Supporting Information for

Green Mechanochemical Synthesis of Imine-Linked Covalent Organic Frameworks for High Iodine Capture

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Materials and Methods

Reagents

Reagents were obtained from different commercial sources and used as received. 2,5-Dimethoxylbenzene-1,4-dicarboxaldehyde (DMTP, 97%), tris(4-formylphenyl) amine (97%), glacial acetic acid, mesitylene, and 1,4-dioxane were purchased from Sigma Aldrich. Tetrahydrofuran (THF), acetone, and tris(4-aminophenyl) amine (97%) were purchased from Fisher Scientific. 1,3,5-triformylbenzene (TFB, 99.6%), 4,4′,4″-(1,3,5,-triazine-2,4,6-triyl) trianiline (TTA, 99.9%), 2,4,6-tris(4-formylphenyl)-1,3,5-triazine (TTB, >98%), and 1,3,5,-tris(4-aminophenyl)benzene (TPB, 99.6%) were purchased from ChemScene. 2,5-dimethyl-1,4-phenylenediamine (PD-CH_3, >98%) was purchased from TCI America. o-Tolidine (95%, pract) was purchased from Acros Organics. *n*-butanol was purchased from Alfa Aesar. 1,2-Dicholorbenzene (99%) and acetonitrile (ACS 99%) were purchased from Thermo Scientific.

Instrumentation

Infrared spectroscopy: Fourier transform infrared (FTIR) spectra were acquired on a Perkin Elmer Spectrum One FT-IR system.

Powder X-ray diffraction (PXRD): PXRD data were collected on a Rigaku MiniFlex 600 X-ray diffractometer with Cu Kα1 radiation (λ = 1.5406 Å) at 40 kV and 15 mA from 2θ = 2° to 30° with 0.02° increment.

Gas sorption measurements: Nitrogen sorption isotherms were measured on a Micromeritics 3Flex ASAP2020 gas sorption analyzer at 77 K. Before measurement, the COF samples were degassed under vacuum at 120 °C overnight. The specific surface areas were calculated using the Brunauer-Emmett-Teller (BET) method.

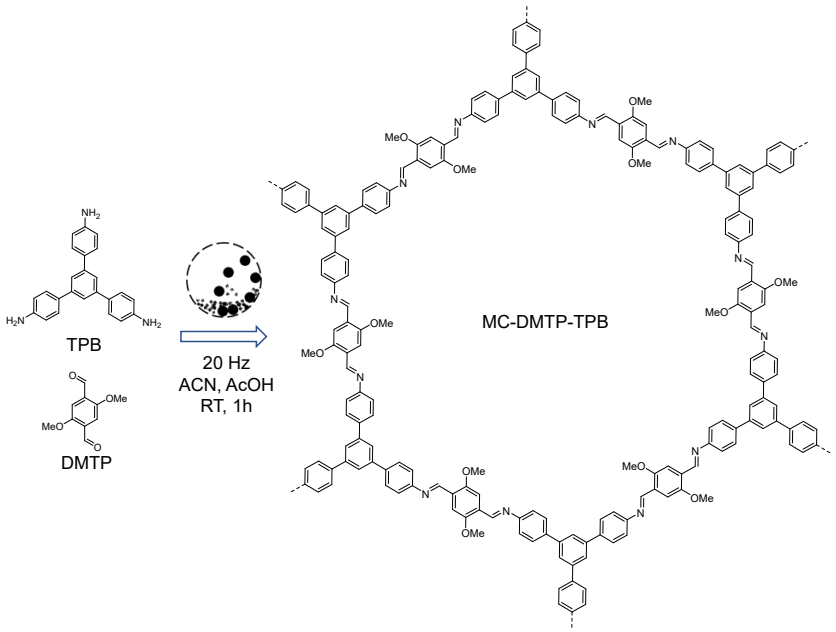
X-ray photoelectron spectroscopy (XPS): XPS measurements were conducted on a Thermo Scientific K-Alpha XPS apparatus equipped with a monochromatic Al K(alpha) source and flood gun for charge compensation.

Thermogravimetric analysis: Thermogravimetric analysis was performed on a TA Instruments Q5000 TGA under N_2 flow (25 mL/min) at a heating rate of 10 °C min^{-1} up to 900 °C.

Scanning electron microscopy (SEM): SEM images were obtained with a Zeiss Gemini Ultra-55 Analytical Field Emission Scanning Electron Microscope operated at 10 kV using an in-lens detector. Some SEM images (Figures S8-S11) were obtained on a TESCAN Vega 3 scanning electron microscope equipped with a Tungsten electron source and operated at 10 kV using a secondary electron detector. COF samples were sputtered with Au before imaging.

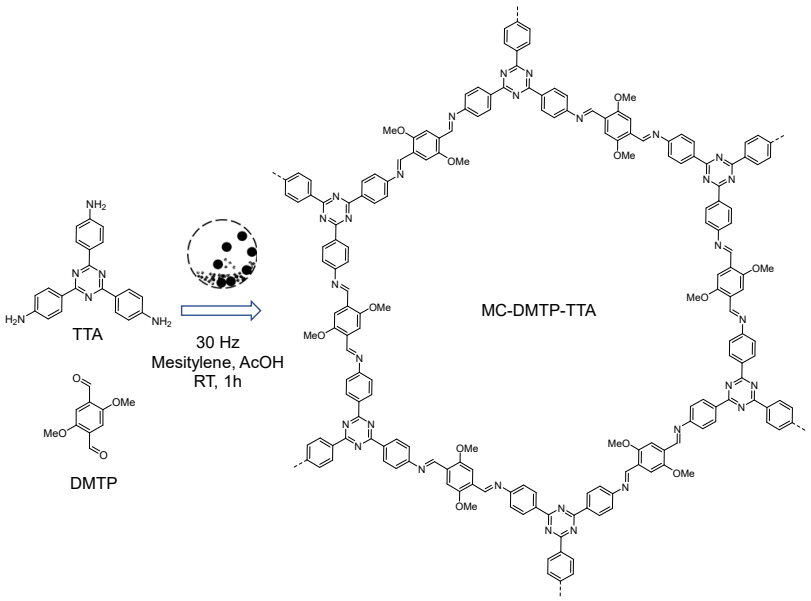
Synthetic Procedures

Mechanochemical synthesis of MC-DMTP-TPB COF



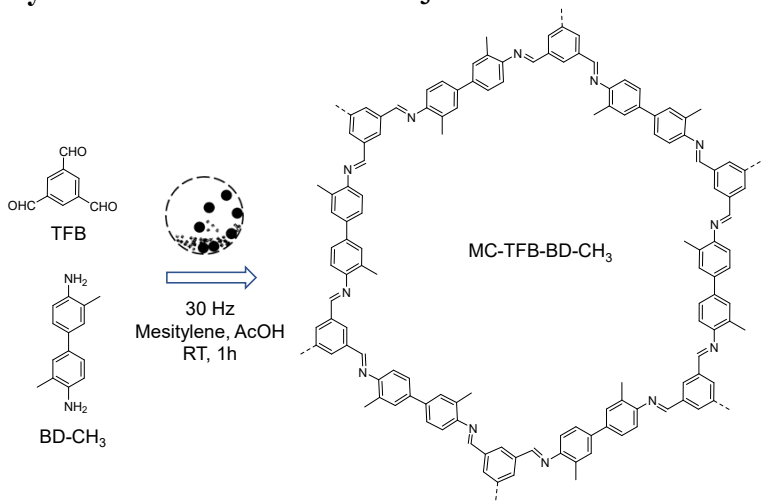
In a 5 mL stainless steel milling jar, 2,5-Dimethoxylbenzene-1,4-dicarboxaldehyde (DMTP) (20.4 mg, 0.105 mmol), 1,3,5,-tris(4-aminophenyl)benzene (TPB) (24.6 mg, 0.07 mmol), acetonitrile (30 µL), and glacial acetic acid (15 µL) were added along with a 5 mm steel ball. The reaction mixture was milled in a Retsch MM 400 Mixer Mill at 20 Hz for 1 hour. The resultant solid was collected via vacuum filtration, washed with THF and acetone, and subjected to Soxhlet extraction using THF as the solvent for 1 day. The collected powder was dried at 120 °C under vacuum overnight to give MC-DMTP-TPB in an isolated yield of 98%.

