Supplementary Information (SI) for Journal of Materials Chemistry C.

-- Supplementary Information –

Tuning the charge transfer and band shape of donor-acceptor covalent organic frameworks for optoelectronics

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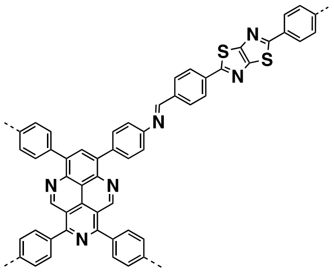
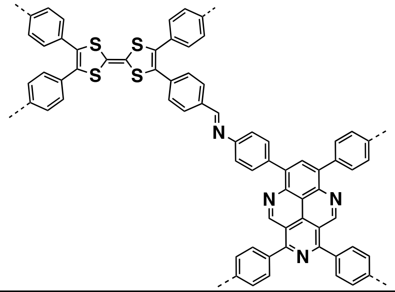
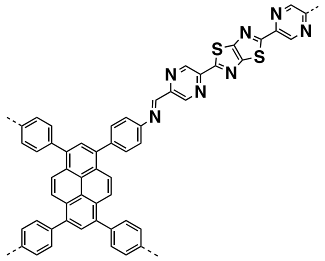
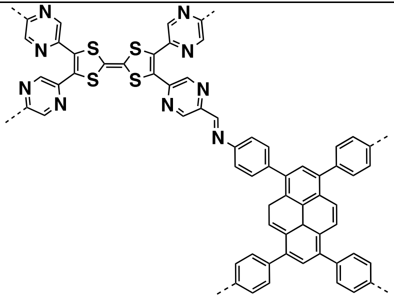
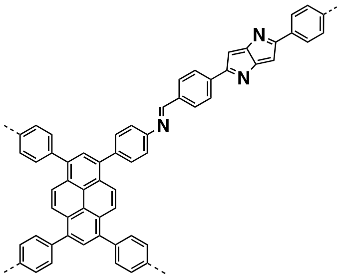
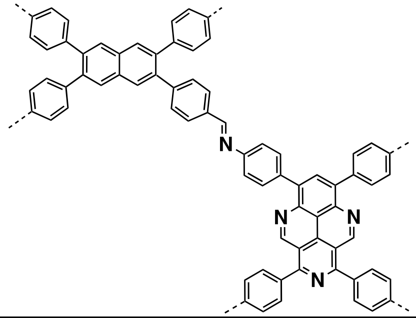
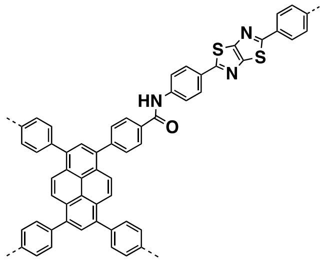
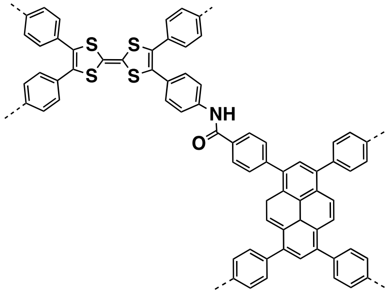
Section 1. Molecular structure of the Mod9-Mod12 monolayers.

Figure S1. Molecular structure of the D-A dimers of COF-1 (left) and COF-2 (right) chemical modifications of the Mod9-to-Mod12.

	COF-1	**COF-2**
Mod9		
Mod10		
Mod11		
Mod12		

Section 2. Optimized cell parameters.

Table S1. Optimized cell parameters of the original (Mod0) and modified (Mod1-Mod12) 2D-COF monolayers of COF-1 and COF-2 given as a, b, c, alpha, beta, gamma in Å and degrees.

	COF-1
Mod0	34.188727 34.079827 25.0 90.0 90.0 77.380512
Mod1	34.319906 34.133621 25.0 90.0 90.0 78.752333
Mod2	34.208113 34.059567 25.0 90.0 90.0 78.455680
Mod3	34.241489 34.095321 25.0 90.0 90.0 78.332226
Mod4	34.288576 34.165730 25.0 90.0 90.0 78.510007
Mod5	34.109156 34.104588 25.0 90.0 90.0 77.341644
Mod6	34.178668 34.065307 25.0 90.0 90.0 76.906174
Mod7	34.181477 34.177451 25.0 90.0 90.0 78.568293
Mod8	34.074860 34.102926 25.0 90.0 90.0 76.955390
Mod9	34.108780 33.997619 25.0 90.0 90.0 78.264896
Mod10	33.877895 33.811213 25.0 90.0 90.0 77.548791
Mod11	33.907578 33.824205 25.0 90.0 90.0 77.309858
Mod12	34.333839 34.300006 25.0 90.0 90.0 74.229264

	COF-2
Mod0	36.181083 20.490657 25.0 89.466041 89.787 55.276740
Mod1	36.025970 20.766860 25.0 89.466534 89.787 55.025096
Mod2	36.018288 21.142236 25.0 89.469066 89.787 53.804915
Mod3	35.976360 21.005028 25.0 89.468311 89.787 54.156663
Mod4	36.050862 21.018469 25.0 89.467948 89.787 54.329322
Mod5	35.973405 20.548020 25.0 89.466768 89.787 54.907234
Mod6	36.060541 20.884280 25.0 89.467428 89.787 54.580560
Mod7	35.965774 21.042067 25.0 89.468441 89.787 54.095513
Mod8	35.985578 21.259343 25.0 89.469079 89.787 53.799158
Mod9	35.859184 20.876537 25.0 89.467491 89.787 54.549919
Mod10	35.552549 21.204959 25.0 89.470178 89.787 53.302931
Mod11	34.852249 20.398075 25.0 89.467819 89.787 54.391247
Mod12	36.282791 20.682527 25.0 89.465974 89.787 55.311626

Section 3. HOCO, LUCO, Band gap and S_1 energies of the 26 monolayers.

