An imidazole-based covalent-organic framework enabling a super-efficiency in sunlight-driving uranium extraction from seawater

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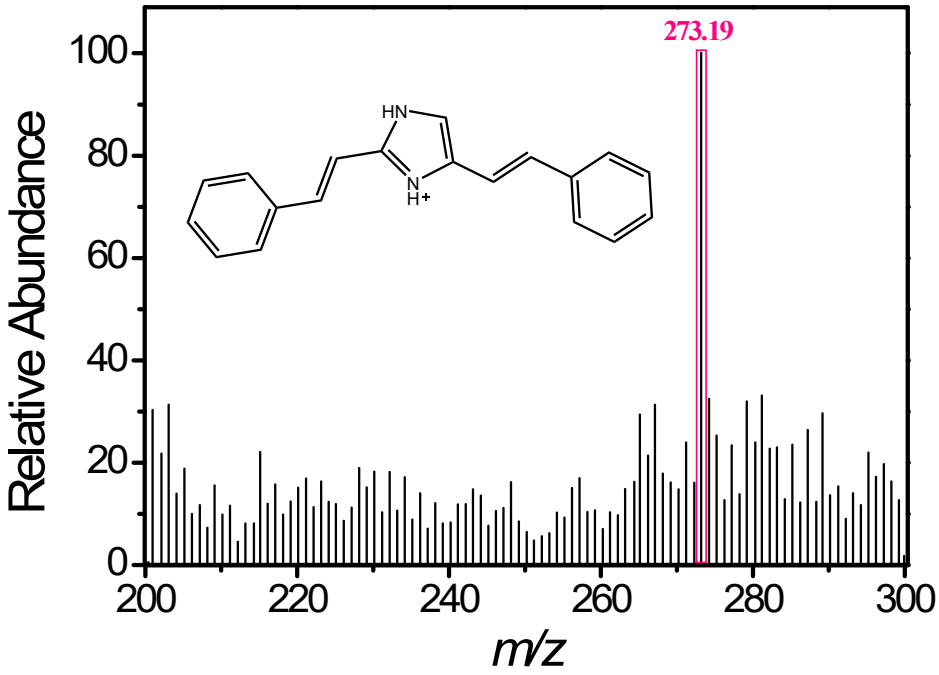

Fig. S1 MS of model compound.

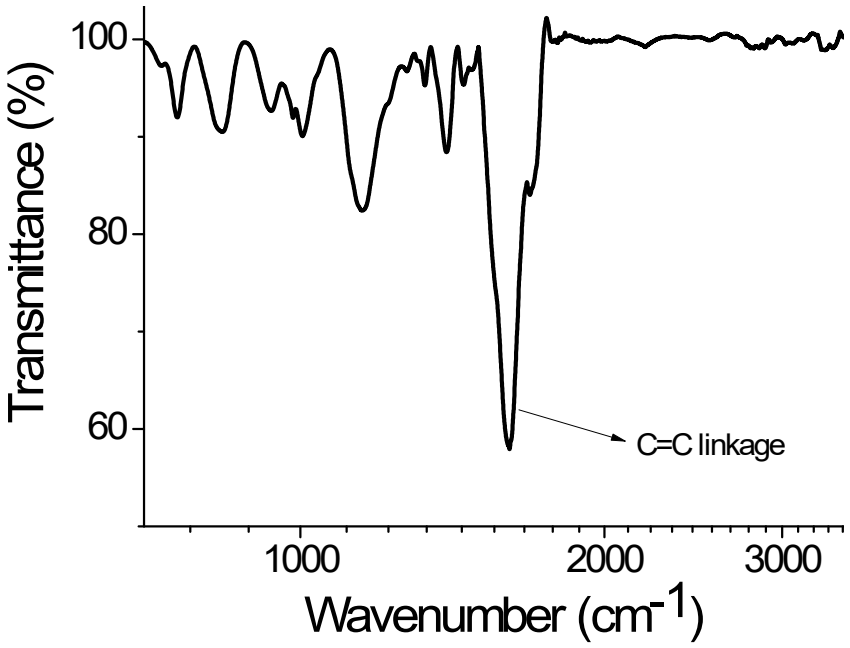

Fig. S2 IR of BI-COF.

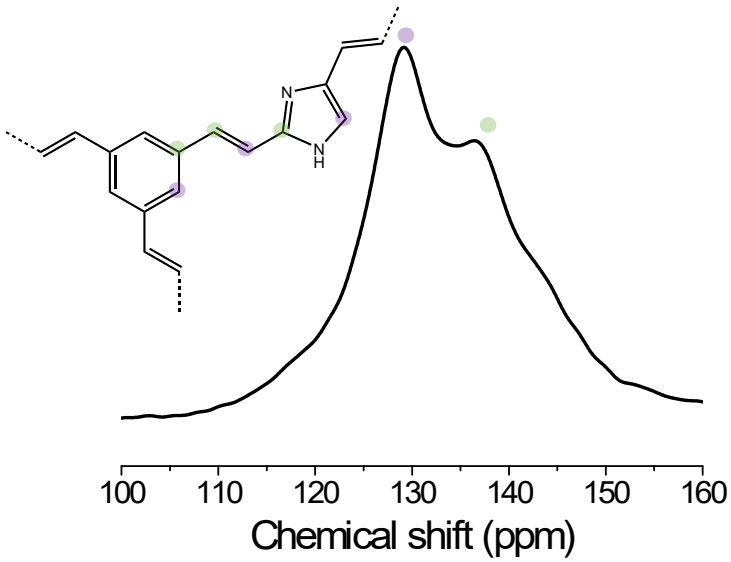

Fig. S3 CP-MAS ^{13}C NMR of BI-COF.