Mechanochemical synthesis of MC-DMTP-TTA COF



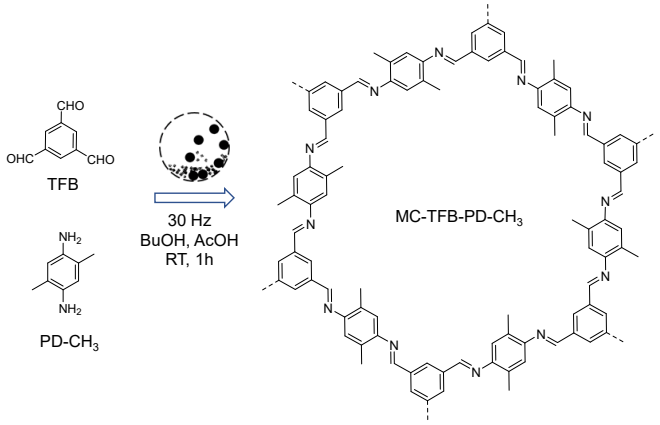
In a 5 mL stainless steel jar, DMTP (20.4 mg, 0.105 mmol), 4,4′,4″-(1,3,5,-triazine-2,4,6-triyl) trianiline (TTA) (24.8 mg, 0.07 mmol), mesitylene (20 µL), and glacial acetic acid (20 µL) were added along with a 5 mm steel ball. The reaction mixture was then milled using Retsch MM 400 Mixer Mill at 30 Hz for 1 hour. The solid was collected via vacuum filtration, washed with THF and acetone, and subjected to Soxhlet extraction using THF for 1 day. The collected powder was dried at 120 °C under vacuum overnight to give MC-DMTP-TTA in an isolated yield of 91%.

Mechanochemical synthesis of MC-TFB-BD-CH_3



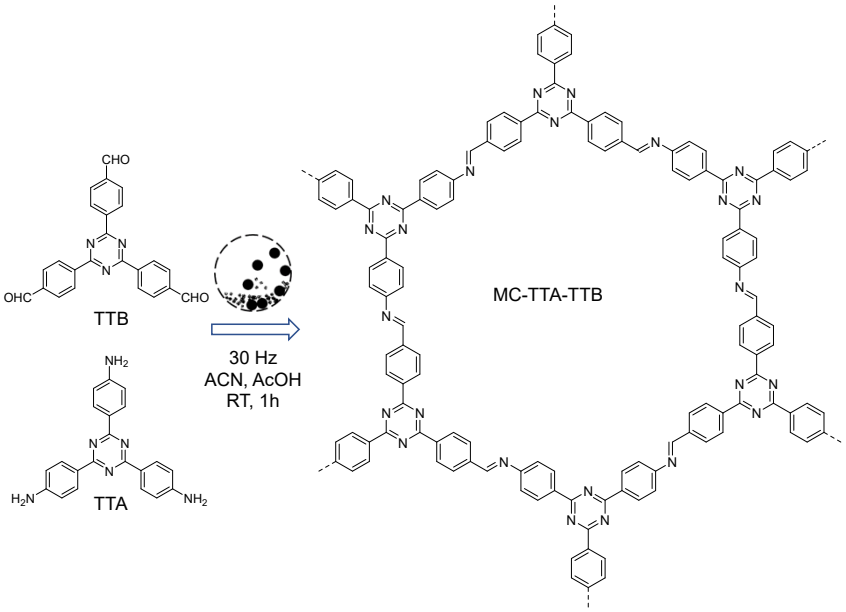
In a 5 mL stainless steel jar, 1,3,5-triformylbenzene (TFB) (11.3 mg, 0.07 mmol), o-tolidine (BD-CH_3, 22.3 mg, 0.105 mmol), mesitylene (15 µL), and glacial acetic acid (15 µL) were added along with a 5 mm steel ball. The reaction mixture was then milled using Retsch MM 400 Mixer Mill at 30 Hz for 1 hour. The solid was collected via vacuum filtration, washed with THF and acetone, and subjected to Soxhlet extraction using THF as the solvent for 1 day. The collected powder was dried at 120 °C under vacuum overnight, resulting in MC-TFB-BD-CH_3 in an isolated yield of ~87%.

Mechanochemical synthesis of MC-TFB-PD-CH_3



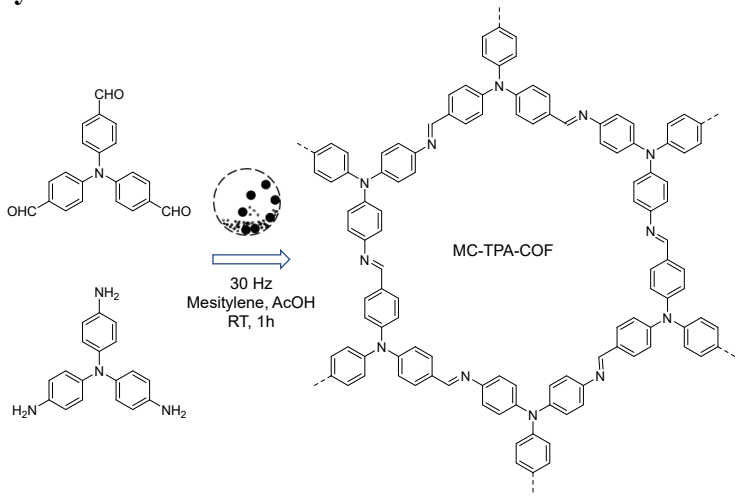
In a 5 mL stainless steel jar, TFB (22.6 mg, 0.14 mmol), 2,5-dimethyl-1,4-phenylenediamine (PD-CH_3) (28.6 mg, 0.21 mmol), *n*-butanol (15 µL), and glacial acetic acid (20 µL) were added along with a 5 mm steel ball. The reaction mixture was then milled in a Retsch MM 400 Mixer Mill at 30 Hz for 1 hour. The resulting solid was collected through vacuum filtration, washed with THF and acetone, and subjected to Soxhlet extraction using THF as the solvent for 1 day. The collected powder was dried at 120 °C under vacuum overnight to get MC-TFB-PD-CH_3 in an isolated yield of 85%.

Mechanochemical synthesis of MC-TTA-TTB COF



In a 5 mL stainless steel jar, TTA (24.9 mg, 0.07 mmol), 2,4,6-tris(4-formylphenyl)-1,3,5-triazine (TTB) (27.5 mg, 0.07 mmol), acetonitrile (15 µL), and glacial acetic acid (20 µL) were added with a 5 mm steel ball. The reaction mixture was milled in a Retsch MM 400 Mixer Mill at 30 Hz for 1 hour. The solid was collected via vacuum filtration, washed with THF and acetone followed by Soxhlet extraction using THF as the solvent for 1 day. The powder collected was dried at 120 °C under vacuum overnight to get MC-TTA-TTB COF in an isolated yield of 91%.

Mechanochemical synthesis of MC-TPA-COF



In a 5 mL stainless steel jar, tris(4-formylphenyl) amine (20.3 mg, 0.07 mmol), tris(4-aminophenyl) amine (23.1 mg, 0.07 mmol), mesitylene (20 µL), and glacial acetic acid (20 µL) were added along with a 5 mm steel ball. The reaction mixture was milled in a Retsch MM 400 Mixer Mill at 30 Hz for 1 hour. The solid was collected via vacuum filtration, washed with THF and acetone followed by Soxhlet extraction using THF as the solvent for 1 day. The powder collected was dried at 120 °C under vacuum overnight to get MC-TPA-COF in an isolated yield of 86%.

Conventional solvothermal synthesis of DMTP-TPB COF

This COF was synthesized based on a reported procedure.[1] In a 5 mL Biotage microwave vial, DMTP (46 mg, 0.24 mmol) and TPB (56 mg, 0.16 mmol) were dissolved by sonication in a mixture of 1,2-dichlorobenezene/*n*-butanol (1 mL/1 mL). Next, 6 M aqueous acetic acid (0.2 mL) was added to the mixture. The vial was sealed, subjected to three cycles of freeze-pump-thaw process, and then heated under vacuum in an oven at 120 ºC for 3 days. The resulting precipitates were collected by vacuum filtration, subjected to Soxhlet extraction using THF as the solvent for 1 day, and dried at 120 ºC under vacuum overnight to give a yellow powder in ~90 % yield.

Static iodine vapor adsorption

An open glass vial (1 mL) containing activated COFs (~25 mg) was placed inside a Scintillation vial (20 mL), which contains iodine solids (2 g). The Scintillation vial was sealed and kept at 75 ºC in an oven. After a certain period of exposure time, the Scintillation vial was removed from the oven and allowed to room temperature. The small vial containing COFs was weighted and placed back into the Scintillation vial. The Scintillation vial was resealed and returned to the oven to continue the iodine adsorption until the weight of the small vial containing COFs remained constant. The static iodine adsorption capacity of the COF was determined using the following equation:

$$Q = (W_f - W_i)/W_i$$

Where:

Q (g g^{-1}) refers to the adsorption capacity

W_f = final weight of COF after the adsorption of iodine

W_i = initial weight of COF before the adsorption of iodine

Recycling the COF adsorbent through thermal desorption

The iodine-captured MC-DMTP-TPB COF sample was placed in a vacuum oven at 135 °C overnight. The successful removal of adsorbed iodine from the COF was confirmed by the apparent changes in both weight and color of the sample. The regenerated COF was subjected to the next cycle.