Table S2. HOCO, LUCO, Band Gap, S1 energies (all in eV) and oscillator strengths at PBE0 level are collected for the optimized COF-1 and COF-2 monolayers obtained with PBE-D3BJ.

	COF-1					**COF-2**				
	HOCO	**LUCO**	**BG**	**S1**	**o. s.**	**HOCO**	**LUCO**	**BG**	**S1**	**o. s.**
Mod0	-5.09	-2.61	2.48	2.25	1.25	-4.47	-2.12	2.36	1.95	0.08
Mod1	-5.29	-3.26	2.03	1.94	0.23	-4.49	-2.14	2.35	2.00	0.13
Mod2	-5.91	-3.17	2.75	2.39	1.47	-4.20	-2.05	2.16	1.91	0.09
Mod3	-5.37	-2.26	3.11	2.65	1.16	-4.44	-2.32	2.12	1.79	0.09
Mod4	-5.78	-2.55	3.23	2.77	1.13	-4.39	-1.96	2.43	2.32	0.05
Mod5	-5.64	-2.70	2.94	2.51	1.35	-4.70	-2.84	1.86	1.43	0.04
Mod6	-5.20	-2.57	2.63	2.40	1.34	-4.53	-2.83	1.71	1.69	0.03
Mod7	-4.91	-1.88	3.04	2.64	1.07	-4.48	-2.17	2.31	1.95	0.12
Mod8	-5.35	-2.49	2.85	2.54	1.54	-5.04	-2.74	2.30	1.90	0.11
Mod9	-5.39	-2.73	2.67	2.23	1.49	-4.54	-2.42	2.12	1.95	0.12
Mod10	-5.26	-3.13	2.13	1.98	0.90	-4.86	-2.81	2.05	1.67	0.13
Mod11	-5.18	-3.46	1.72	1.66	0.38	-5.12	-2.29	2.83	2.43	1.36
Mod12	-5.42	-2.48	2.94	2.67	1.16	-4.07	-2.11	1.96	1.94	0.02

Section 4. Characterization of the orbital contributions to the S_1 state.

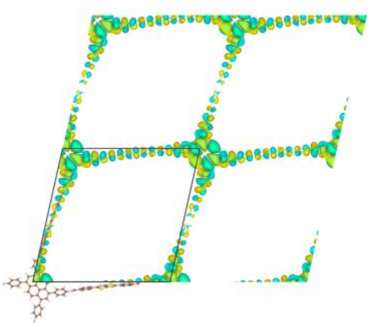
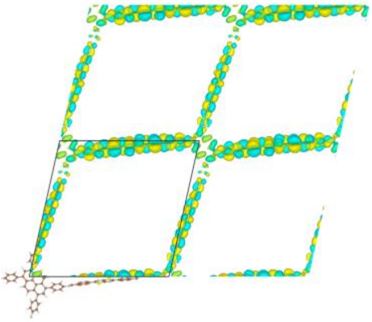
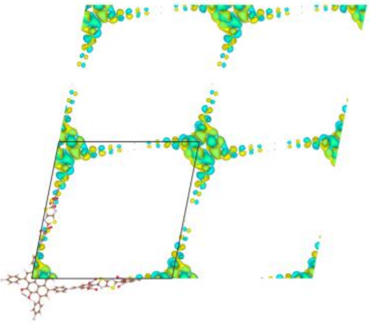
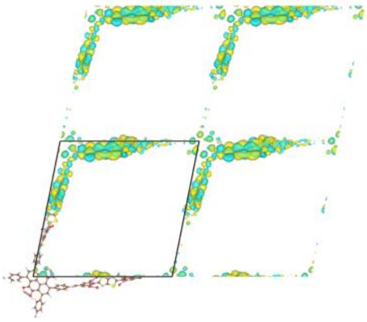
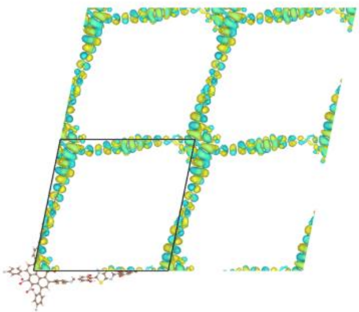
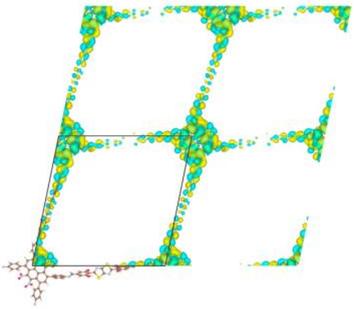
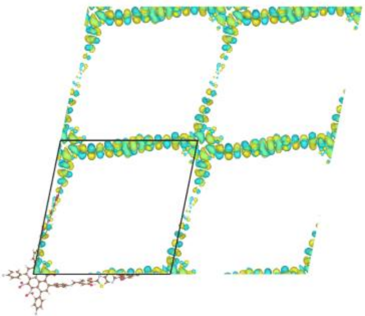
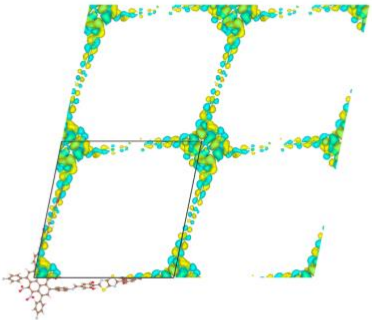
Table S3. Occupied-virtual orbital main contributions (with coefficient > 0.2) to the lowest excited state singlet (S1) as computed with TD-DFPT implemented in CP2K with truncated PBE0 functional for the COF-1 monolayers. The overlap and CT number (1-Λ) associated with each contribution is given. The energy difference between the HOCO and HOCO-1, and the LUCO and LUCO+1 is provided in eV.

