424 | 0.62513 | 0.65536 | 0.92608 |
| C151 | 0.60778 | 0.1711  | 0.94813 | C425 | 0.60667 | 0.67193 | 0.94906 |
| N152 | 0.36191 | 0.18316 | 0.8803  | N426 | 0.36    | 0.69967 | 0.88226 |
| C153 | 0.33357 | 0.16807 | 0.88085 | C427 | 0.33243 | 0.66699 | 0.88274 |
| H154 | 0.47141 | 0.24206 | 0.92964 | H428 | 0.47005 | 0.74573 | 0.93131 |
| H155 | 0.55238 | 0.44044 | 0.92059 | H429 | 0.55104 | 0.93995 | 0.92159 |
| H156 | 0.5846  | 0.40703 | 0.87957 | H430 | 0.58309 | 0.90624 | 0.88047 |
| H157 | 0.64712 | 0.07338 | 0.92657 | H431 | 0.646   | 0.57483 | 0.92743 |
| H158 | 0.61436 | 0.10205 | 0.96735 | H432 | 0.61336 | 0.60319 | 0.96829 |
| C159 | 0.71691 | 0.18623 | 0.80194 | C433 | 0.71527 | 0.68788 | 0.80261 |
| C160 | 0.73494 | 0.18076 | 0.77613 | C434 | 0.73314 | 0.68355 | 0.7767  |
| C161 | 0.76617 | 0.15289 | 0.77604 | C435 | 0.76438 | 0.65753 | 0.77645 |
| C162 | 0.72805 | 0.25032 | 0.82666 | C436 | 0.72644 | 0.75254 | 0.82726 |
| C163 | 0.68806 | 0.13129 | 0.80224 | C437 | 0.68652 | 0.63154 | 0.80304 |
| C164 | 0.7222  | 0.20808 | 0.74951 | C438 | 0.72019 | 0.71059 | 0.75016 |
| H165 | 0.71979 | 0.3149  | 0.86936 | H439 | 0.71828 | 0.81722 | 0.86998 |
| H166 | 0.64831 | 0.1013  | 0.82583 | H440 | 0.64693 | 0.60014 | 0.82679 |
| H167 | 0.75034 | 0.29504 | 0.82715 | H441 | 0.74867 | 0.79818 | 0.82765 |
| H168 | 0.67902 | 0.07643 | 0.78358 | H442 | 0.6775  | 0.576   | 0.78443 |
| F169 | 0.78117 | 0.12005 | 0.80035 | F443 | 0.77958 | 0.62499 | 0.80066 |
| F170 | 0.69228 | 0.24133 | 0.74676 | F444 | 0.69024 | 0.74212 | 0.74758 |
| H171 | 0.32142 | 0.14771 | 0.90067 | H445 | 0.3214  | 0.62785 | 0.90232 |
| C172 | 0.83433 | 0.18832 | 0.32178 | C446 | 0.83652 | 0.71111 | 0.32069 |
| C173 | 0.82212 | 0.25208 | 0.34678 | C447 | 0.8221  | 0.74458 | 0.34616 |
| C174 | 0.79261 | 0.32421 | 0.3462  | C448 | 0.79134 | 0.79102 | 0.34554 |
| C175 | 0.93789 | 0.19473 | 0.47306 | C449 | 0.93611 | 0.69654 | 0.47332 |
| C176 | 0.96937 | 0.20573 | 0.47335 | C450 | 0.96761 | 0.70582 | 0.47388 |
| C177 | 0.9858  | 0.22238 | 0.44843 | C451 | 0.98438 | 0.72072 | 0.44911 |
| C178 | 0.91963 | 0.19204 | 0.4475  | C452 | 0.91823 | 0.69361 | 0.44756 |
| C179 | 0.073   | 0.37876 | 0.42715 | C453 | 0.07168 | 0.87632 | 0.42846 |
| C180 | 0.09123 | 0.39775 | 0.40438 | C454 | 0.09017 | 0.89409 | 0.40585 |
| C181 | 0.11938 | 0.30787 | 0.40245 | C455 | 0.11844 | 0.80516 | 0.4044  |
| C182 | 0.1289  | 0.20161 | 0.425   | C456 | 0.12783 | 0.70149 | 0.4273  |
| C183 | 0.11062 | 0.18357 | 0.44775 | C457 | 0.10928 | 0.68474 | 0.44991 |
| N184 | 0.86666 | 0.18788 | 0.37406 | N458 | 0.86671 | 0.69319 | 0.37346 |
| C185 | 0.83888 | 0.24552 | 0.37289 | C459 | 0.83821 | 0.7323  | 0.37258 |
| H186 | 0.9741  | 0.22685 | 0.42844 | H460 | 0.97294 | 0.72409 | 0.42899 |