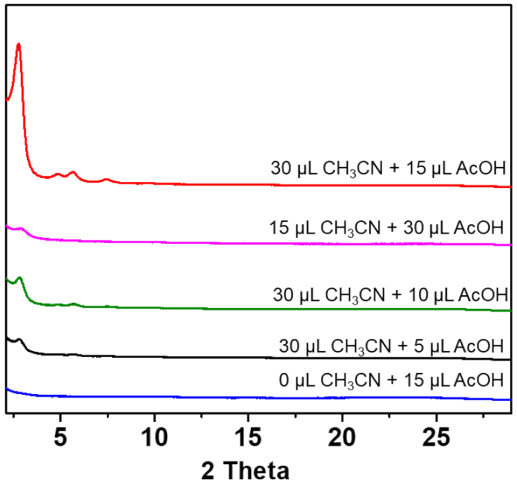


Figure S1. The effect of catalyst and LAG additive amount on the crystallinity of MC-DMTP-TPB COF, which was made by milling DMTP (20.4 mg, 0.105 mmol) with TPB (24.6 mg, 0.07 mmol) at 20 Hz for 1 hour using glacial acetic acid and acetonitrile as the LAG additive.

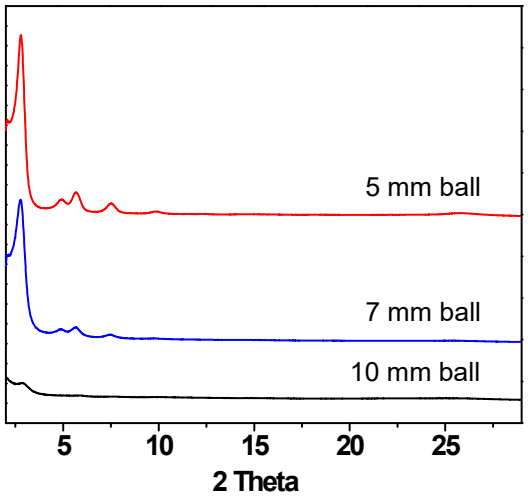


Figure S2. The effect of grinding ball size on the crystallinity of MC-DMTP-TPB COF, which was made by milling DMTP (20.4 mg, 0.105 mmol) with TPB (24.6 mg, 0.07 mmol) at 20 Hz for 1 hour using glacial acetic acid (15 μL) and acetonitrile (30 μL) as the LAG additive.

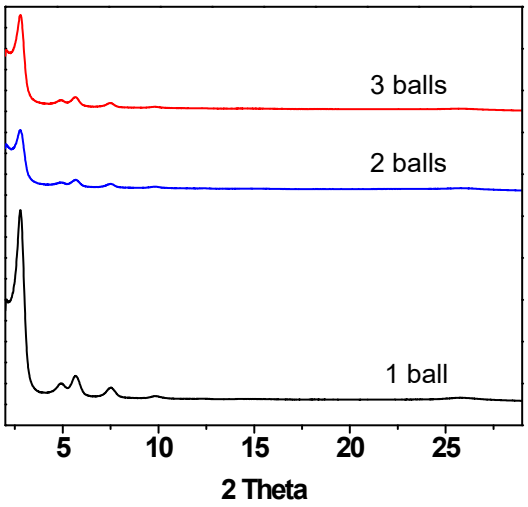


Figure S3. The effect of the number of the ball (5 mm in diameter) on the crystallinity of MC-DMTP-TPB COF, which was made by milling DMTP (20.4 mg, 0.105 mmol) with TPB (24.6 mg, 0.07 mmol) at 20 Hz for 1 hour using glacial acetic acid (15 μL) and acetonitrile (30 μL) as the LAG additive.

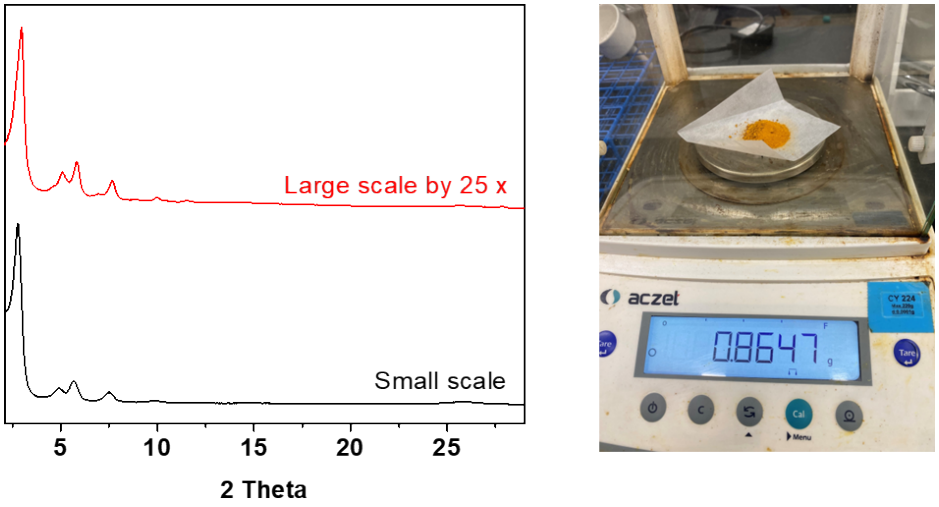


Figure S4. Scalable synthesis of MC-DMTP-TPB COF, which was made by milling DMTP (510 mg, 2.625 mmol) with TPB (615 mg,1.75 mmol) at 20 Hz for 1 hour using a 10-mm diameter stainless steel ball in the presence of glacial acetic acid (375 μL) and acetonitrile (750 μL). Inset digital photo shows the large-scale production of COF (0.8647g).

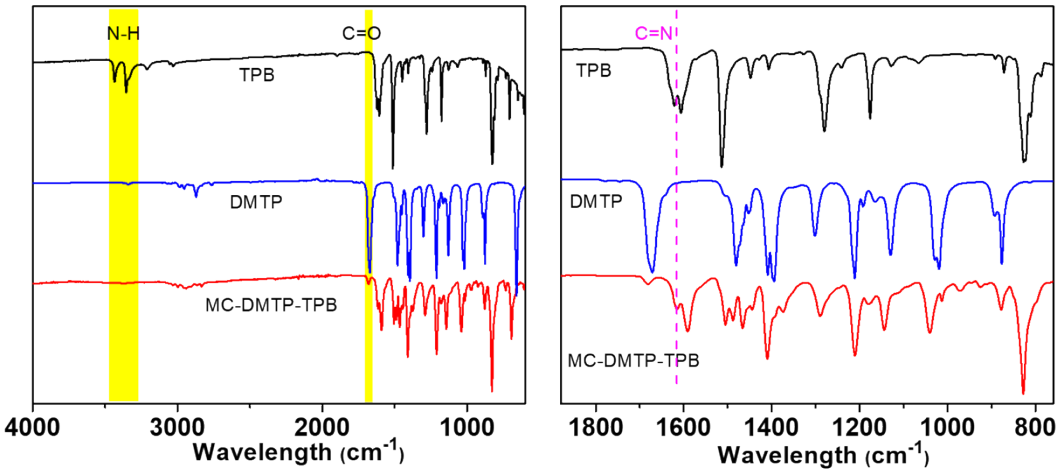


Figure S5. FTIR spectra of MC-DMTP-TPB COF and its monomers, TPB and DMTP.

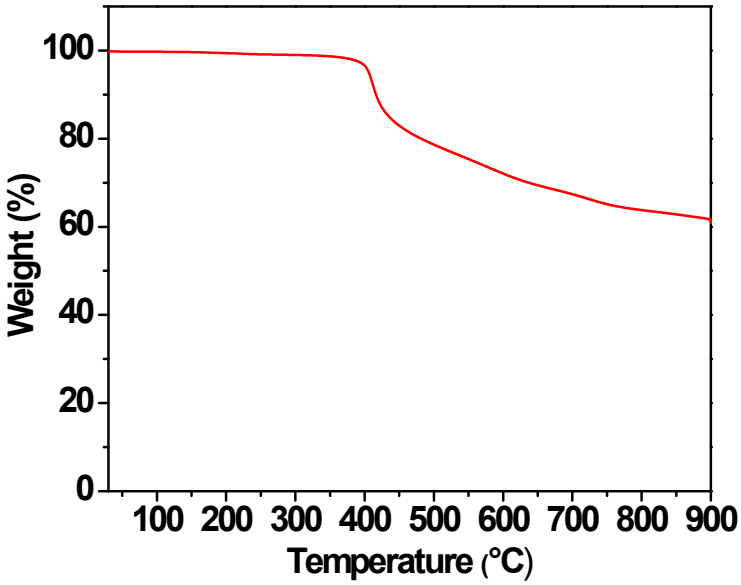


Figure S6. TGA curve of MC-DMTP-TPB COF under an N_2 atmosphere.