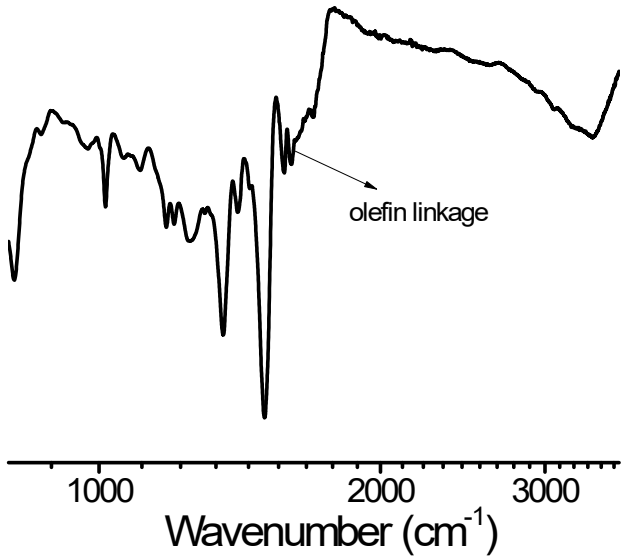

Fig. S4 IR of TI-COF.

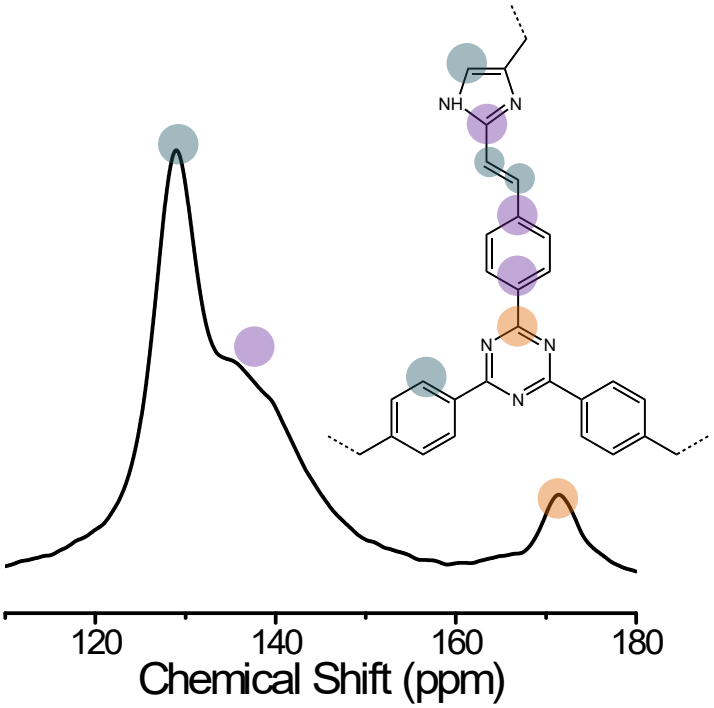

Fig. S5 CP-MAS ^{13}C NMR of TI-COF.

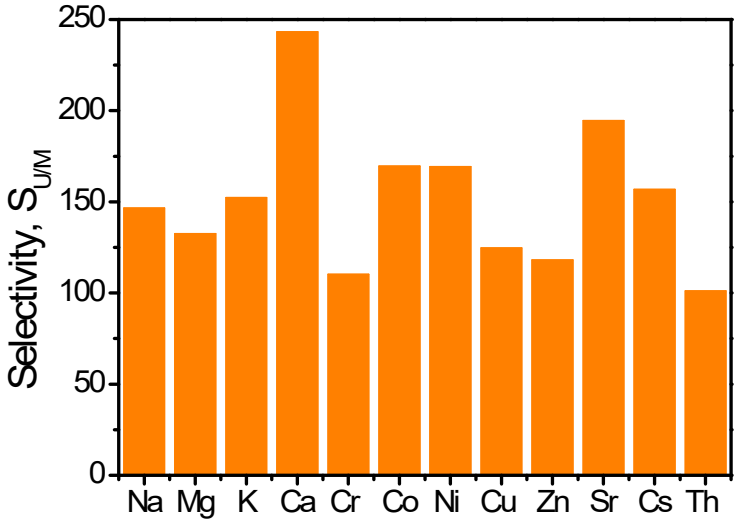

Fig. S6 Selectivity of $S_{U/M}$.

