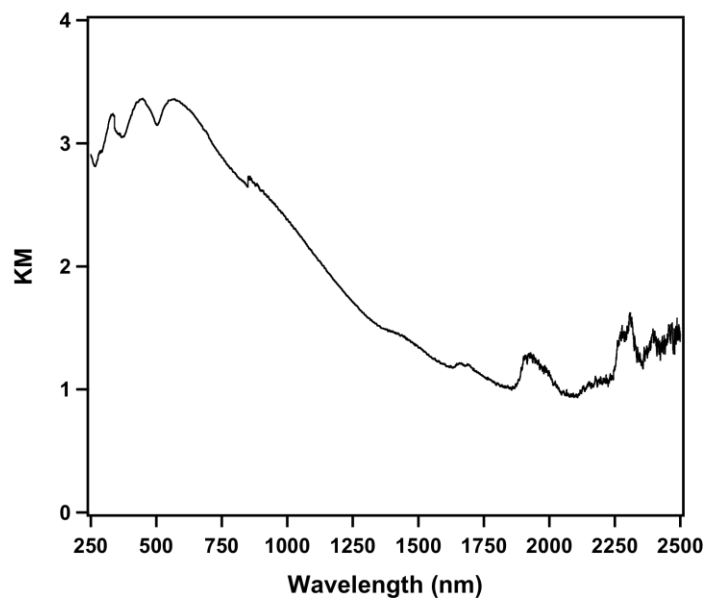
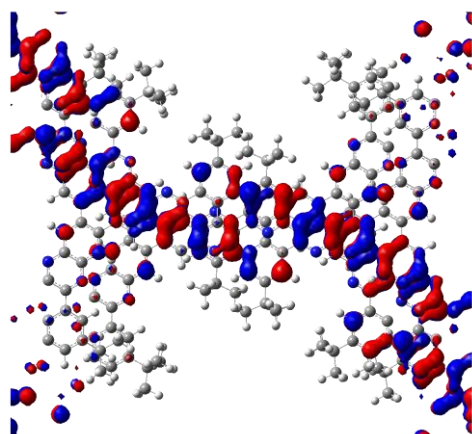


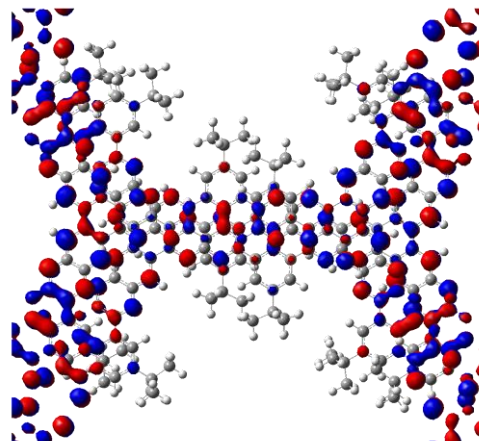
Supplementary Figure S1 | FE-SEM and AFM Images of CS-COF. a, FE SEM image. b, AFM image.



Suppelementary Figure S2 | Electronic absorption spectrum of CS-COF. CS-COF displayed broad absorption profile covering from UV to IR regions.