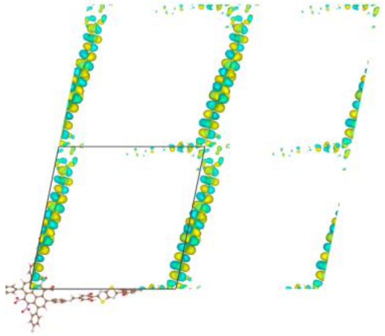
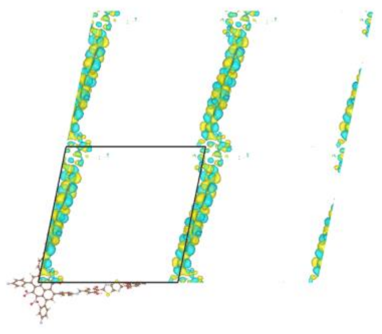
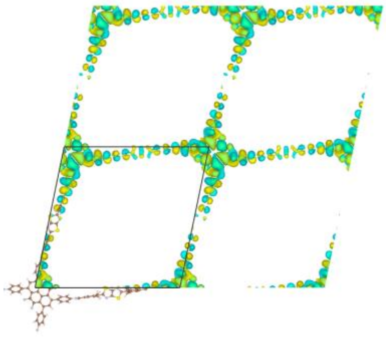
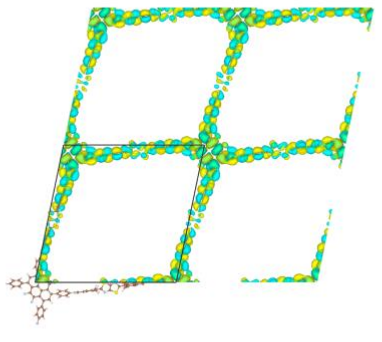
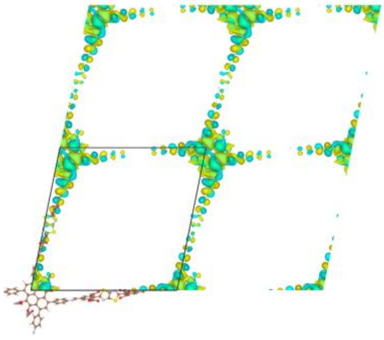
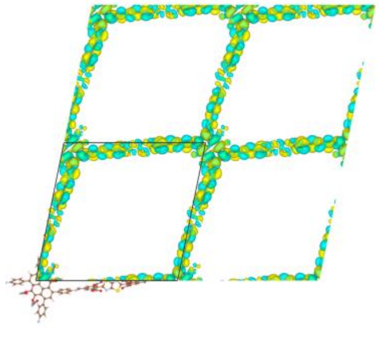
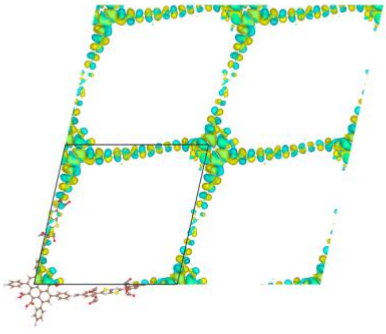
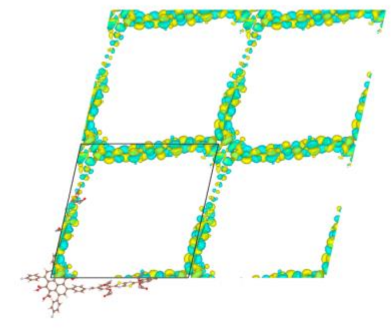
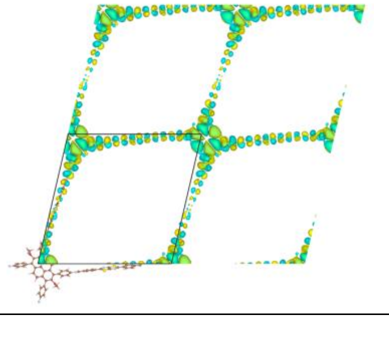
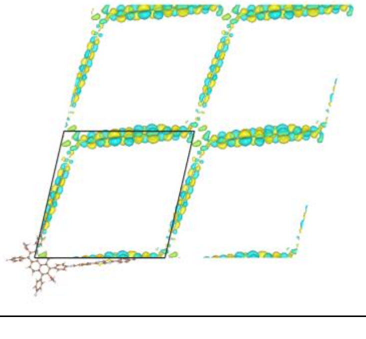
COF-1	**HOCO**	**Orbitals**	**coefficient**	**overlap**	**1-Λ**	**HOCO-HOCO-1**	**LUCO+1-LUCO**
Mod0	205	**205 206**	**0.817108**	**0.3976**	0.6024	0.58	0.01
		203 206	0.309677	0.7629	0.2371		
		205 208	0.232210	0.6160	0.3840		
		205 207	0.213095	0.3061	0.6939		
		204 207	0.200053	0.7629	0.2371		
Mod1	281	**281 283**	**-0.970332**	**0.2271**	0.7729	0.87	0.00
Mod2	261	**259 262**	**-0.589776**	**0.6247**	0.3753	0.10	0.46
		260 262	**-0.525749**	**0.4632**	0.5368		
		261 263	**-0.516994**	**0.7566**	0.2434		
Mod3	241	**241 242**	**0.901355**	**0.8231**	0.1769	0.57	0.35
Mod4	301	**301 302**	**0.792492**	**0.7020**	0.2980	0.15	0.15
		300 305	0.259007	0.4084	0.5916		
		301 306	-0.258275	0.7355	0.2645		
Mod5	301	**301 302**	**0.686099**	**0.6067**	0.3933	0.39	0.06
		301 304	0.389841	0.7037	0.2963		
		299 303	-0.310574	0.7091	0.2909		
		300 302	-0.225452	0.6165	0.3835		
Mod6	217	**217 219**	**0.704793**	**0.3609**	0.6391	0.46	0.02
		215 219	0.388294	0.7192	0.2808		
		216 218	-0.321057	0.7478	0.2522		
		217 220	0.280295	0.6044	0.3956		
Mod7	301	**301 302**	**0.905602**	**0.7405**	0.2595	0.89	0.22
Mod8	241	**239 242**	**0.672703**	**0.7714**	0.2286	0.21	0.08
		240 243	0.418502	0.7370	0.2630		
		241 243	0.376339	0.3629	0.6371		
		241 244	-0.226791	0.4696	0.5304		
		241 246	-0.207065	0.8164	0.1836		
Mod9	205	**205 206**	**0.795703**	**0.6636**	0.3364	0.31	0.09
		205 208	0.274005	0.7627	0.2373		
		204 207	-0.255027	0.7300	0.2700		
		203 208	0.207529	0.7046	0.2954		
Mod10	205	**205 207**	**-0.847040**	**0.3316**	0.6684	0.76	0.01
		205 206	0.393103	0.3047	0.6953		
		203 207	0.216480	0.6865	0.3135		
Mod11	203	**203 205**	**-0.959019**	**0.2192**	0.7808	0.69	0.00
		203 204	-0.231472	0.2407	0.7593		
Mod12	217	**215 218**	**0.591903**	**0.8419**	0.1581	0.01	0.42
		217 218	**0.569320**	**0.3412**	0.6588		
		217 220	-0.346323	0.7293	0.2707		
		216 219	-0.328975	0.7289	0.2711		