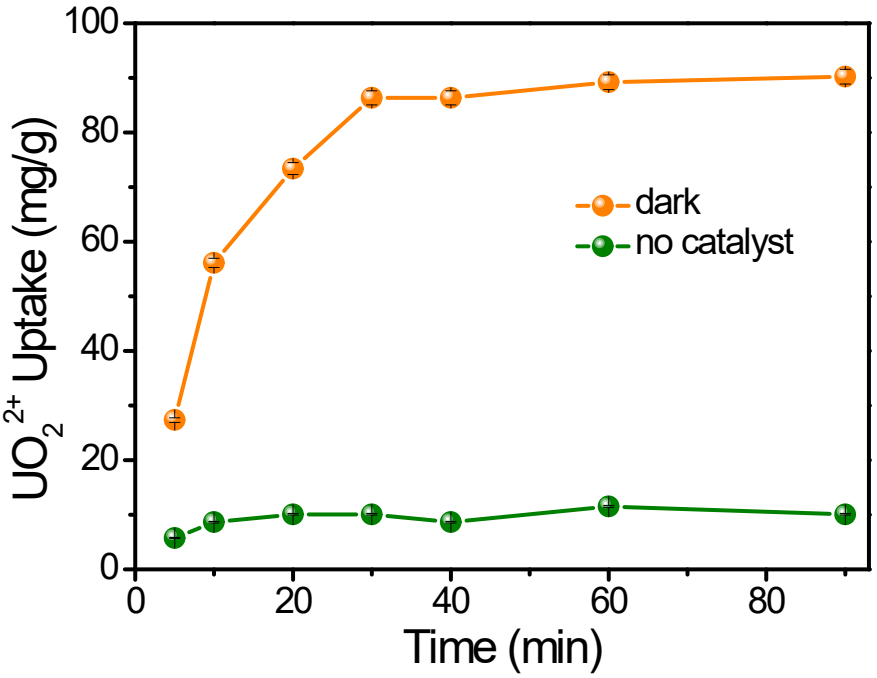

Fig. S7 Uranium adsorption under controlled conditions upon TI-COF. The error bars is based on the average vaule of three times.

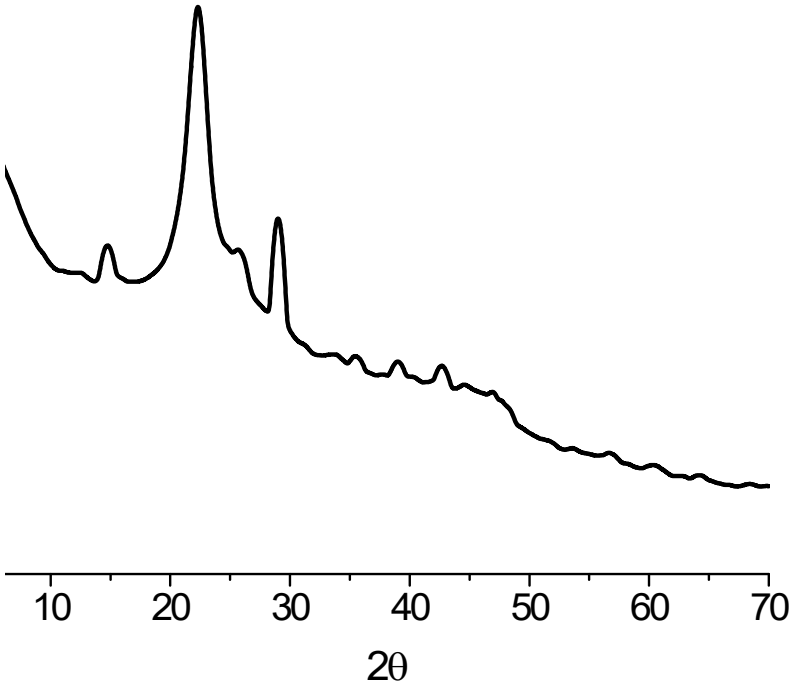

Fig. S8 The PXRD patterns of the uranium-extracted samples after photocatalysis.

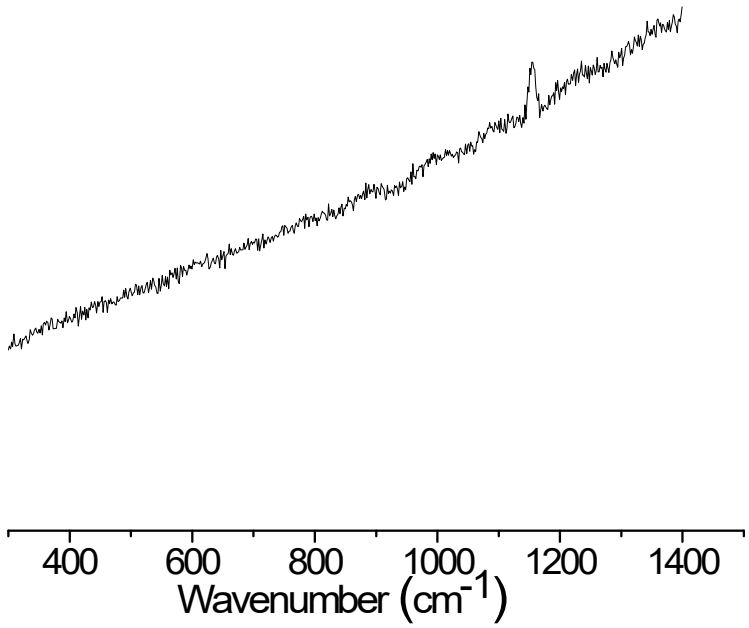

Fig. S9 The Raman spectra of the uranium-extracted samples after photocatalysis.