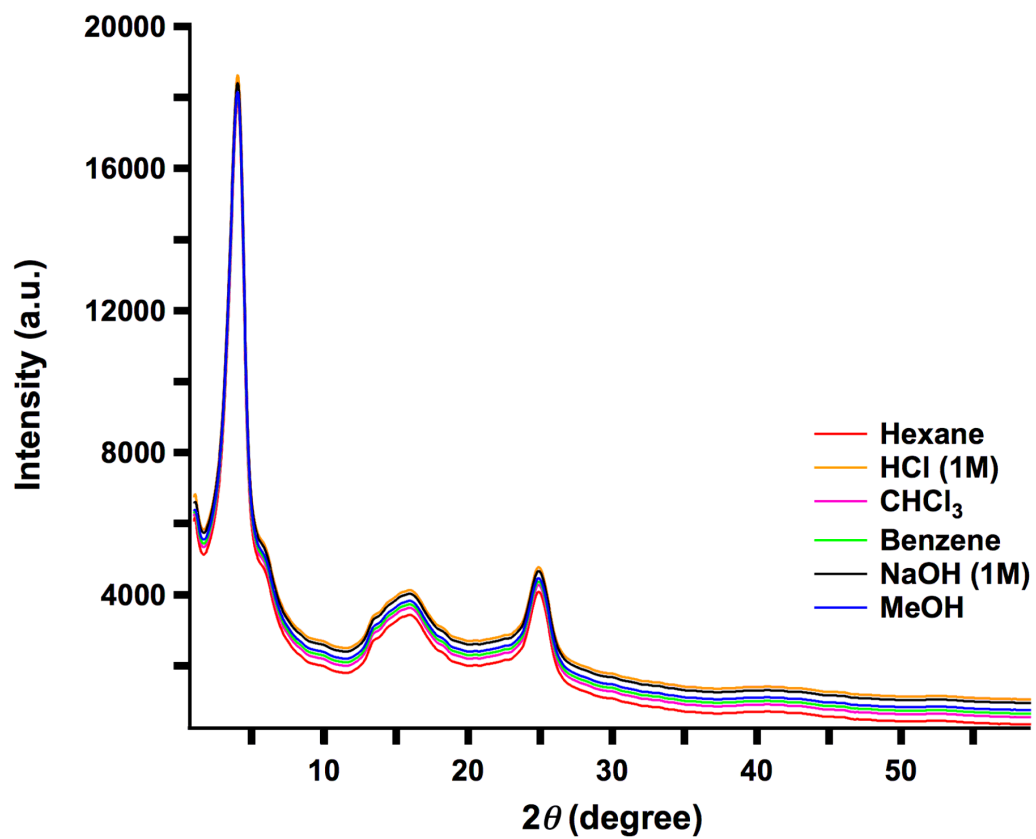


HOMO

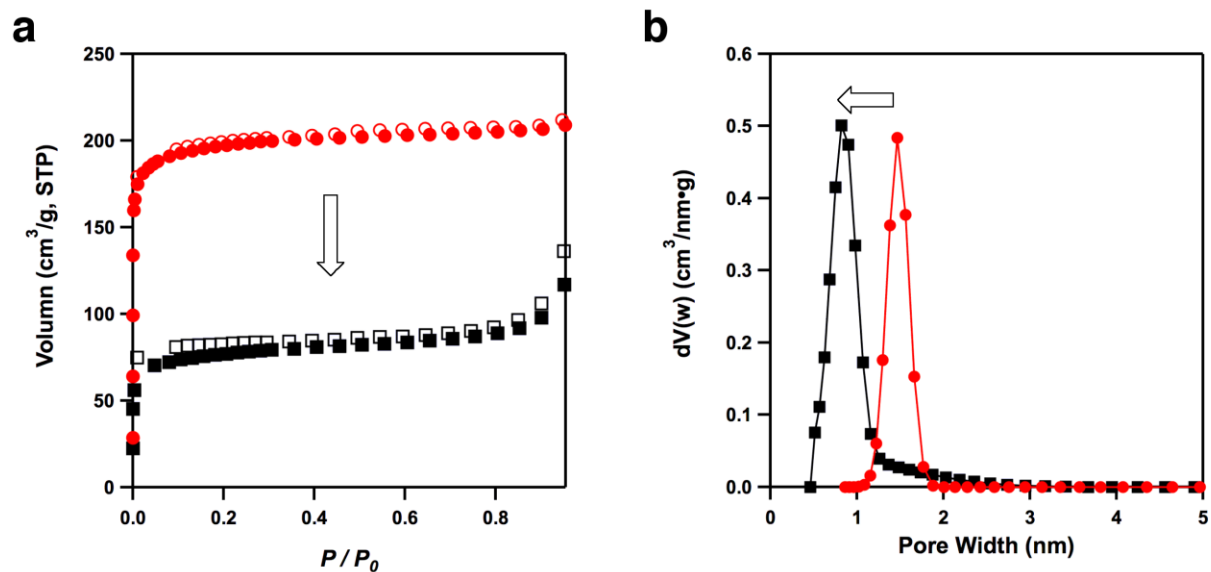


LUMO

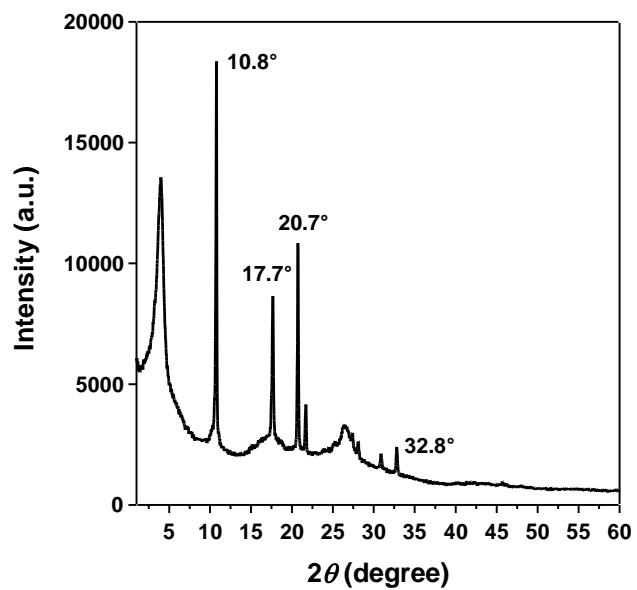
Supplementary Figure S3 | Frontier molecular orbitals (MOs). The π clouds are delocalized in the 2D skeleton. The 0.8-Å