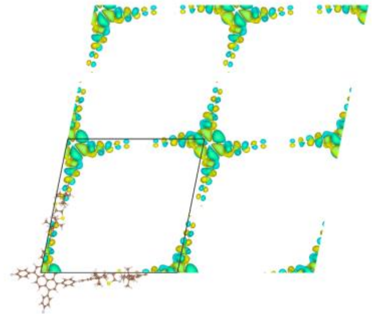
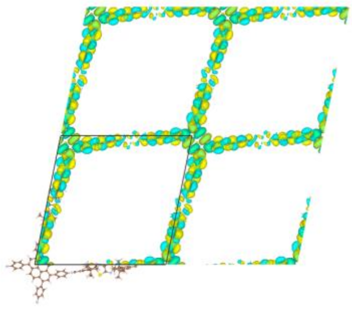
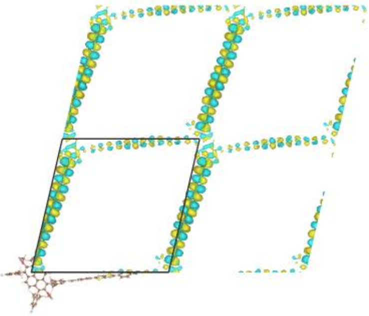
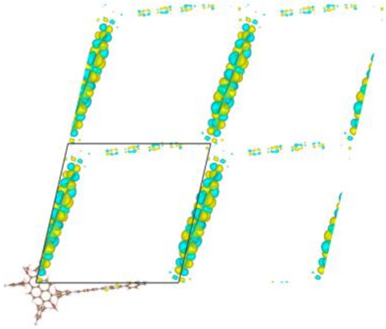
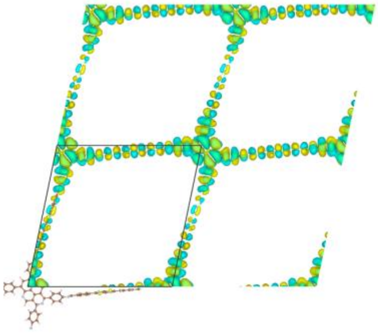
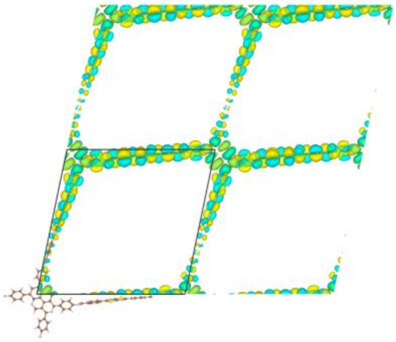
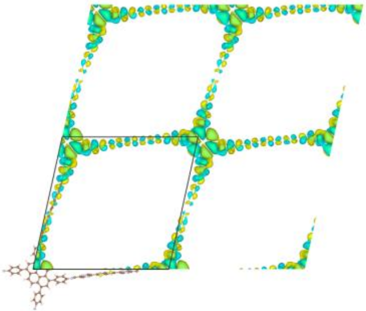
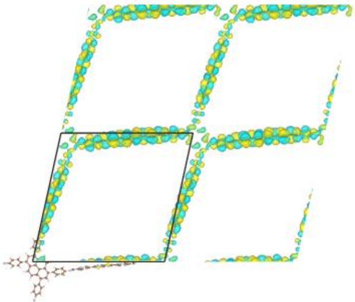
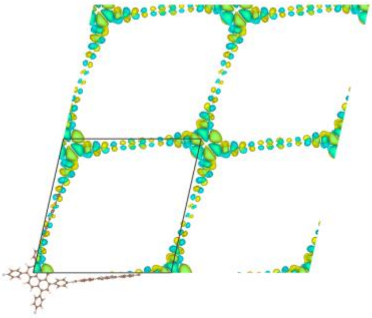
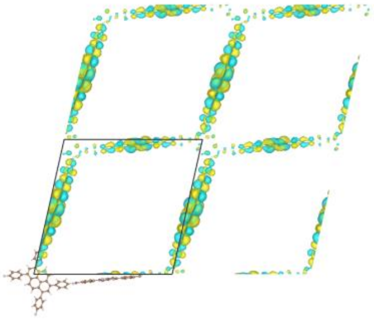
Table S4. Occupied-virtual orbital main contributions (with coefficient > 0.2) to the lowest excited state singlet (S1) as computed with TD-DFPT implemented in CP2K with truncated PBE0 functional for the COF-2 monolayers. The overlap and CT number (1-Λ) associated with each contribution is given. The energy difference between the HOCO and HOCO-1, and the LUCO and LUCO+1 is provided in eV.