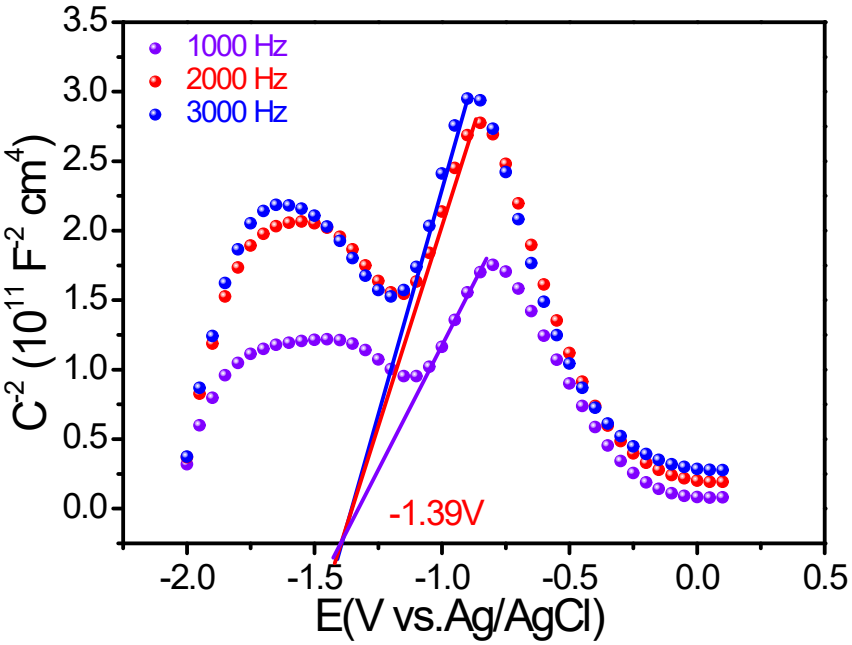

Fig. S10 Mott-Schottky plots of BI-COF.

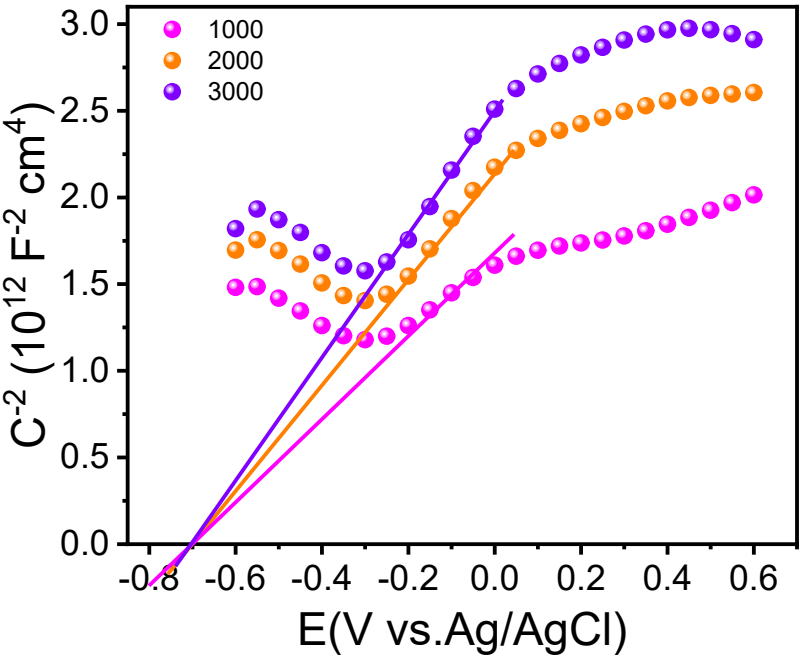

Fig. S11 Mott-Schottky plots of TI-COF.

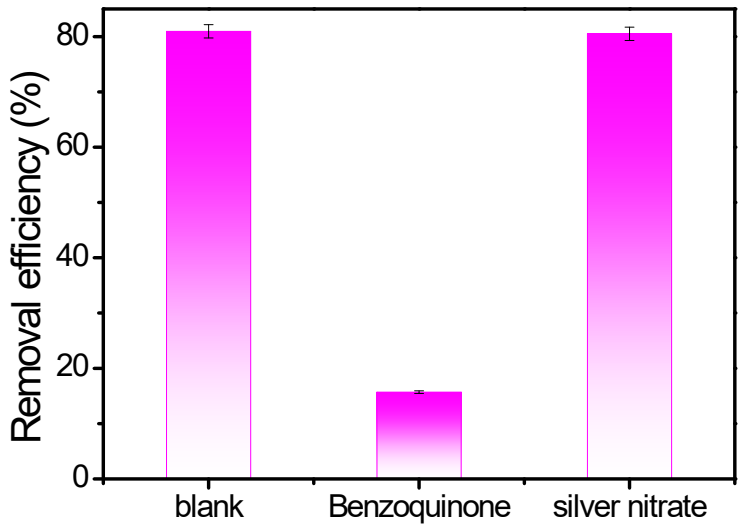

Fig. S12 The active intermediate test. The error bars is based on the average vaule of three times.