slipped AA stacking unit was shown for both HOMO and LUMO.



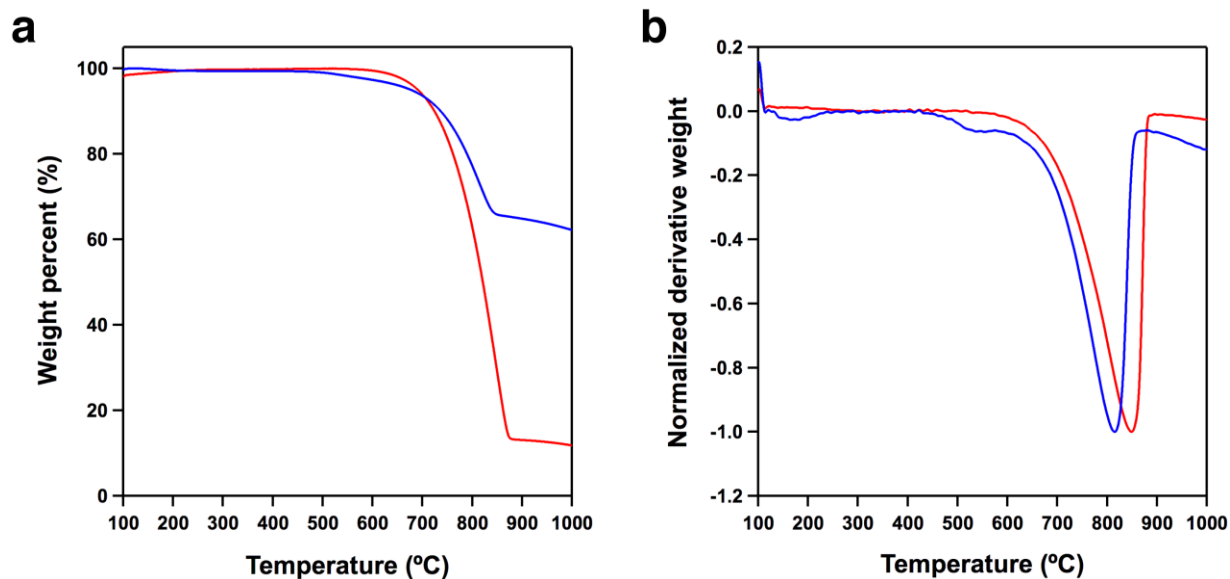
Supplementary Figure S4 | Chemical stability in different solvents. XRD patterns of the CS-COF samples, upon keeping in the solvent for 24 h at 25 °C.