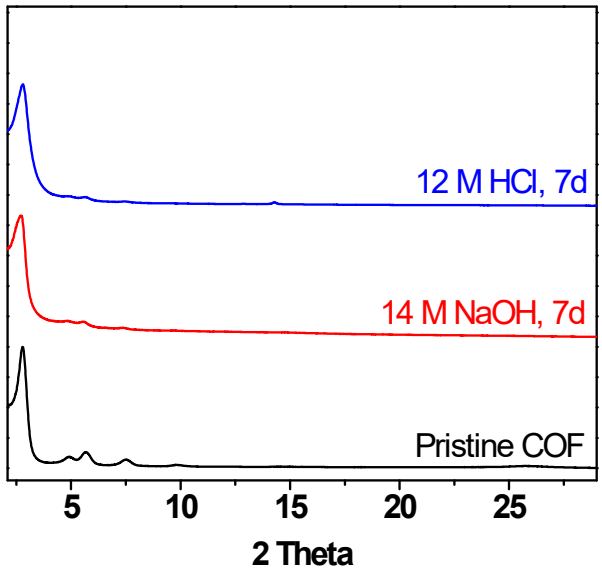


Figure S7. PXRD patterns of MC-DMTP-TPB COF after being exposed to 12 M HCl and 14 M NaOH aqueous solution for 7 days.

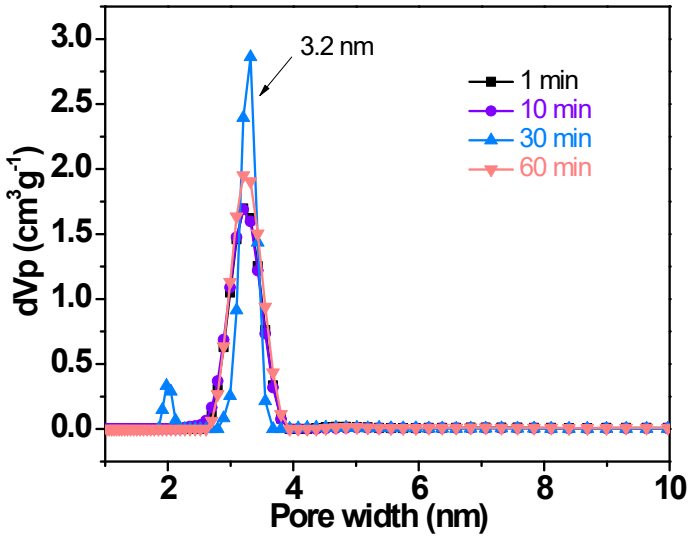


Figure S8. Pore size distribution profiles of MC-DMTP-TPB at different milling times, which were obtained by fitting the N_2 sorption isotherms using a quenched solid-state density functional theory (QSDFT) model.

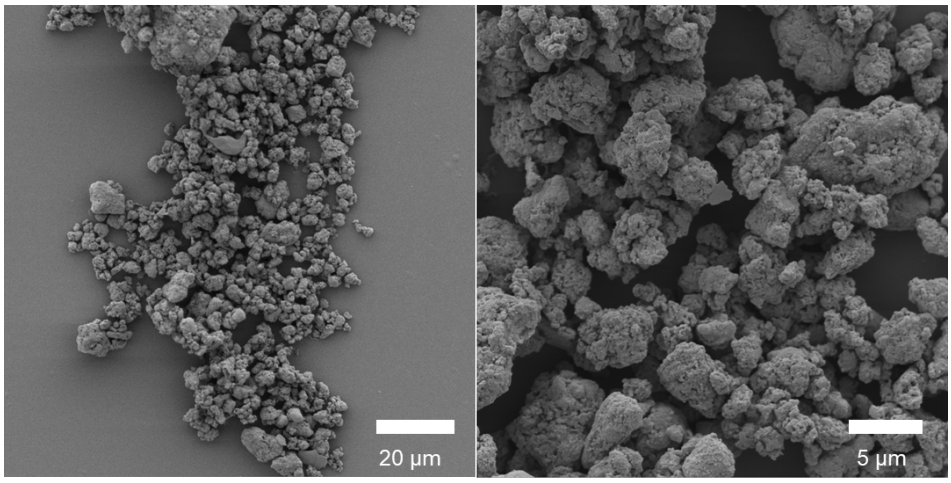


Figure S9. SEM images of MC-DMTP-TPB COF (1 h) at different magnifications.

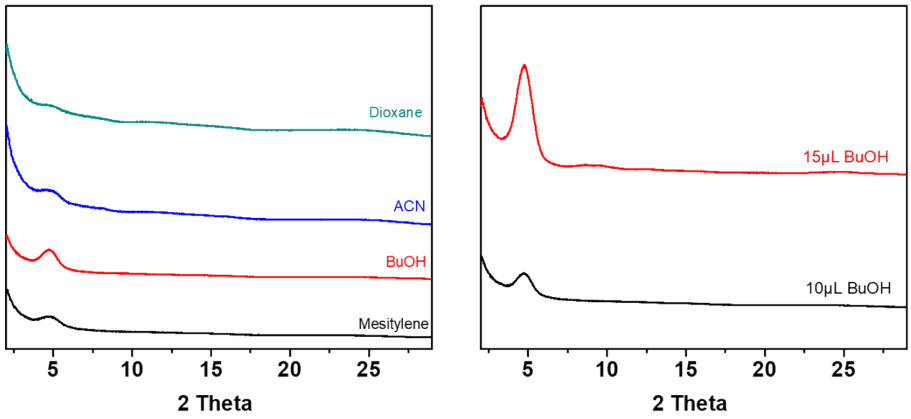


Figure S10. Condition screening for MC-TFB-PD-CH_3, which was made by milling 1,3,5-triformylbenzene (TFB) and 2,5-dimethyl-1,4-phenylenediamine (PD-CH_3) at 30 Hz for 1 hour using various LAG additives and glacial acetic acid with one 5 mm diameter stainless steel ball per jar.

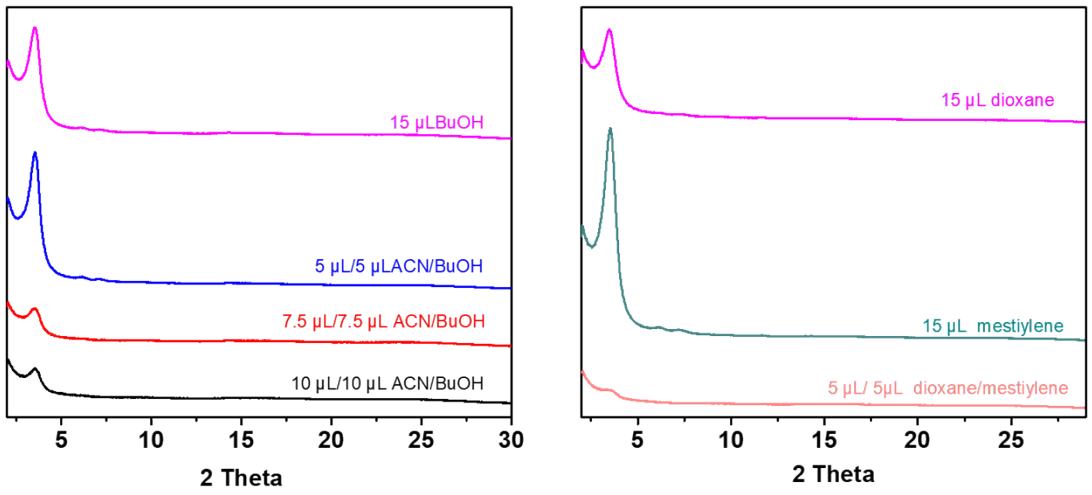


Figure S11. Condition screening for MC-TFB-BD-CH_3, which was constructed by milling 1,3,5-triformylbenzene (TFB) (11.3 mg, 0.07 mmol) and o-tolidine (22.3 mg, 0.105 mmol) at 30 Hz for 1 h using various LAG additive and glacial acetic acid (15 µL) with a 5 mm diameter stainless steel ball per jar.