|      |         |         |         |      |         |         |         |
|------|---------|---------|---------|------|---------|---------|---------|
| H187 | 0.05185 | 0.45666 | 0.42828 | H461 | 0.05044 | 0.95357 | 0.42922 |
| H188 | 0.08377 | 0.48545 | 0.38753 | H462 | 0.08282 | 0.98006 | 0.38873 |
| H189 | 0.15081 | 0.13139 | 0.4249  | H463 | 0.14986 | 0.63254 | 0.42758 |
| H190 | 0.11842 | 0.09702 | 0.46468 | H464 | 0.11699 | 0.60028 | 0.46714 |
| C191 | 0.21158 | 0.30442 | 0.29898 | C465 | 0.21115 | 0.80042 | 0.30135 |
| C192 | 0.22925 | 0.32101 | 0.27323 | C466 | 0.22888 | 0.81769 | 0.27565 |
| C193 | 0.26058 | 0.30292 | 0.27295 | C467 | 0.2602  | 0.79884 | 0.27535 |
| C194 | 0.22193 | 0.19383 | 0.32062 | C468 | 0.22134 | 0.68872 | 0.32291 |
| C195 | 0.18399 | 0.39748 | 0.30244 | C469 | 0.18364 | 0.89423 | 0.30486 |
| C196 | 0.2162  | 0.35475 | 0.24686 | C470 | 0.21587 | 0.85321 | 0.24931 |
| H197 | 0.21359 | 0.08657 | 0.36079 | H471 | 0.21279 | 0.58009 | 0.36294 |
| H198 | 0.14608 | 0.45472 | 0.32876 | H472 | 0.14571 | 0.95156 | 0.33115 |
| H199 | 0.24309 | 0.11759 | 0.31859 | H473 | 0.24244 | 0.61196 | 0.32085 |
| H200 | 0.17547 | 0.48676 | 0.28624 | H474 | 0.17523 | 0.98446 | 0.28875 |
| F201 | 0.27604 | 0.27386 | 0.29711 | F475 | 0.27564 | 0.76812 | 0.29948 |
| F202 | 0.18611 | 0.37121 | 0.24412 | F476 | 0.18579 | 0.87048 | 0.24653 |
| H203 | 0.82615 | 0.29175 | 0.39159 | H477 | 0.82458 | 0.75513 | 0.39174 |
| C204 | 0.67006 | 0.39273 | 0.17007 | C478 | 0.67659 | 0.9094  | 0.16664 |
| C205 | 0.68247 | 0.34538 | 0.14432 | C479 | 0.68584 | 0.82309 | 0.14244 |
| C206 | 0.71303 | 0.29628 | 0.14356 | C480 | 0.71427 | 0.73685 | 0.14248 |
| C207 | 0.56233 | 0.33682 | 0.023   | C481 | 0.56184 | 0.83687 | 0.02412 |
| C208 | 0.53058 | 0.34459 | 0.02331 | C482 | 0.53008 | 0.84427 | 0.02471 |
| C209 | 0.51391 | 0.39241 | 0.04743 | C483 | 0.5136  | 0.89058 | 0.049   |