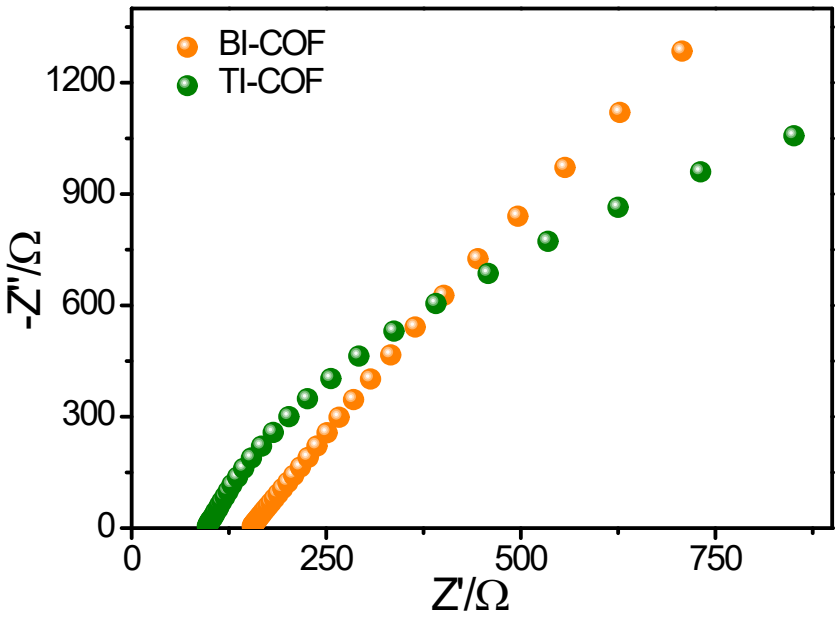

Fig. S13 Impedance plots of TI-COF and BI-COF.

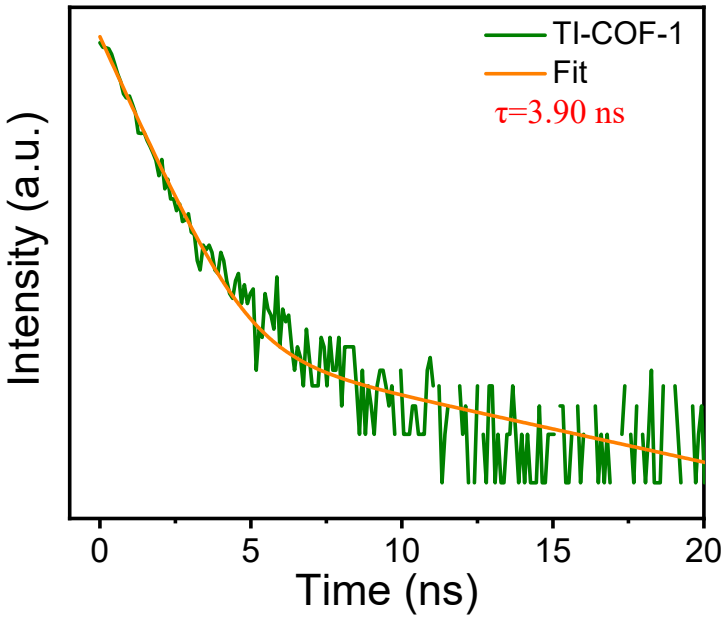

Fig. S14 Fluorescence lifetimes of TI-COF.

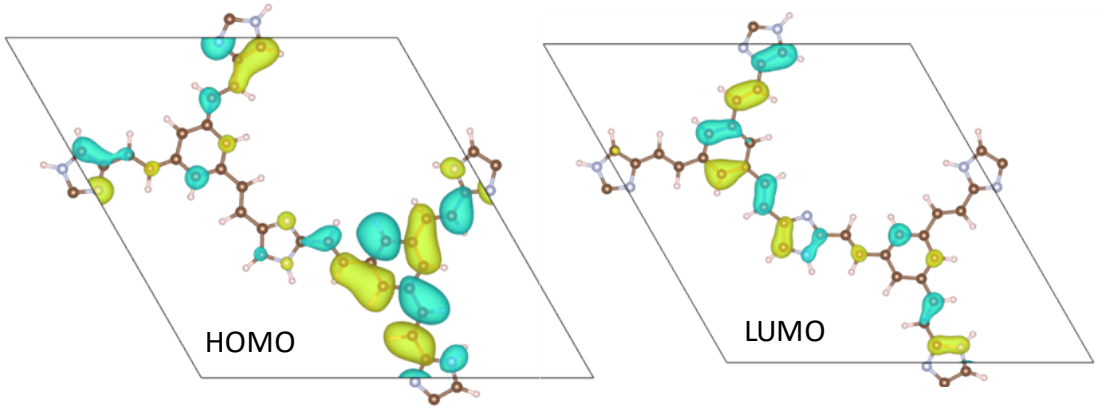

Fig. S15 View of the HOMO and LUMO levels of BI-COF.