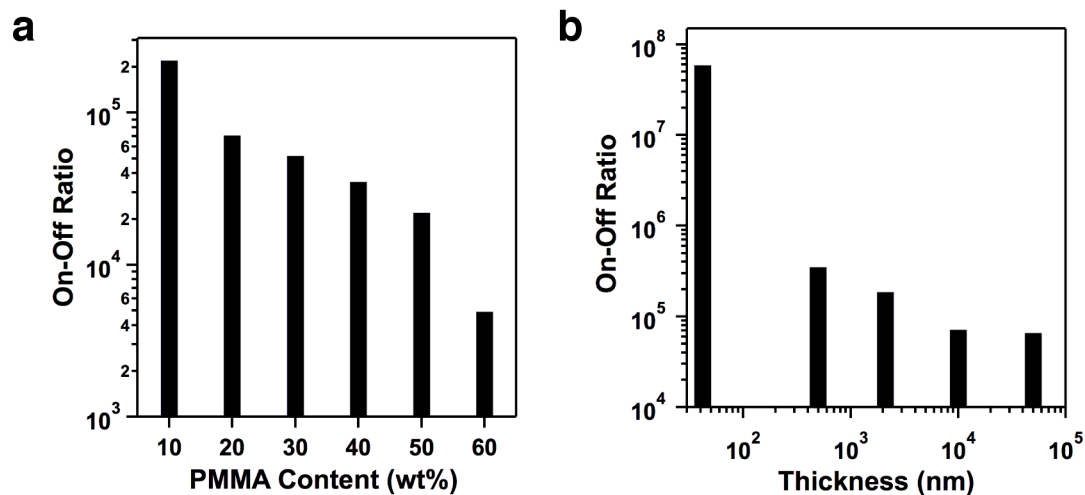
Supplementary Figure S5 | Nitrogen sorption isotherm profiles. **a**, Nitrogen adsorption (filled black square) and desorption (open black square) isotherm profiles of CS-COF \supset C60 (black) at 77 K, in comparison with CS-COF (red). **b**, Pore size of CS-COF \supset C60 (black) calculated using the NLDFT analysis method from the N₂ adsorption isotherm, in comparison with CS-COF (red). Upon loading C60, CS-COF showed significant decreases in specific surface area (776 \rightarrow 291 m² g⁻¹), pore volume (0.34 \rightarrow 0.2 cm³ g⁻¹), and pore size.



Supplementary Figure S6 | XRD pattern of CS-COF@C₆₀. The peaks with values are owing to C₆₀, while other peaks are due to CS-COF. Small amount of C₆₀ aggregates existed between CS-COF particles may also contribute to the XRD peaks.



Supplementary Figure S7 | TGA and DTA profiles. **a**, TGA profile of CS-COF@C₆₀ (blue curve) showed a decrease in weight due to the loss of C₆₀, which is in the similar temperature region to that of C₆₀ (red curve). From the comparison of the two curves, the weight content of C₆₀ in CS-COF@C₆₀ was calculated to be 25 wt%. **b**, DTA curve of CS-COF@C₆₀ (blue curve) showed that the loss of C₆₀ from CS-COF@C₆₀ takes place at 810 °C. The red curve is the DTA profile of C₆₀.



Supplementary Figure S8 | Performance of on-off photoswitches. a, Effect of the PMMA content of CS-COF \rhd C60 film (thickness = 100 μ m) on the photocurrent on-off ratio. **b**, Effect of the thickness of the CS-COF \rhd C60 film (PMMA content = 50 wt%) on the photocurrent on-off ratio.

Supplementary Table S1 | The total DFTB energies, LJ, and the crystal stacking energies per monolayer as well as the corresponding energy gap for CS-COF.

Stacking	c [Å]	Total DFTB Energy [a.u.]	LJ energy [a.u.]	Total crystal stacking energy [kcal mol ⁻¹]	HOMO-LU MO gap [eV]
Monolayer		-243.027702	1.1731		2.046
AA	6.07	-486.147851	2.2503	29.01	2.050
Slipped AA (0.8 Å)	4.01	-486.406628	1.9768	110.20	2.013
AB	4.49	-486.291736	2.1057	74.15	1.977

Suppelementary Table S2 | Atomistic coordinates and net charges for the 0.8-Å slipped AA stacking structure.