| C210 | 0.58085 | 0.34956 | 0.0481  | C484 | 0.5809  | 0.85025 | 0.04886 |
| C211 | 0.4275  | 0.27582 | 0.07673 | C485 | 0.42694 | 0.77452 | 0.07862 |
| C212 | 0.40945 | 0.28625 | 0.09999 | C486 | 0.40894 | 0.78403 | 0.10192 |
| C213 | 0.38038 | 0.36481 | 0.09849 | C487 | 0.38    | 0.8646  | 0.10066 |
| C214 | 0.3704  | 0.43741 | 0.07296 | C488 | 0.37012 | 0.94014 | 0.07532 |
| C215 | 0.38869 | 0.42765 | 0.04985 | C489 | 0.38835 | 0.93103 | 0.05214 |
| N216 | 0.6357  | 0.36717 | 0.11984 | N490 | 0.63865 | 0.86813 | 0.11862 |
| C217 | 0.66433 | 0.34074 | 0.11901 | C491 | 0.66648 | 0.81574 | 0.11795 |
| H218 | 0.52553 | 0.43589 | 0.06571 | H492 | 0.52538 | 0.93267 | 0.06729 |
| H219 | 0.44942 | 0.20597 | 0.07826 | H493 | 0.44874 | 0.70326 | 0.07996 |
| H220 | 0.41744 | 0.22747 | 0.11961 | H494 | 0.41688 | 0.72302 | 0.12139 |
| H221 | 0.34815 | 0.50327 | 0.07139 | H495 | 0.348   | 1.00773 | 0.07392 |
| H222 | 0.38049 | 0.48695 | 0.03029 | H496 | 0.38019 | 0.99253 | 0.03273 |
| C223 | 0.28207 | 0.35792 | 0.19648 | C497 | 0.28178 | 0.85707 | 0.19892 |
| C224 | 0.2642  | 0.34941 | 0.22226 | C498 | 0.2639  | 0.84785 | 0.22471 |
| C225 | 0.2329  | 0.36778 | 0.22251 | C499 | 0.2326  | 0.86715 | 0.22499 |
| C226 | 0.27157 | 0.29735 | 0.17124 | C500 | 0.27113 | 0.79912 | 0.17355 |
| C227 | 0.31017 | 0.42272 | 0.19672 | C501 | 0.31004 | 0.91995 | 0.19925 |
| C228 | 0.27726 | 0.31669 | 0.24863 | C502 | 0.27691 | 0.81341 | 0.25105 |
| H229 | 0.2804  | 0.24705 | 0.12812 | H503 | 0.27978 | 0.75162 | 0.13032 |
| H230 | 0.34933 | 0.47262 | 0.17352 | H504 | 0.3493  | 0.96897 | 0.17614 |
| H231 | 0.24985 | 0.2455  | 0.17034 | H505 | 0.24929 | 0.74883 | 0.17253 |
| H232 | 0.31862 | 0.4752  | 0.21584 | H506 | 0.31865 | 0.97048 | 0.21846 |
| F233 | 0.21759 | 0.40436 | 0.19848 | F507 | 0.21729 | 0.90549 | 0.201   |
| F234 | 0.30732 | 0.2914  | 0.25111 | F508 | 0.30696 | 0.78717 | 0.25355 |