Fig. S16 View of the HOMO and LUMO levels of BI-COF and TI-COF, and their photocatalytic mechanism.

Table S1. Structural data of BI-COF.

```
data_ BI-COF
_audit_creation_date                    2023-10-15
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_symmetry_cell_setting                  hexagonal
loop_
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   -y,x-y,z
   -x+y,-x,z
   x,y,-z
   -y,x-y,-z
   -x+y,-x,-z
_cell_length_a                          20.8009
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_cell_length_c                          3.4407
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_atom_site_fract_z
_atom_site_U_iso_or_equiv
_atom_site_adp_type
_atom_site_occupancy
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C3      C       0.37609     0.43837     0.50000     0.00000     Uiso    1.00
C4      C       0.33671     0.36202     0.50000     0.00000     Uiso    1.00
N5      N       0.38541     0.33760     0.50000     0.00000     Uiso    1.00
C6      C       0.52474     0.40080     0.50000     0.00000     Uiso    1.00
C7      C       0.34332     0.48678     0.50000     0.00000     Uiso    1.00
C8      C       0.53010     0.33915     0.50000     0.00000     Uiso    1.00
C9      C       0.40908     0.69007     0.50000     0.00000     Uiso    1.00
C10     C       0.38618     0.74332     0.50000     0.00000     Uiso    1.00
C11     C       0.38572     0.56095     0.50000     0.00000     Uiso    1.00
C12     C       0.73134     0.32859     0.50000     0.00000     Uiso    1.00
C13     C       0.73686     0.39886     0.50000     0.00000     Uiso    1.00
H14     H       0.27697     0.32671     0.50000     0.00000     Uiso    1.00
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H17	H	0.28355	0.45969	0.50000	0.00000	Uiso	1.00
H18	H	0.47968	0.28579	0.50000	0.00000	Uiso	1.00
H19	H	0.46725	0.70685	0.50000	0.00000	Uiso	1.00
H20	H	0.44548	0.58476	0.50000	0.00000	Uiso	1.00
H21	H	0.78047	0.32352	0.50000	0.00000	Uiso	1.00

Table S2. Structural data of TI-COF.

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_symmetry_cell_setting hexagonal
loop_
_symmetry_equiv_pos_as_xyz
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-x+y,-x,z
x,y,-z
-y,x-y,-z
-x+y,-x,-z
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_cell_length_b 34.0869
_cell_length_c 6.6018
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loop_
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_atom_site_fract_z
_atom_site_U_iso_or_equiv
_atom_site_adp_type
_atom_site_occupancy

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N2	N	0.41642	0.42199	-0.00000	0.00000	Uiso	1.00
C3	C	0.37221	0.40958	-0.00000	0.00000	Uiso	1.00
C4	C	0.34625	0.36298	-0.00000	0.00000	Uiso	1.00

N5	N	0.37438	0.34601	-0.00000	0.00000	Uiso	1.00
C6	C	0.45931	0.38170	-0.00000	0.00000	Uiso	1.00
C7	C	0.35533	0.44186	-0.00000	0.00000	Uiso	1.00
N8	N	0.37675	0.67562	-0.00000	0.00000	Uiso	1.00
C9	C	0.36808	0.71043	-0.00000	0.00000	Uiso	1.00
C10	C	0.35220	0.59416	-0.00000	0.00000	Uiso	1.00
C11	C	0.39679	0.79396	-0.00000	0.00000	Uiso	1.00
C12	C	0.45123	0.76816	-0.00000	0.00000	Uiso	1.00
C13	C	0.48673	0.81315	-0.00000	0.00000	Uiso	1.00
C14	C	0.43216	0.83880	-0.00000	0.00000	Uiso	1.00
C15	C	0.47753	0.84910	-0.00000	0.00000	Uiso	1.00