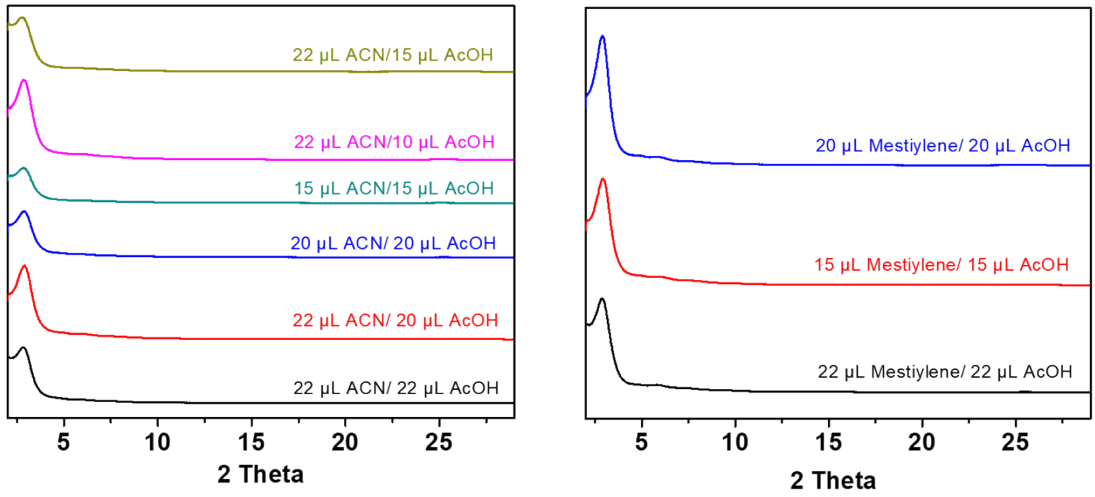


Figure S12. Condition screening for MC-DMTP-TTA, which was made by milling DMTP (20.4 mg, 0.105 mmol) and TTA (24.8 mg, 0.07 mmol) at 30 Hz for 1 hour using various LAG additives and glacial acetic acid with one 5 mm diameter stainless steel ball per jar.

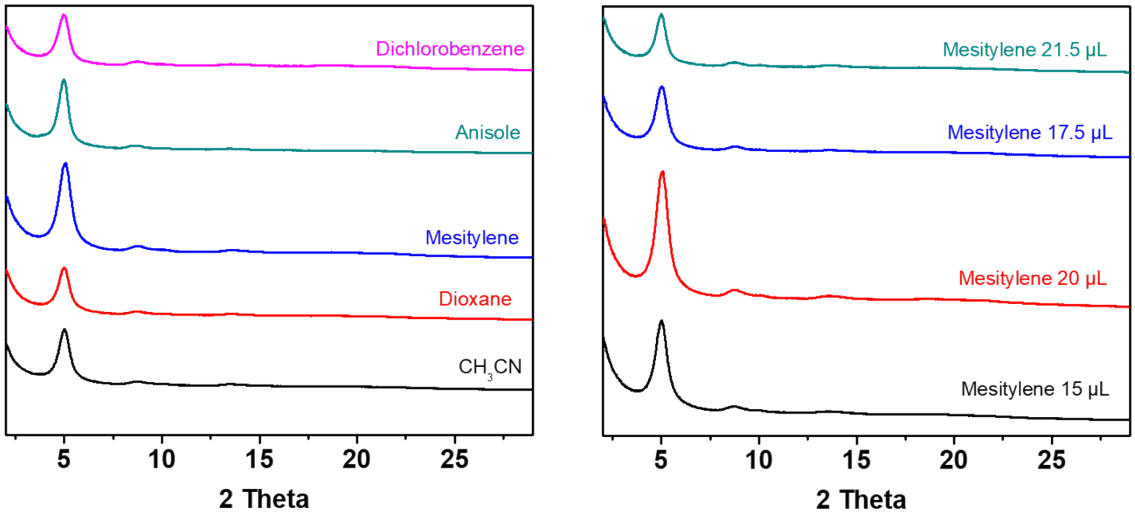


Figure S13. Condition screening for MC-TPA COF, which was made by milling tris(4-formylphenyl) amine and tris(4-aminophenyl) amine at 30 Hz for 1 hour using various LAG additives and glacial acetic acid with one 5 mm diameter stainless steel ball per jar.

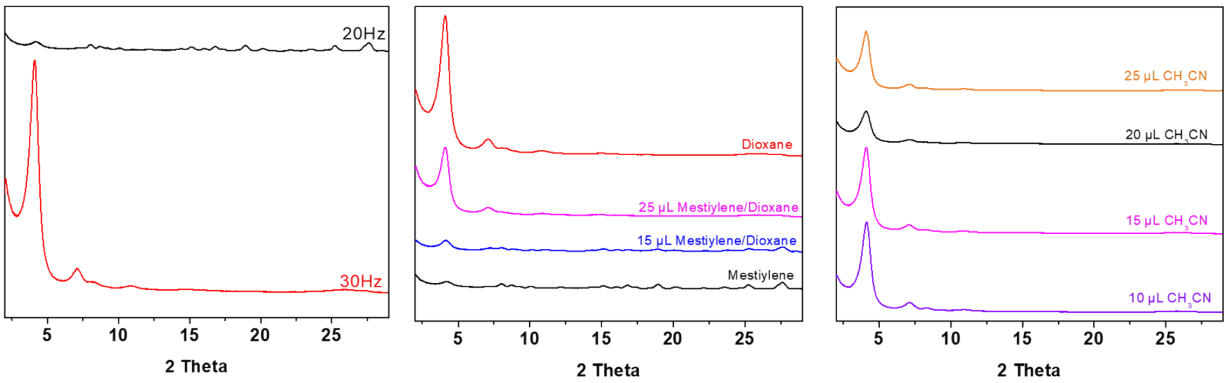


Figure S14. Condition screening for MC-TTA-TTB COF, which was made by milling TTA and TTB for 1 hour using various LAG additives and glacial acetic acid with one 5 mm diameter stainless steel ball.

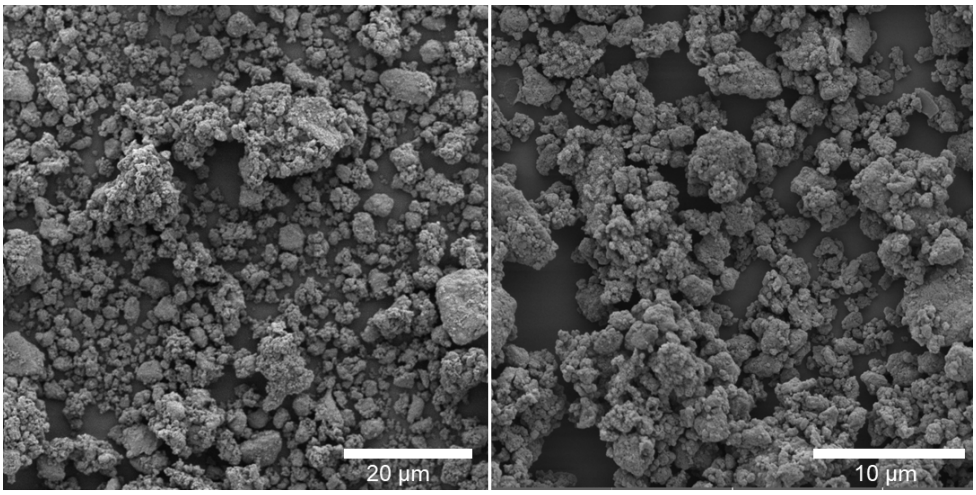


Figure S15. SEM images of MC-DMTP-TTA COF at different magnifications.

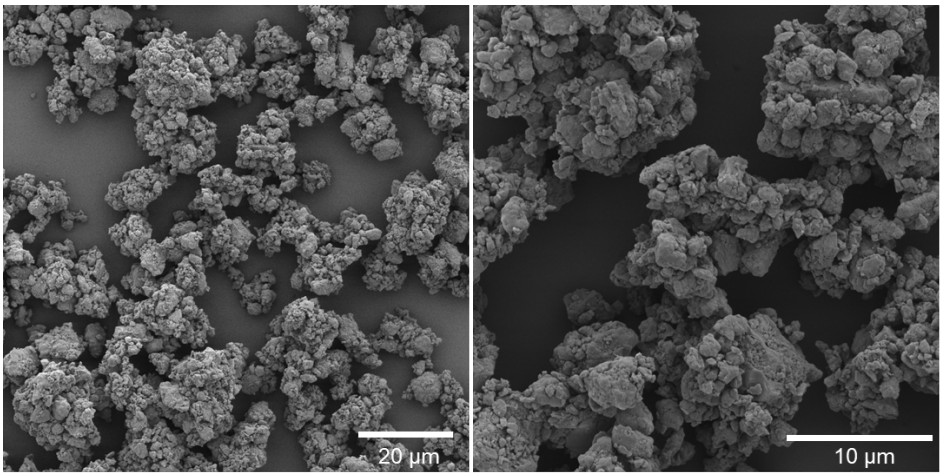


Figure S16. SEM images of MC-TFB-BD-CH_3 COF at different magnifications.