	HOCO	**Orbitals**	**coefficient**	**overlap**	**1-Λ**	**HOCO-HOCO-1**	**LUCO+1-LUCO**
Mod0	191	**191 192**	**0.907205**	**0.3798**	0.6202	0.35	0.12
		191 193	-0.325119	0.3577	0.6423		
Mod1	203	**203 204**	**0.946137**	**0.3781**	0.6219	0.12	0.21
Mod2	215	**215 216**	**0.959384**	**0.3105**	0.6895	0.65	0.37
Mod3	227	**227 228**	**0.962029**	**0.3797**	0.6203	0.38	0.15
Mod4	227	**227 228**	**0.862089**	**0.1302**	0.8698	0.65	0.39
		227 230	-0.411954	0.3236	0.6764		
Mod5	255	**255 256**	**0.731396**	**0.3689**	0.6311	0.46	0.02
		255 257	**0.640217**	**0.3799**	0.6201		
Mod6	223	**223 224**	**0.992261**	**0.0955**	0.9045	1.15	0.65
Mod7	239	**239 240**	**0.931339**	**0.3793**	0.6207	0.52	0.25
Mod8	239	**239 240**	**0.954064**	**0.4245**	0.5755	0.30	0.25
Mod9	191	**191 192**	**0.906080**	**0.2352**	0.7648	0.65	0.46
		191 193	0.335835	0.4636	0.5364		
Mod10	191	**191 192**	**0.961880**	**0.4860**	0.5140	0.22	0.12
Mod11	189	**189 190**	**0.920132**	**0.7515**	0.2485	0.55	0.41
Mod12	203	**203 204**	**0.995369**	**0.0798**	0.9202	1.28	0.78

Figure S2. Occupied and virtual crystal orbitals involved in the major contribution(s) to the S1 state of all COF-1 monolayers as computed with TD-DFPT implemented in CP2K with truncated PBE0 functional.

		Occupied		**Virtual**
Mod0	205		206	
Mod1	281		283	
Mod2	259		262	
	260		262	

	261		263	
Mod3	241		242	
Mod4	301		302	
Mod5	301		302	
Mod6	217		219	

Mod7	301		302	
Mod8	239		242	
Mod9	205		206	
Mod10	205		207	
Mod11	203		205	

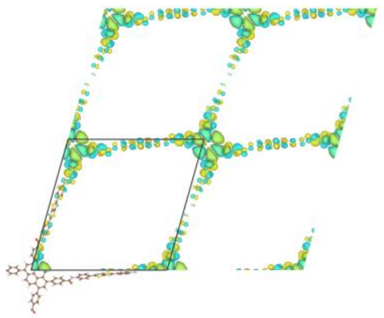
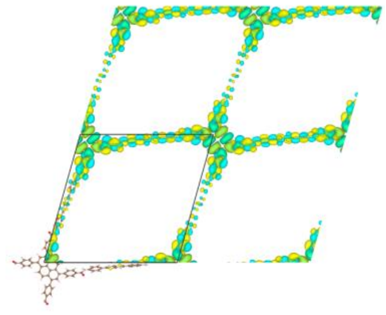
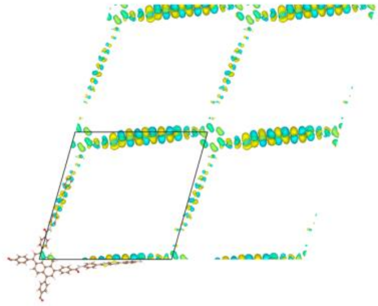
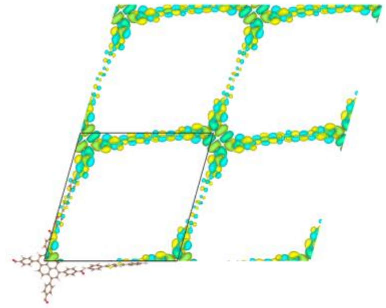
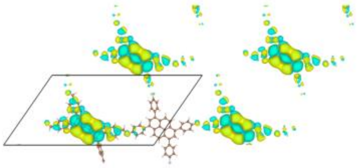
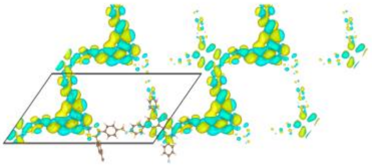
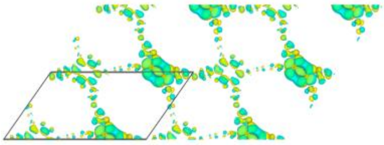
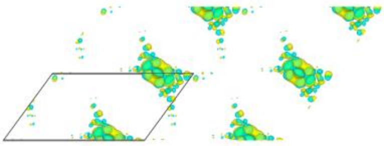
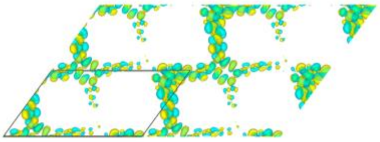
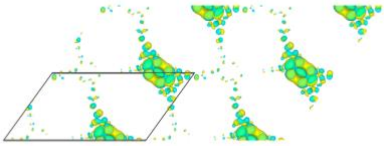
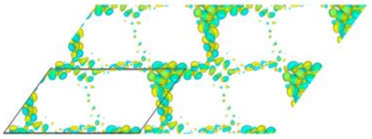
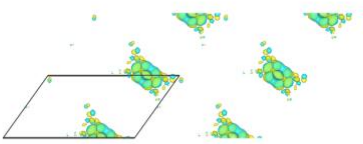
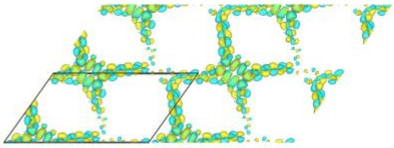
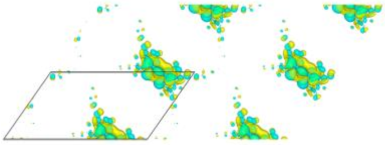
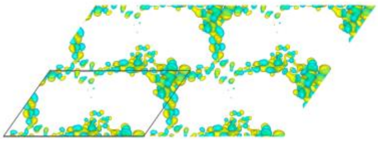
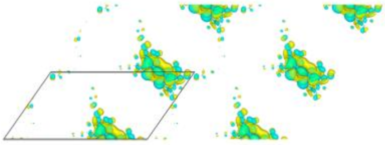
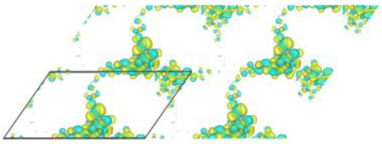
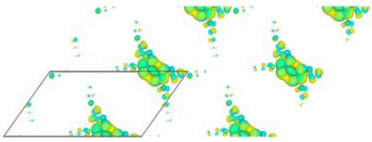
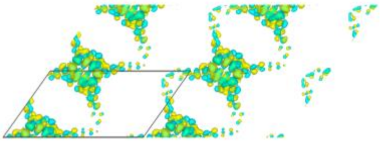
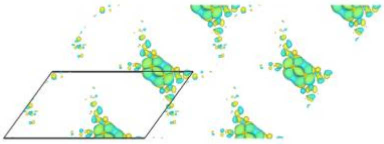
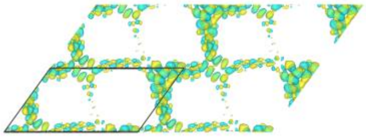
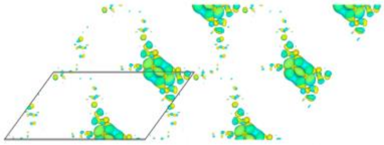
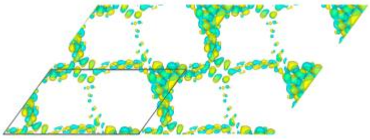
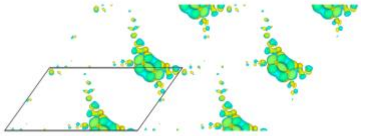
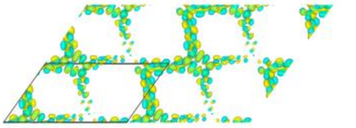
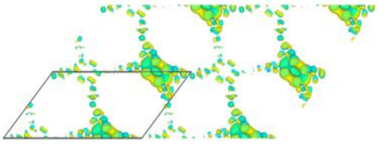
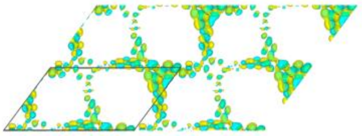
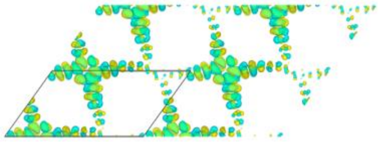
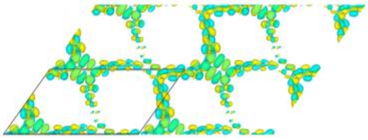
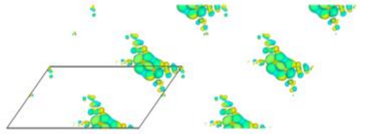
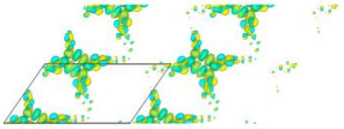
Mod12	215		218	
	217		218	