C16	C	0.75542	0.45703	-0.00000	0.00000	Uiso	1.00
C17	C	0.79242	0.41226	-0.00000	0.00000	Uiso	1.00
C18	C	0.83461	0.45182	-0.00000	0.00000	Uiso	1.00
C19	C	0.79766	0.49667	-0.00000	0.00000	Uiso	1.00
C20	C	0.83788	0.49476	-0.00000	0.00000	Uiso	1.00
N21	N	0.70520	0.33112	-0.00000	0.00000	Uiso	1.00
C22	C	0.70772	0.37217	-0.00000	0.00000	Uiso	1.00
C23	C	0.66199	0.24766	-0.00000	0.00000	Uiso	1.00
C24	C	0.51331	0.89765	-0.00000	0.00000	Uiso	1.00
C25	C	0.46186	0.34373	-0.00000	0.00000	Uiso	1.00
H26	H	0.30965	0.34285	-0.00000	0.00000	Uiso	1.00
H27	H	0.36486	0.31177	-0.00000	0.00000	Uiso	1.00
H28	H	0.48951	0.41455	-0.00000	0.00000	Uiso	1.00
H29	H	0.31912	0.42818	-0.00000	0.00000	Uiso	1.00
H30	H	0.36230	0.78755	-0.00000	0.00000	Uiso	1.00
H31	H	0.45945	0.74131	-0.00000	0.00000	Uiso	1.00
H32	H	0.52097	0.81915	-0.00000	0.00000	Uiso	1.00
H33	H	0.42391	0.86565	-0.00000	0.00000	Uiso	1.00
H34	H	0.72520	0.45991	-0.00000	0.00000	Uiso	1.00
H35	H	0.79134	0.37997	-0.00000	0.00000	Uiso	1.00
H36	H	0.86403	0.44781	-0.00000	0.00000	Uiso	1.00
H37	H	0.79889	0.52905	-0.00000	0.00000	Uiso	1.00
H38	H	0.50168	0.92201	-0.00000	0.00000	Uiso	1.00
H39	H	0.43082	0.31132	-0.00000	0.00000	Uiso	1.00
C40	C	0.74968	0.04956	0.50000	0.00000	Uiso	1.00
N41	N	0.74952	0.08898	0.50000	0.00000	Uiso	1.00
C42	C	0.70529	0.07653	0.50000	0.00000	Uiso	1.00
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C46	C	0.68839	0.10874	0.50000	0.00000	Uiso	1.00
N47	N	0.71006	0.34229	0.50000	0.00000	Uiso	1.00
C48	C	0.70138	0.37705	0.50000	0.00000	Uiso	1.00

C49	C	0.68550	0.26094	0.50000	0.00000	Uiso	1.00
C50	C	0.72999	0.46045	0.50000	0.00000	Uiso	1.00
C51	C	0.78441	0.43460	0.50000	0.00000	Uiso	1.00
C52	C	0.81991	0.47951	0.50000	0.00000	Uiso	1.00
C53	C	0.76538	0.50524	0.50000	0.00000	Uiso	1.00
C54	C	0.81072	0.51545	0.50000	0.00000	Uiso	1.00
C55	C	1.08905	0.12379	0.50000	0.00000	Uiso	1.00
C56	C	1.12598	0.07897	0.50000	0.00000	Uiso	1.00
C57	C	1.16823	0.11851	0.50000	0.00000	Uiso	1.00
C58	C	1.13136	0.16344	0.50000	0.00000	Uiso	1.00
C59	C	1.17157	0.16149	0.50000	0.00000	Uiso	1.00
N60	N	1.03851	-0.00228	0.50000	0.00000	Uiso	1.00
C61	C	1.04112	0.03884	0.50000	0.00000	Uiso	1.00
C62	C	0.99518	-0.08586	0.50000	0.00000	Uiso	1.00
C63	C	0.84648	0.56392	0.50000	0.00000	Uiso	1.00
C64	C	0.79510	0.01077	0.50000	0.00000	Uiso	1.00
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H66	H	0.69799	-0.02121	0.50000	0.00000	Uiso	1.00
H67	H	0.82272	0.08159	0.50000	0.00000	Uiso	1.00
H68	H	0.65217	0.09506	0.50000	0.00000	Uiso	1.00
H69	H	0.69552	0.45404	0.50000	0.00000	Uiso	1.00
H70	H	0.79259	0.40772	0.50000	0.00000	Uiso	1.00
H71	H	0.85413	0.48547	0.50000	0.00000	Uiso	1.00
H72	H	0.75717	0.53212	0.50000	0.00000	Uiso	1.00
H73	H	1.05886	0.12673	0.50000	0.00000	Uiso	1.00
H74	H	1.12486	0.04665	0.50000	0.00000	Uiso	1.00
H75	H	1.19765	0.11449	0.50000	0.00000	Uiso	1.00
H76	H	1.13263	0.19583	0.50000	0.00000	Uiso	1.00
H77	H	0.83487	0.58829	0.50000	0.00000	Uiso	1.00
H78	H	0.76405	-0.02162	0.50000	0.00000	Uiso	1.00

Table S3. A comparison of U uptake capacity among established adsorbents/photocatalysts and our case.

Adsorbents	U uptake capacity (mg/g)	Reference
TI-COF	**902**	**Our case**
i-MZIF90(50)	1.3 g/g	Energy Environ. Sci., 2022,15, 3462
USC-CP-1	562	Angew. Chem. Int. Ed. 2019,58,52
POP-pNH_2-AO	580	Nat. Commun. 2018, 9,1644