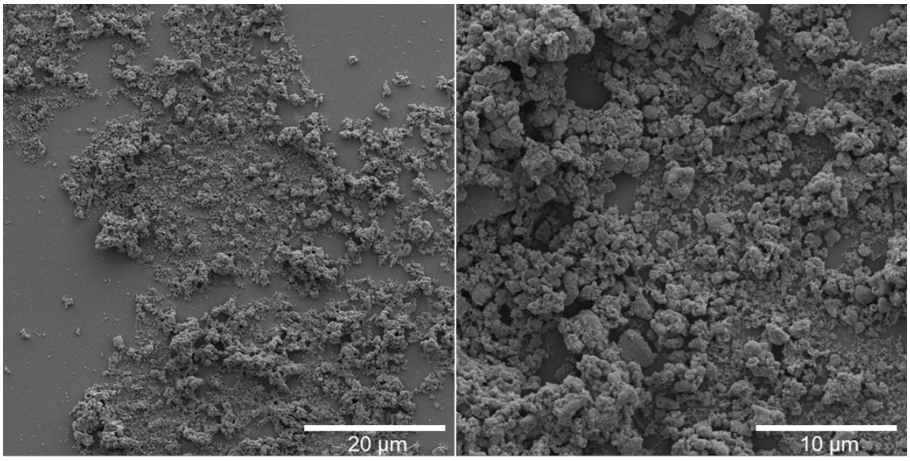


Figure S17. SEM images of MC-TFB-PD-CH_3 COF at different magnifications.

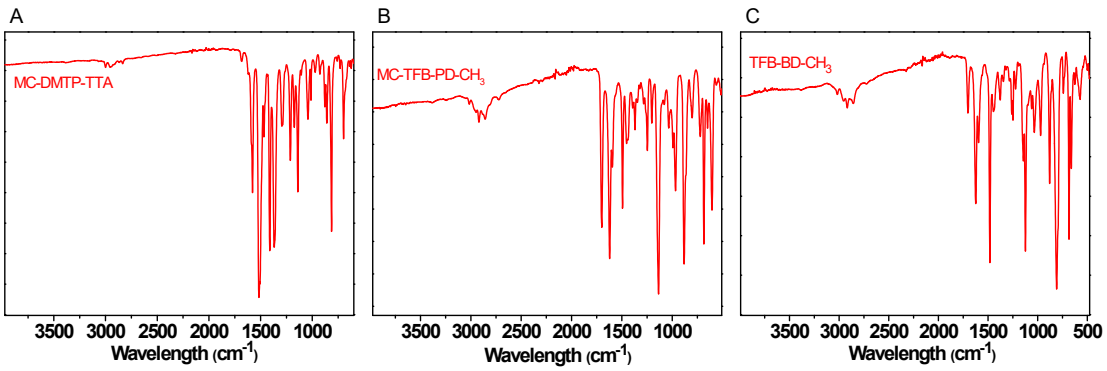


Figure S18. FTIR spectra of (A) MC-DMTP-TTA, (B) MC-TFB-BD-CH_3, and (C) MC-TFB-PD-CH_3.

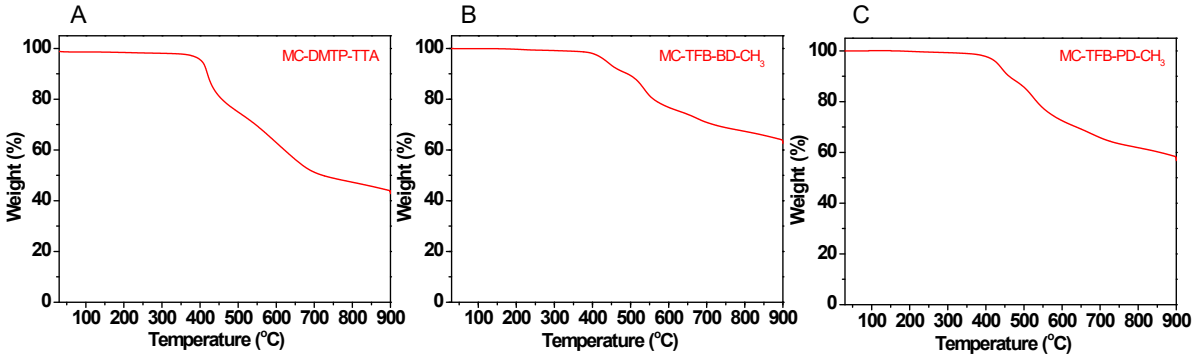


Figure S19. TGA curves of (A) MC-DMTP-TTA, (B) MC-TFB-BD-CH_3, and (C) MC-TFB-PD-CH_3 COFs under an N_2 atmosphere.

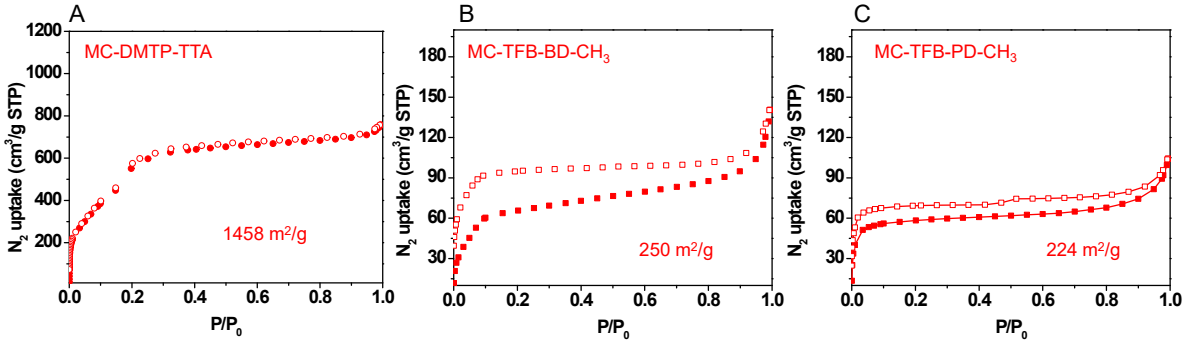


Figure S20. N_2 sorption isotherms of (A) MC-DMTP-TTA, (B) MC-TFB-BD-CH_3, and (C) MC-TFB-PD-CH_3 COFs.

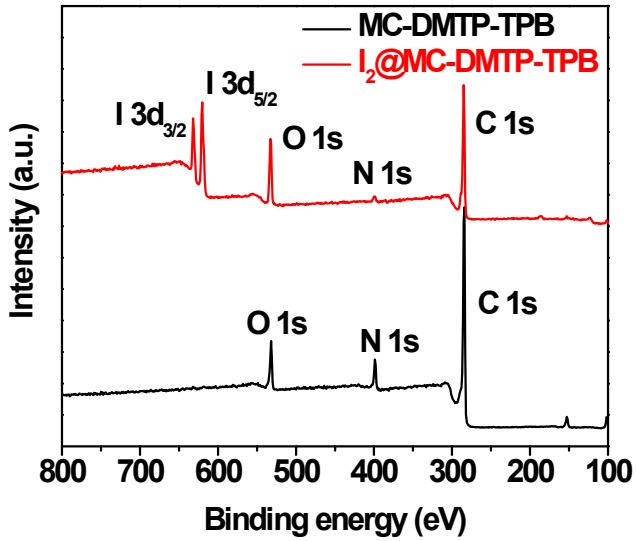


Figure S21. XPS survey spectra of MC-DMTP-TPB before and after iodine capture.

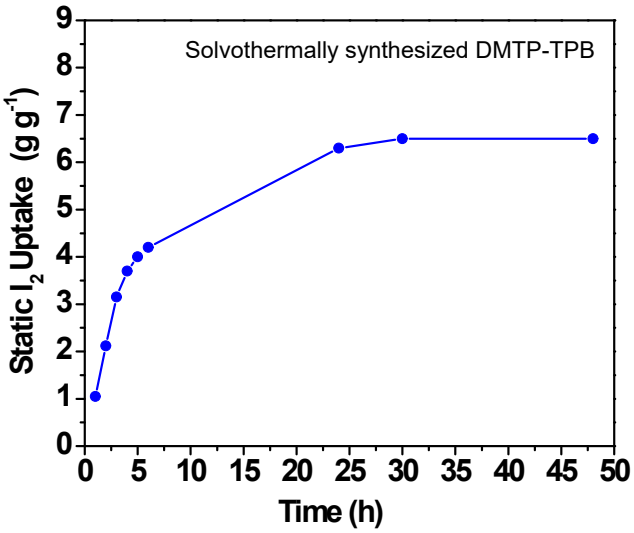


Figure S22. The gravimetric iodine adsorption capacity of solvothermally synthesized DMTP-TPB COF as a function of exposure time at 75 °C and ambient pressure. DMTP-TPB COF showed an iodine adsorption capacity of 6.4 g g^{-1}, which is consistent with the previous report.[2]