|      |         |          |         |      |         |         |         |
|------|---------|----------|---------|------|---------|---------|---------|
| H235 | 0.67647 | 0.30441  | 0.09946 | H509 | 0.67689 | 0.75426 | 0.09914 |
| C236 | 0.17159 | 0.24819  | 0.6739  | C510 | 0.16265 | 0.77046 | 0.67923 |
| C237 | 0.17924 | 0.13642  | 0.6521  | C511 | 0.17249 | 0.67061 | 0.65657 |
| C238 | 0.20528 | 0.02766  | 0.65522 | C512 | 0.20002 | 0.57533 | 0.65916 |
| C239 | 0.06381 | 0.22701  | 0.52563 | C513 | 0.06121 | 0.73378 | 0.52713 |
| C240 | 0.03227 | 0.21841  | 0.5254  | C514 | 0.0298  | 0.7206  | 0.52654 |
| C241 | 0.01573 | 0.20374  | 0.5503  | C515 | 0.01311 | 0.70195 | 0.55126 |
| C242 | 0.08239 | 0.22603  | 0.55104 | C516 | 0.079   | 0.73905 | 0.55289 |
| C243 | 0.92788 | 0.05995  | 0.57231 | C517 | 0.92461 | 0.55966 | 0.57207 |
| C244 | 0.90942 | 0.04712  | 0.59508 | C518 | 0.906   | 0.54843 | 0.59477 |
| C245 | 0.88131 | 0.13806  | 0.59623 | C519 | 0.87848 | 0.64541 | 0.5962  |
| C246 | 0.87197 | 0.23759  | 0.57285 | C520 | 0.8699  | 0.7503  | 0.57321 |
| C247 | 0.89052 | 0.24981  | 0.55015 | C521 | 0.88853 | 0.76038 | 0.55052 |
| N248 | 0.1385  | 0.23955  | 0.62238 | N522 | 0.13168 | 0.76388 | 0.62622 |
| C249 | 0.16048 | 0.1267   | 0.62733 | C523 | 0.15484 | 0.65934 | 0.63117 |
| H250 | 0.02736 | 0.1994   | 0.57032 | H524 | 0.02461 | 0.69465 | 0.57135 |
| H251 | 0.94897 | -0.01883 | 0.57182 | H525 | 0.94522 | 0.47666 | 0.57135 |
| H252 | 0.91669 | -0.03612 | 0.61256 | H526 | 0.91268 | 0.46162 | 0.61197 |
| H253 | 0.84998 | 0.30695  | 0.57229 | H527 | 0.84847 | 0.82572 | 0.5729  |
| H254 | 0.88288 | 0.33173  | 0.53259 | H528 | 0.88151 | 0.84588 | 0.5332  |
| C255 | 0.7883  | 0.186    | 0.69923 | C529 | 0.78597 | 0.69312 | 0.69952 |
| C256 | 0.77034 | 0.18156  | 0.72502 | C530 | 0.7682  | 0.68759 | 0.72541 |
| C257 | 0.73909 | 0.2087   | 0.72512 | C531 | 0.73692 | 0.7127  | 0.72568 |
| C258 | 0.77896 | 0.29358  | 0.67687 | C532 | 0.7762  | 0.79909 | 0.67709 |
| C259 | 0.8152  | 0.08506  | 0.69649 | C533 | 0.8131  | 0.59439 | 0.69672 |
| C260 | 0.78308 | 0.15286  | 0.75164 | C534 | 0.78113 | 0.65909 | 0.75196 |
| H261 | 0.78858 | 0.38939  | 0.6362  | H535 | 0.78514 | 0.89303 | 0.63615 |
| H262 | 0.85297 | 0.01213  | 0.67079 | H536 | 0.85078 | 0.52323 | 0.67082 |
| H263 | 0.75832 | 0.37533  | 0.6783  | H537 | 0.75542 | 0.87952 | 0.67859 |
| H264 | 0.82289 | -0.0023  | 0.71326 | H538 | 0.82116 | 0.5085  | 0.71356 |
| F265 | 0.72389 | 0.23386  | 0.70074 | F539 | 0.72153 | 0.73739 | 0.7014  |
| F266 | 0.8131  | 0.12846  | 0.75454 | F540 | 0.81119 | 0.63651 | 0.75472 |
| H267 | 0.16601 | 0.01598  | 0.61357 | H541 | 0.16241 | 0.5547  | 0.61683 |
| C268 | 0.48291 | 0.30948  | 0.99915 | C542 | 0.48213 | 0.81079 | 1.00082 |
| C269 | 0.42097 | 0.29937  | 0.00077 | C543 | 0.42014 | 0.80346 | 0.00275 |
| H270 | 0.39696 | 0.28803  | 0.00171 | H544 | 0.3961  | 0.79349 | 0.00379 |
| C271 | 0.98502 | 0.20544  | 0.49921 | C545 | 0.98297 | 0.70575 | 0.49988 |
| C272 | 0.92317 | 0.18641  | 0.49852 | C546 | 0.92103 | 0.69042 | 0.49863 |
| H273 | 0.89916 | 0.17427  | 0.49817 | H547 | 0.89695 | 0.68074 | 0.49808 |
| H274 | 0.30347 | 0.21635  | 0.66899 | H548 | 0.80459 | 1.01391 | 0.16238 |