atom	x	y	z	population
H	-2.26072953	4.60735469	4.17854197	0.9122687
H	4.46614391	4.54094576	4.17974516	0.91319645
H	4.42140739	0.18259102	4.09171623	0.9134171
H	-2.31222895	0.24661488	4.14964901	0.90963096
H	-1.49092432	-3.57807145	4.19959604	0.90799854
H	3.51558444	-3.63445859	4.27276397	0.91299108
H	-1.40220622	8.37561207	4.34461959	0.90747193
H	3.60340717	8.39288914	4.37520873	0.90726421
H	-2.45219629	-5.21779861	4.43230721	0.90798127
H	0.03677724	-9.54330152	4.02761189	0.90692375
H	4.42183807	-5.28756186	4.65370267	0.91138829
H	1.87005292	-9.57030663	4.1865389	0.90596618
H	-2.3422249	9.99423884	4.28676009	0.90670395
H	0.11690421	14.30403276	4.94149312	0.91146701
H	4.52992152	10.04136906	4.25753754	0.90792347
H	2.0133882	14.32968688	4.82125479	0.91323713
H	8.25830917	11.23465776	4.07764722	0.90904984
H	12.00492877	13.45945976	3.89984499	0.91114568
H	-6.17372919	-6.41600626	4.44039499	0.91250839
H	-9.93474838	-8.60140616	4.50894276	0.91158402
H	-6.1214765	11.15902838	4.11700589	0.90977986
H	-9.87856982	13.36302336	4.09756457	0.90853632
H	8.08627355	-6.51448048	4.5944103	0.90911573
H	11.85875236	-8.69635486	4.68964753	0.91346704
H	6.55592268	0.37201326	4.92854617	0.93351878
H	6.50309296	0.41257481	3.14912801	0.93371103
H	7.9167471	1.02282427	4.01420375	0.93761137
H	6.34202746	4.20371916	2.83346363	0.93260486
H	7.7649435	3.15837219	2.7428746	0.9376714
H	6.2672325	2.68925885	1.91397561	0.93327209

H	7.83920765	3.11961841	5.37272909	0.93742556
H	6.40701634	4.15595414	5.40797025	0.93319912
H	6.39886378	2.60951495	6.27278915	0.93332067
H	-4.45181058	0.53112686	3.27154955	0.93307815
H	-4.38857618	0.51104526	5.05157529	0.93080021
H	-5.79397314	1.18402468	4.21772983	0.93992536
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H	-4.09108064	2.78505582	6.35046649	0.92779602
H	-5.62805066	3.28939783	2.86137841	0.93920668
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H	-4.17009667	2.76417722	1.99276352	0.93052493
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H	11.07060218	-6.22988969	2.35832589	0.93244897
H	12.29600906	-5.19289891	3.11597148	0.93655943

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C	3.94276623	3.57118554	4.16605327	4.09356803
C	2.53708781	3.60406859	4.17802109	4.00771259
C	1.80219864	2.3858348	4.16158755	4.00313815
C	2.51507381	1.15728191	4.1439241	4.00723276
C	3.92342429	1.1632579	4.12015252	4.09648899

C	1.82243091	4.88224614	4.20739931	3.85641772
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C	-0.3931473	-3.62985322	4.23140471	4.13564077
C	2.41606505	-3.66047548	4.27326296	4.13529394
C	1.71904527	-4.87235089	4.31829137	3.978439
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C	1.82130674	7.18704016	4.28266095	3.85272702
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C	2.43840081	-6.15551364	4.3780224	3.98029694
C	-2.60040123	-7.3638601	4.28850882	3.8539121
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C	3.8270562	-6.2050109	4.53306472	4.14029407
C	-1.88616365	-8.59998578	4.15285002	3.85294694
C	2.39682035	-8.60975378	4.26852362	4.1367185
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C	3.80767496	-8.66577258	4.38251713	3.85311858

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C	-0.3702091	10.90797979	4.4952174	3.98325433
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C	2.54750889	10.92538974	4.47738022	3.97843181
C	-2.51334431	12.1244114	4.52655047	3.84932298
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C	3.94300183	10.96160573	4.38703577	4.13348474
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C	4.6678933	12.18117518	4.43986972	3.85121438
C	3.9478069	13.41108945	4.59927412	3.85194331
C	6.65753093	13.34681561	4.3661181	3.85319502
C	-4.58512842	-8.5314128	4.33371146	3.85730823
C	6.50323911	-8.62595534	4.58665301	3.85993738
C	-4.51905045	13.26453602	4.51125861	3.85310913
C	5.93095677	14.59076691	4.50541808	3.8534672
C	-3.79926753	14.50731108	4.69010467	3.85892511
C	-3.86264877	-9.77429553	4.19740994	3.8542244
C	5.77234009	-9.86539139	4.45241904	3.85208495
C	8.11628455	13.36565136	4.24460289	4.01196943
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C	8.84180286	12.1675988	4.09216701	4.09399996
C	10.2163706	14.6119841	4.14926971	4.00790739
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