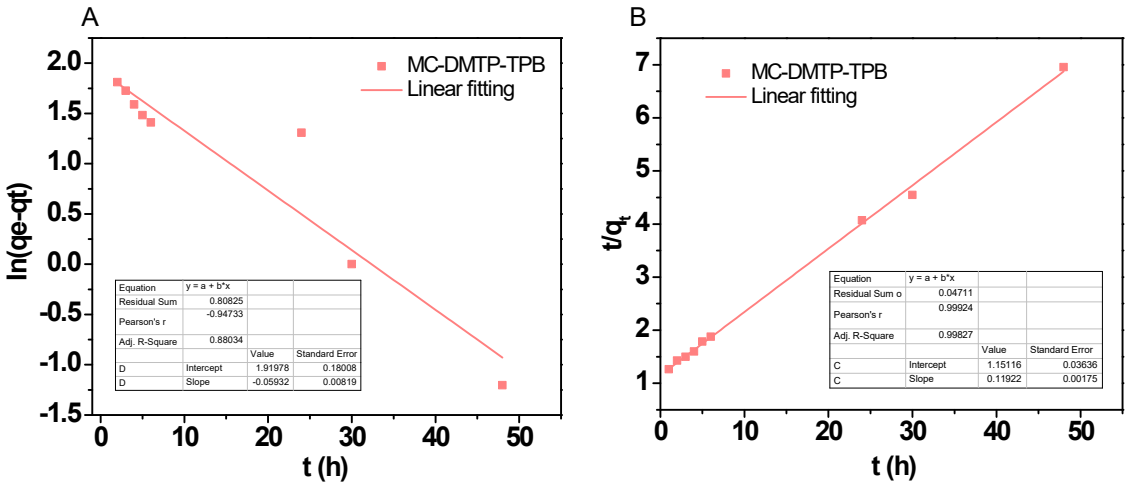


Figure S23. The fitting curves for (A) pseudo-first-order and (B) pseudo-second-order kinetic model of iodine adsorption on MC-DMTP-TPB COF.

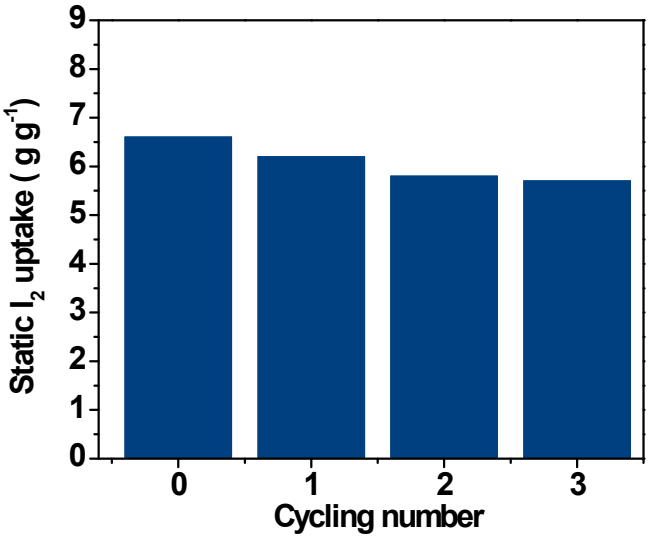


Figure S24. The recycling test of MC-DMTP-TPB COF in three successive adsorption-desorption cycles at 75 °C.

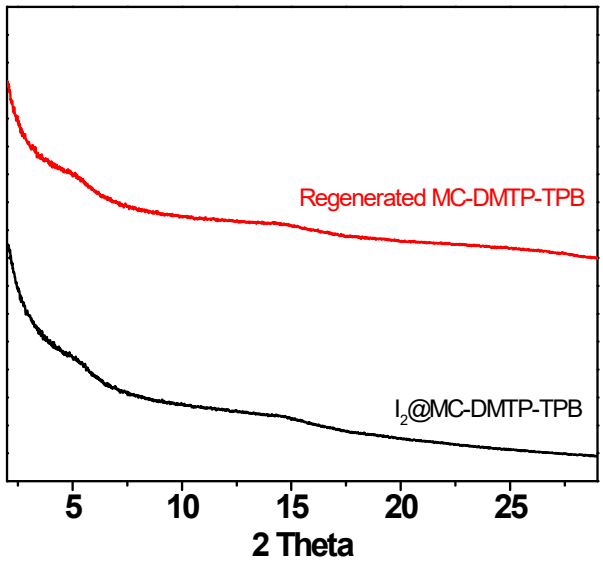


Figure S25. PXRD patterns of I_2@MC-DMTP-TPB (black curve) and regenerated MC-DMTP-TPB COF (red curve).

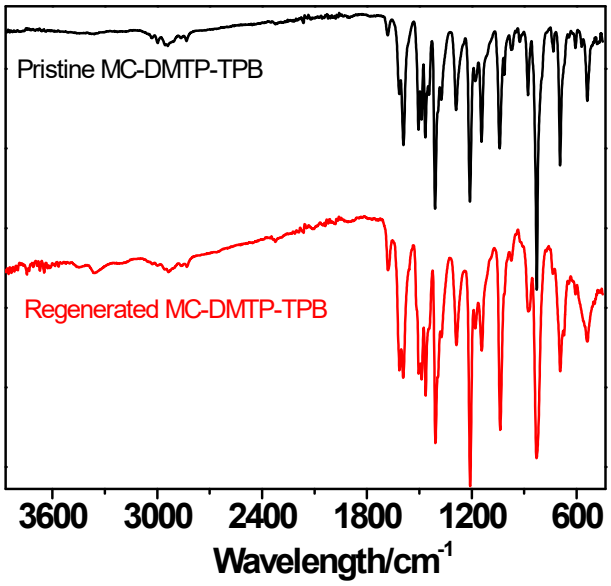


Figure S26. FTIR spectra of pristine MC-DMTP-TPB COF (black curve) and regenerated MC-DMTP-TPB COF (red curve).

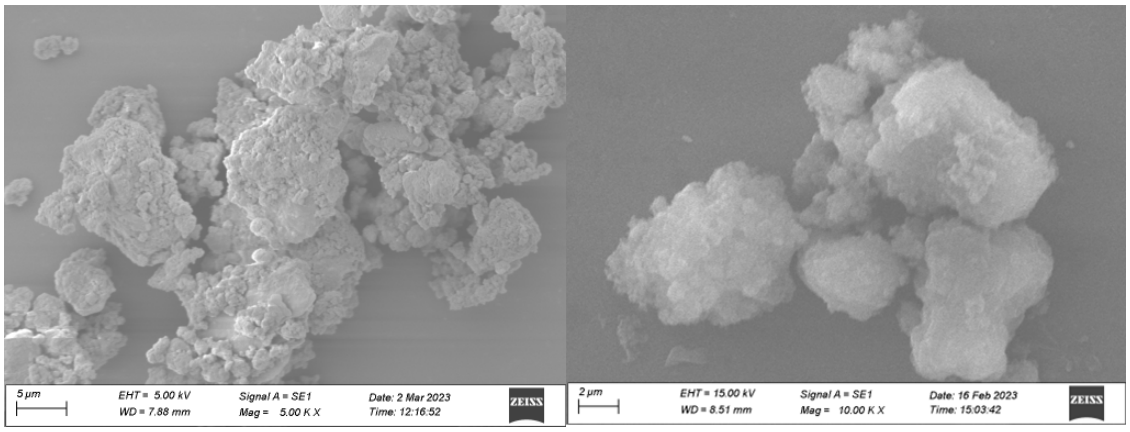


Figure S27. SEM images of regenerated MC-DMTP-TPB COF at different magnifications.

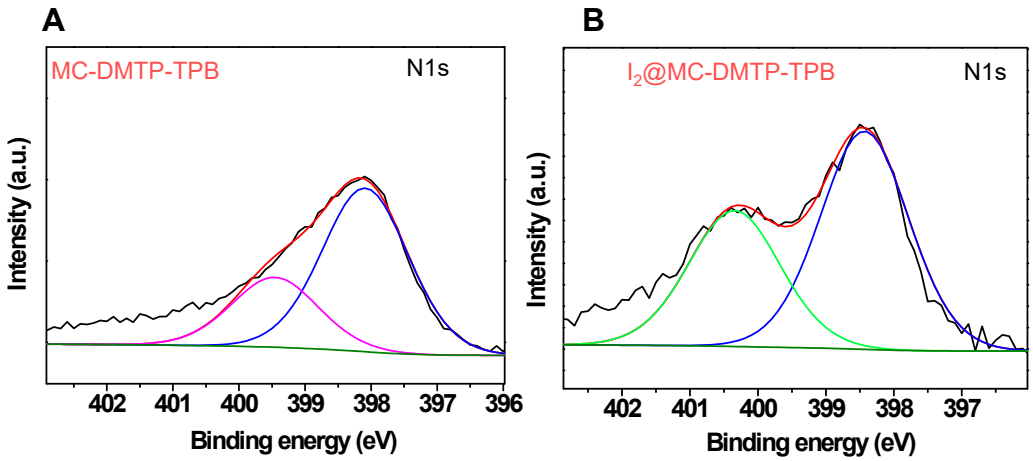


Figure S28. N 1s XPS spectra of (A) pristine MC-DMTP-TPB and (B) I_2@MC-DMTP-TPB. Note: The N1s peak centered at 399.5 eV is assigned to the unreacted -NH_2 at the defective edges of COF.