**Supplementary Table 3.** Pore parameters of Py-TFImI-25 COF and Py-TFIm-25 COF.

| Adsorbents  | Surface area (m <sup>2</sup> /g) | Microporous pore volume (cm <sup>3</sup> /g) | Pore volume (cm <sup>3</sup> /g) |
|-------------|----------------------------------|----------------------------------------------|----------------------------------|
| Py-TFImI-25 | 1324.05                          | 0.013                                        | 0.68                             |
| Py-TFIm-25  | 1430.15                          | 0.028                                        | 0.74                             |

**Supplementary Table 4.** Th(IV) adsorption performances over various materials.

| Category      | Sorbents                                                                                         | Equilibrium time | c <sub>0</sub><br>(mg L <sup>-1</sup> ) | q <sub>e</sub><br>(mg g <sup>-1</sup> ) | Ref.        |
|---------------|--------------------------------------------------------------------------------------------------|------------------|-----------------------------------------|-----------------------------------------|-------------|
| Zeolite       | NKF-6                                                                                            | 3 h              | 40                                      | 51                                      | 17          |
| Polymer       | P (DVB-VPA)                                                                                      | 1 h              | 26                                      | 54                                      | 18          |
| Porous silica | SBA/SA                                                                                           | 5 h              | 50                                      | 12                                      | 19          |
|               | SBA/EnSA                                                                                         | 2 h              | 50                                      | 49                                      |             |
| GO            | Reduced graphene oxide                                                                           | 50 min           | 35                                      | 12                                      | 20          |
| MOF           | MOFs with O <sup>-</sup> groups                                                                  | 10 min           | 41                                      | 67                                      | 21          |
| Composites    | Tannin-modified poly(glycidylmethacrylate)-grafted zirconium oxide-densified cellulose (TMPGZDC) | 2 h              | 50                                      | 23                                      | 22          |
|               | Fe <sub>3</sub> O <sub>4</sub> @AMCA-MIL53(Al)                                                   | 90 min           | 20                                      | 22                                      | 23          |
| COF           | [NH <sub>4</sub> ] <sup>+</sup> [COF-SO <sub>3</sub> <sup>-</sup> ]                              | 1 h              | 50                                      | 100                                     | 24          |
|               | COF-DL229                                                                                        | 1 min            | 50                                      | 133                                     | 25          |
|               | <b>Py-TFImI-25 COF</b>                                                                           | <b>4 min</b>     | <b>50</b>                               | <b>136</b>                              | <b>This</b> |
|               | <b>Py-TFIm-25 COF</b>                                                                            | <b>1 min</b>     | <b>47</b>                               | <b>141</b>                              | <b>work</b> |

**Supplementary Table 5.** Analysis of nitrogen sites (Solid-liquid ratio = 1:3000 g mL<sup>-1</sup>, Initial concentration of Th(IV) ≈ 22 mg L<sup>-1</sup>, pH = 4, T = 25°C).