Figure S3. Occupied and virtual crystal orbitals involved in the major contribution(s) to the S1 state of all COF-2 monolayers as computed with TD-DFPT implemented in CP2K with truncated PBE0 functional.

		Occupied		**Virtual**
Mod0	191		192	
Mod1	203		204	
Mod2	215		216	
Mod3	227		228	
Mod4	227		228	
Mod5	255		256	
	255		257	
Mod6	223		224	
Mod7	239		240	
Mod8	239		240	

Mod9	191		192	
Mod10	191		192	
Mod11	189		190	
Mod12	203		204	

Section 5. HOMO and LUMO energies of the constituent D and A monomers.

Table S5. PBE0/def2svp HOMO and LUMO orbital energies of the Py and Tz isolated monomers extracted from Mod0-to-Mod12 COF-1 monolayers. The local LUMO-HOMO energy differences evaluated as $L_{Py} - H_{Py}$, and $L_{Tz} - H_{Tz}$ are also provided. The CT LUMO-HOMO energy differences evaluated as $L_{Py} - H_{Tz}$ and $L_{Tz} - H_{Py}$ are also provided. Finally, the difference between the smallest local and smallest CT orbital gap is provided.

	H_{Py}	L_{Py}	$L_{Py} - H_{Py}$	H_{Tz}	L_{Tz}	$L_{Tz} - H_{Tz}$	$L_{Tz} - H_{Py}$	$L_{Py} - H_{Tz}$	local – CT
Mod0	-5.46	-2.16	3.30	-6.32	-2.81	3.51	2.65	4.16	0.65
Mod1	-5.13	-2.15	2.98	-7.13	-3.48	3.65	1.65	4.98	1.33
Mod2	-6.52	-3.38	3.14	-6.61	-2.43	4.17	4.08	3.23	-0.09
Mod3	-5.81	-2.48	3.33	-6.71	-2.14	4.57	3.67	4.23	-0.34
Mod4	-6.02	-2.51	3.51	-6.64	-2.55	4.09	3.47	4.13	0.04
Mod5	-5.97	-2.62	3.35	-6.67	-2.85	3.83	3.12	4.05	0.23
Mod6	-5.56	-2.07	3.49	-6.32	-2.80	3.52	2.75	4.25	0.73
Mod7	-5.46	-2.13	3.33	-6.85	-1.85	5.01	3.61	4.72	-0.28
Mod8	-5.73	-1.98	3.75	-6.34	-2.78	3.55	2.95	4.36	0.61
Mod9	-5.80	-2.74	3.06	-6.32	-2.81	3.51	2.99	3.58	0.07
Mod10	-5.46	-2.16	3.29	-6.78	-3.37	3.41	2.08	4.62	1.21
Mod11	-5.46	-2.16	3.29	-6.35	-3.75	2.60	1.70	4.19	0.90
Mod12	-5.85	-2.43	3.42	-5.79	-2.15	3.64	3.70	3.37	0.06