MUU_{re}	475	Angew. Chem. Int. Ed. 2022,61,82
COF-TpDb-AO	408	Adv. Mater. 2018, 1705479
SCU-19	557	Angew. Chem. Int. Ed. 2019,58,45
SZ-2/SZ-3	58	Nat. Commun. 2017, 8, 15369
MISS-PAF-1	79.8	ACS Cent. Sci. 2019, 5,1432
PIDO NF	860	Adv. Energy Mater. 2018,8,1802607
COF-TpAb-AO	127	Adv. Mater. 2018, 30, 1705479
MIPAF-11c	14.5	Adv. Mater. 2018, 30, 1706507
NDA-TN-AO	600	Angew. Chem. Int. Ed. 2020,59,40

Table S4. A comparison of U adsorption kinetics among established photocatalysts and our case.

Adsorbents	Adsorption equilibrium time (min)	Reference
TI-COF	**40**	**Our case**
i-MZIF90(50)	120	Energy Environ. Sci., 2022,15, 3462
SCU-19	500	Angew. Chem. Int. Ed. 2019,58,45
COF-4	200	Nat. Commun. 2023, 14,1106
Tp-DBD	300	Small 2021, 17, 2006882
WO_{3-x}	120	Appl. Catal. B. 2023,324,122202
BP@CNF-MOF	600	Adv. Fun. Mater. 2021, 31, 2100106
NDA-TN-AO	900	Angew. Chem. Int. Ed. 2020,59,40

Table S5. A comparison in U uptake capacity from natural seawater among established adsorbents and our case. All the photocatalysis was performed under simulated sunlight.

Materials	Adsorbent dosage	Time (d)	Capacity for U (mg/g/day)	References
TI-COF	**5 mg sample**	**1**	**8.8**	**This work**
i-MZIF90(50)	10 mg sample	25	1.1	Energy Environ. Sci., 2022,15, 3462
COF 4P	10 mg sample	3	8.02	Angew. Chem. Int. Ed. 2023, e202303129
COF-HHTF-AO	5 mg sample	25	0.21	Sci. Bull. 2021, 66, 1994
Tp-DBD	5 mg sample	8	1.28	Small 2021, 17, 2006882
COF 4-Pd-AO	10 mg sample	3	4.62	CCS Chem. 2022, 4, 2294
COF-R5	10 mg sample	15	0.75	Cell Rep. Phys. Sci. 2023, 4, 101220

NDA-TN-AO	5 mg sample	27	0.22	Angew. Chem. Int. Ed. 2020, 59, 17684
COF 2-Ru-AO	10 mg sample	3	2.45	JACS. Au. 2023, 3, 239
COF-4	9 mg sample	56	6.84	Nat. Commun. 2023, 14, 1106
Ui0-66-3C4N	20 mg sample	28	0.24	Angew. Chem. Int. Ed. 2020, 59, 4262
UiO-66-A0	1 mg sample	3	0.89	ACS. Appl. Mater. Interfaces. 2017, 9, 32446
Anti-UiO-66	5 mg sample	30	0.15	Adv. Sci. 2019, 6, 1900002
MIL-101-0A	100 mg sample	5	0.92	Micropor. Mesopor. Mater. 2019, 288, 109567
PPA@MISS-PAF-1			0.38	Chem. 2020, 6, 1683
BP-PAO	10 mg sample	56	0.21	Angew. Chem. Int. Ed. 2019, 59, 1220
AO-PIM-1	10 mg sample	28	0.32	Nat. Sustain. 2022, 5, 71
POP1-AO	2.5 mg sample	56	0.15	ACS Cent. Sci. 2021, 7, 1650
PAF-CS	5 mg sample	21	0.28	Chem. Sci. 2020, 11, 4747
MISS-PAF-1	5 mg sample	56	0.10	ACS Cent. Sci. 2019, 5, 1432
POP-oNH_2- AO	5 mg sample	56	0.07	Nat. Commun. 2018, 9, 1644
P(2DVB-VBC)-2PAN	10 mg sample	27	0.07	Angew. Chem. Int. Ed. 2013, 52, 13458
AO-HNTs	10 mg sample	30	0.30	Angew. Chem. Int. Ed. 2019, 58, 14979
MIGPAF-13	4 mg sample	28	0.28	J. Am. Chem. Soc. 2021, 143, 14523
Fe-Nx-C-R	6 mg sample	1	1.20	Adv. Mater. 2021, 33, 2106621
Cp-1:12	10 mg sample	28	0.02	Sci. China Chem. 2013, 56, 1510
DSUP	10 mg sample	3	5.82	Angew. Chem. Int. Ed. 2020, 59, 15997
SSUP	10 mg sample	3.5	3.52	Angew. Chem. Int. Ed. 2019, 58, 11785
H-ABP	10 mg sample	90	0.13	Energy Environ. Sci. 2019, 12, 1979
SMON-PAO	10 mg sample	56	0.17	Adv. Funct. Mater. 2019, 29, 1805380
Zn^{2+}-PAO	36 mg sample	28	0.33	Adv. Mater. 2020, 32, 1906615
PPH-OP	10 mg sample	21	0.34	Nat. Sustain. 2021, 4, 708
DNA-UEH	10 mg sample	6	1.01	Nat. Commun. 2020, 11, 5708
MS@PIDO/Alg sponge		56	0.10	Adv. Funct. Mater. 2019, 29, 1901009
AUPM membrane	36 mg sample	25	0.35	ACS. Appl. Mater. Interfaces. 2021, 13, 21272