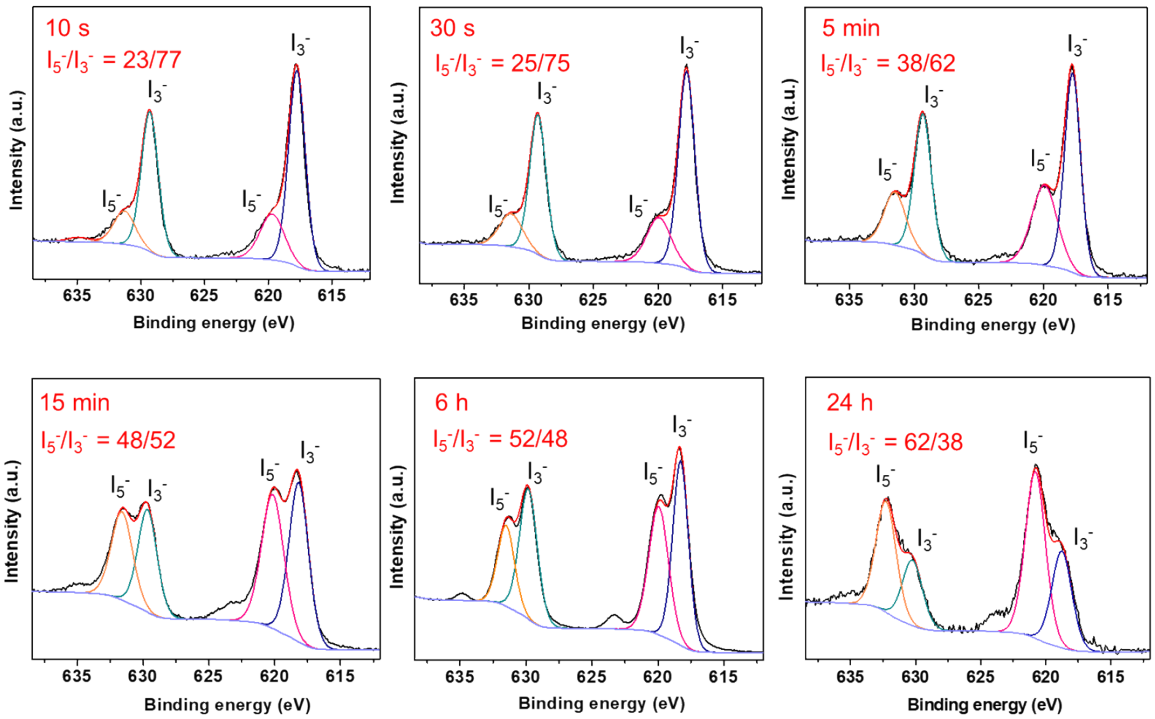


Figure S29. I3d XPS spectra of I_2@MC-DMTP-TPB after different exposure times of iodine vapor.

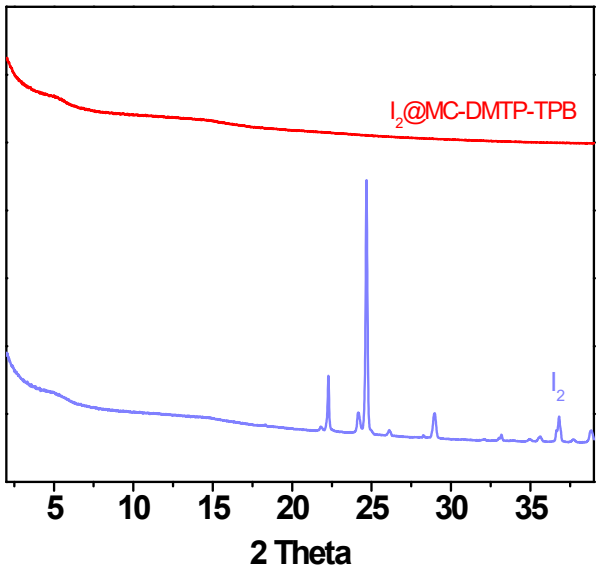


Figure S30. PXRD patterns of molecular iodine (blue curve) and I_2@MC-DMTP-TPB (red curve).

Table S1. Summary of reported COFs made rapidly (< 1 h) at ambient temperature.

Entry	COFs	Temperature	Condition	Time	S_{BET} ($m^2 g^{-1}$)	Activation	Reference
1	MC-DMTP-TPB	RT	Air, ball mill nearly solventless AcOH	1 min	1387	Vacuum activation	This work
				10 min	1400		
				30 min	1517		
				60 min	1554		
2	TpBa series	RT Then 170 °C	Air, ball mill *p*-toluenesulfonic acid	1 min then 1 h	538-3109	Vacuum activation	3
3	TAPB-PDA	20 °C	Air Mesitylene/dioxane $Sc(OTf)_3$	10 min	2175	Supercritical CO_2 activation	4
4	sonoCOF-1	RT	Air, ultrasound 6 M AcOH	60 min	2059	Vacuum activation	5
5	UV-COF-5	RT	Air, UV Mesitylene/dioxane	60 min	2026	Vacuum activation	6
6	JUC-523	RT	Air, H_2O AcOH	30 min	1435	Vacuum activation	7
7	TpBD	RT	Air, EtOH AcOH	30 min	885	Vacuum activation	8
8	EB-COF-1	RT	N_2, 100 kGy 2-dichlorobenzene/*n*-butanol 6 M AcOH	160 s	738	Vacuum activation	9
9	COF-42-B	RT	Air Mesitylene/dioxane $Sc(OTf)_3$	30 min	367	Vacuum activation	10
10	RT-COF-1	RT	Air, DMSO AcOH	1 min	329	Vacuum activation	11
11	TpPa-1 (MC)	RT	Air, solventless grinding	40 min	61	Vacuum activation	12

Table S2. Summary of static iodine vapor capture capacities and synthetic conditions of high-performing COF adsorbents.

Entry	COF adsorbents	Temperature (°C)	S_{BET} ($m^2 g^{-1}$)	I_2 uptake ($g g^{-1}$)	Synthetic condition	Reference
1	MC-DMTP-TPB	75	1554	6.9	RT, 1 h, ball mill Air, nearly solventless	This work
	MC-DMTP-TTA		1458	7.1		
	MC-TPB-BD-CH_3		250	6.5		
	MC-TPB-PD-CH_3		223	6.4		
2	iCOF-AB-50	75	1390	10.21	1. 120 °C, 3 d, vacuum 2. PSM, 140 °C, 3 d	13
3	iCOF-AB-33	75	1580	9.00		
4	COF-TAPT	75	2348	8.61	120 °C, 3 d, vacuum	14
5	JUC-561	75	2359	8.19	120 °C, 3 d, vacuum	15
6	COFA-1 aerogel	75	254.6	8.15	RT, 1 d, freeze-dry	16
7	COF-TAPB	75	2290	7.94	120 °C, 3 d, vacuum	14
8	Mw-TPB-BD-CH_3	75	210	7.83	90 °C, 1 h microwave, air	17
9	TAPB-PDA aerogel	75	2273	7.7	80 °C, 12 h $ScCO_2$ activation	18
10	COF-AB-33	75	2172	6.81	120 °C, 3 d, vacuum	13
11	COF-AB-50	75	2103	6.49		
12	QTD-COF-V	75	/	6.29	1. PSM, RT, 2 d 2. 120 °C, 7 d	19
13	DMTP-TPB	77	1927	6.26	120 °C, 3 d, vacuum	2
14	C-TP-BPDA-COF	75	436	6.11	120 °C, 3 d, vacuum	20
15	SCU-COF-2	75	413.4	6.0	120 °C, 3 d, vacuum	21
16	PB-TT COF	75	1305.3	5.97	120 °C, 3 d, vacuum	22
17	JUC-609	75	1012.81	5.90	120 °C, 3 d, vacuum	23
18	USTB-1c	75	1454	5.80	120 °C, 3 d, vacuum	24
19	TJNU-201	77	2510	5.625	1. PSM, 65 °C, 30 h 2. 120 °C 3 d, vacuum	25

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