Table S6. PBE0/def2svp HOMO and LUMO orbital energies of the TTF and Py isolated monomers extracted from Mod0-to-Mod12 COF-2 monolayers. The local LUMO-HOMO energy differences evaluated as L TFF – H TTF, and L Py – H Py are also provided. The CT LUMO-HOMO energy differences evaluated as L Py – H TTF and L TTF – H Py are also provided. Finally, the difference between the smallest local and smallest CT orbital gap is provided.

	H_{TTF}	L_{TTF}	$L_{TTF} - H_{TTF}$	H_{Py}	L_{Py}	$L_{Py} - H_{Py}$	$L_{Py} - H_{TTF}$	$L_{TTF} - H_{Py}$	local – CT
Mod0	-5.01	-2.35	2.66	-5.38	-2.13	3.25	2.88	3.03	-0.22
Mod1	-5.22	-2.43	2.79	-5.08	-2.06	3.02	3.16	2.65	0.14
Mod2	-4.88	-2.26	2.62	-5.41	-2.14	3.28	2.74	3.15	-0.12
Mod3	-5.23	-2.78	2.45	-5.24	-2.06	3.19	3.17	2.47	-0.02
Mod4	-5.15	-1.92	3.23	-5.70	-2.44	3.26	2.69	3.79	0.53
Mod5	-5.59	-3.52	2.07	-5.41	-2.13	3.28	3.46	1.90	0.18
Mod6	-5.23	-2.45	2.78	-6.46	-3.32	3.14	1.91	4.01	0.87
Mod7	-5.00	-2.44	2.57	-5.43	-2.19	3.24	2.82	2.99	-0.25
Mod8	-5.77	-3.10	2.68	-5.93	-2.68	3.25	3.09	2.83	-0.15
Mod9	-5.22	-2.37	2.85	-5.77	-2.73	3.04	2.49	3.40	0.36
Mod10	-5.64	-3.32	2.32	-5.43	-2.14	3.29	3.49	2.12	0.20
Mod11	-6.52	-2.21	4.31	-5.74	-2.71	3.03	3.81	3.53	-0.50
Mod12	-4.64	-1.51	3.13	-5.79	-2.37	3.42	2.27	4.29	0.86

Section 6. CT values of the isolated D-A pairs at PBE0 and ωB97XD level.

Table S7. CT values of COF-1 evaluated at (a) the 2D monolayers with PBE0, (b) the isolated D-A pairs in gas phase at the 2D geometry with PBE0, (c) the isolated D-A pairs in gas phase at the 2D geometry with ωB97XD, (d) the isolated D-A pairs in gas phase at the ωB97XD optimized ground state geometry with ωB97XD, and (e) the isolated D-A pairs in gas phase at the ωB97XD optimized excited state geometry with ωB97XD.

Column	**(a)**	**(b)**	**(c)**	**(d)**	**(e)**
Environment:	2D	Gas phase (GP)			
DFT functional:	PBE0		wB97XD		
Geometry:	2D (GS)			GP(GS)	GP(ES)
Mod0	0.53	0.74	0.30	0.28	0.79
Mod1	0.77	0.82	0.71	0.61	0.59
Mod2	0.39	0.64	0.42	0.26	0.37
Mod3	0.21	0.18	0.17	0.17	0.15
Mod4	0.35	0.48	0.29	0.26	0.18
Mod5	0.36	0.50	0.35	0.23	0.28
Mod6	0.49	0.75	0.33	0.41	0.79
Mod7	0.27	0.25	0.22	0.22	0.17
Mod8	0.33	0.78	0.27	0.49	0.60
Mod9	0.34	0.50	0.37	0.39	0.27
Mod10	0.64	0.83	0.47	0.44	0.80
Mod11	0.77	0.87	0.38	0.47	*
Mod12	0.37	0.63	0.32	0.21	0.14

*the ES could not be optimized

Table S8. CT values of COF-2 evaluated at (a) the 2D monolayers with PBE0, (b) the isolated D-A pairs in gas phase at the 2D geometry with PBE0, (c) the isolated D-A pairs in gas phase at the 2D geometry with ωB97XD, (d) the isolated D-A pairs in gas phase at the ωB97XD optimized ground state geometry with ωB97XD, and (e) the isolated D-A pairs in gas phase at the ωB97XD optimized excited state geometry with ωB97XD.

	(a)	**(b)**	**(c)**	**(d)**	**(e)**
Environment:	2D	Gas phase (GP)			
DFT functional:	PBE0		wB97XD		
Geometry:	2D (GS)			GP(GS)	GP(ES)
Mod0	0,62	0,58	0,57	0,57	0,53
Mod1	0,62	0,52	0,55	0,54	0,57
Mod2	0,69	0,68	0,70	0,53	0,60
Mod3	0,62	0,59	0,57	0,59	0,57
Mod4	0,81	0,84	0,53	0,60	0,56
Mod5	0,63	0,68	0,69	0,65	0,86
Mod6	0,90	0,96	0,66	0,55	0,55
Mod7	0,62	0,65	0,62	0,54	0,57
Mod8	0,58	0,55	0,52	0,52	0,53
Mod9	0,73	0,75	0,57	0,54	0,53
Mod10	0,52	0,58	0,54	0,52	0,58
Mod11	0,27	0,22	0,22	0,22	0,19
Mod12	0,92	0,96	0,62	0,61	0,58

Section 7. Orbital contributions to the optimized S$_1$ state of the Mod0, Mod6 and Mod10 of COF-1 D-A dimers.