| COFs        | N <sub>C=N</sub> content<br>(mmol L <sup>-1</sup> ) | N <sub>Im</sub> content<br>(mmol L <sup>-1</sup> ) | Th(IV)<br>concentration<br>(mmol L <sup>-1</sup> ) | H <sup>+</sup><br>concentration<br>(mmol L <sup>-1</sup> ) | Th(IV)<br>uptake<br>capacity<br>(mg g <sup>-1</sup> ) |
|-------------|-----------------------------------------------------|----------------------------------------------------|----------------------------------------------------|------------------------------------------------------------|-------------------------------------------------------|
| Py-TFImI-25 | 1.28                                                | 0                                                  | 0.1                                                | 0.1                                                        | 43.1                                                  |
| Py-TFIm-25  | 1.39                                                | 0.35                                               | 0.1                                                | 0.1                                                        | 61.2                                                  |

**Supplementary Table 6.** Analysis of nitrogen sites (Solid-liquid ratio = 1:3000 g mL<sup>-1</sup>, Initial concentration of Th(IV) ≈ 273 mg L<sup>-1</sup>, pH = 4, T = 25°C).

| COFs        | N <sub>C=N</sub> content<br>(mmol L <sup>-1</sup> ) | N <sub>Im</sub> content<br>(mmol L <sup>-1</sup> ) | Th(IV)<br>concentration<br>(mmol L <sup>-1</sup> ) | H <sup>+</sup><br>concentration<br>(mmol L <sup>-1</sup> ) | Th(IV)<br>uptake<br>capacity<br>(mg g <sup>-1</sup> ) |
|-------------|-----------------------------------------------------|----------------------------------------------------|----------------------------------------------------|------------------------------------------------------------|-------------------------------------------------------|
| Py-TFImI-25 | 1.28                                                | 0                                                  | 1.2                                                | 0.1                                                        | 468.3                                                 |
| Py-TFIm-25  | 1.39                                                | 0.35                                               | 1.2                                                | 0.1                                                        | 822.6                                                 |

#### 4. Supplementary Note 1

Since the imidazole nitrogen site would be protonated before the imine nitrogen, we analyzed the occupation of the nitrogen sites at different thorium concentrations. When the initial concentration of Th(IV) is about 22 mg L<sup>-1</sup> and the pH is 4, the initial concentration is 0.1 mmol L<sup>-1</sup> for Th(IV) and 0.1 mmol L<sup>-1</sup> for H<sup>+</sup>, while the content of imidazole nitrogen (N<sub>Im</sub>) is 0.35 mmol L<sup>-1</sup> (Supplementary Table 5). Moreover, the hydrogen ions will not be completely bound to imidazole nitrogen sites due to the presence of protonation constant. Therefore, at this concentration, the main adsorption site of Py-TFIm-25 COF is the imidazole nitrogen, while the adsorption site of Py-TFImI-25 COF is the imine nitrogen.

When the initial Th(IV) concentration increased 12 times, the uptake capacity of Py-TFImI-25 COF for Th(IV) increased by 10.9 times and the uptake capacity of Py-TFIm-25 COF for Th(IV) increased by 13.4 times (Supplementary Table 5 and 6). In this adsorption system, there are hydrolysis equilibriums of thorium ions, proton equilibriums, and reaction equilibriums. When the pH is fixed, the difference between Py-TFImI-25 COF and Py-TFIm-25 COF adsorption systems should focus on the difference of the reaction equilibrium constants,  $k_1$  for the imine site and  $k_2$  for the imidazole site. The 10.9-fold increase in the uptake capacity of Py-TFImI-25 COF is due to the contribution of  $k_1$ , while the 13.4-fold increase in the uptake capacity of Py-TFIm-25 COF is due to the combined contribution of  $k_1$  and  $k_2$ . Therefore, at higher concentrations, the contribution of imidazole nitrogen cannot be neglected.

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