Figure S4. Major orbital contributions to the S$_1$ state of the D-A pair of Mod0 of COF-1 at the ground state geometry (GS opt) and at the optimized S$_1$ excited state geometry (ES opt).

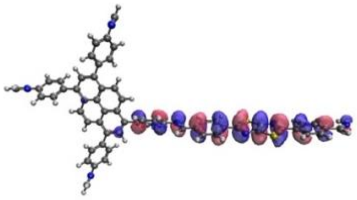
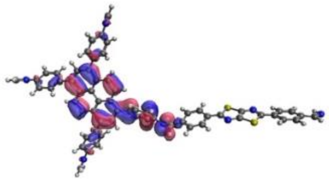
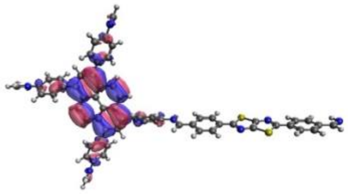
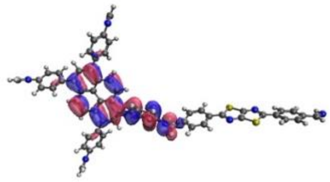
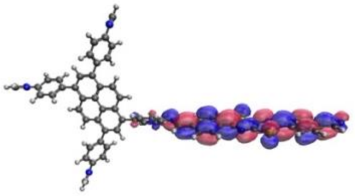
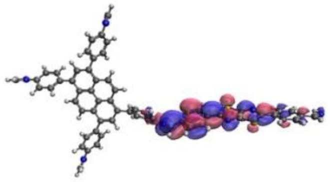
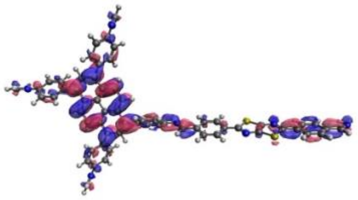
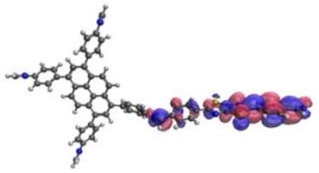
Mod0		
Geometry	**GS opt**	**ES opt**
Major contributions to S1	**242→ 244 0.48** **243 → 245 -0.37**	**243 → 244 0.48** **242 → 244 0.36**
MO 242		
MO 243		
MO 244		
MO 245		

Figure S5. Major orbital contributions to the S_1 state of the D-A pair of Mod6 of COF-1 at the ground state geometry (GS opt) and at the optimized S_1 excited state geometry (ES opt).

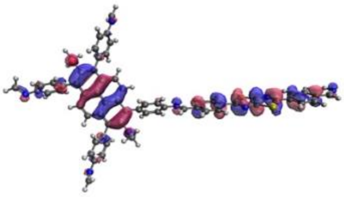
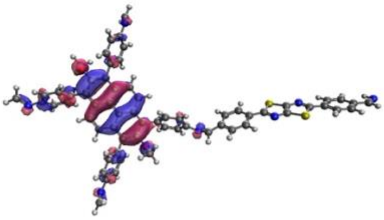
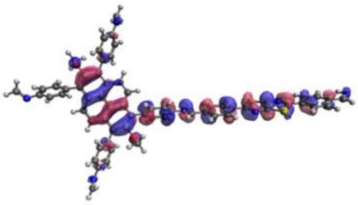
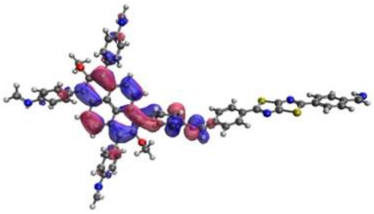
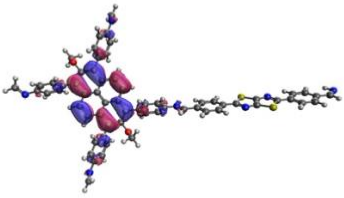
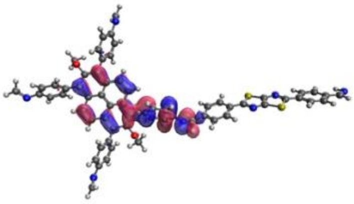
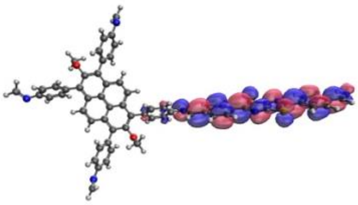
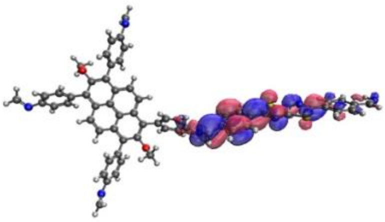
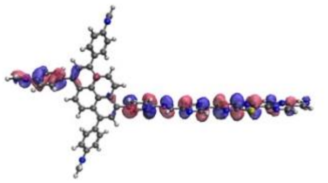
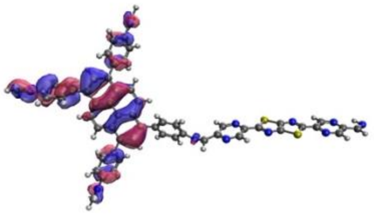
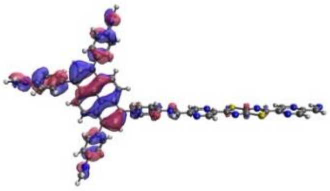
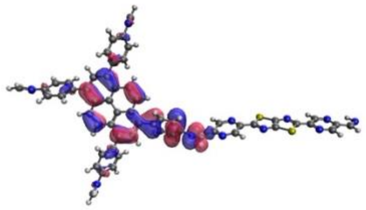
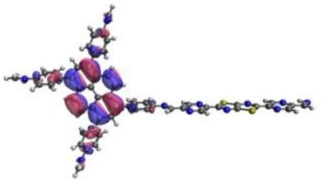
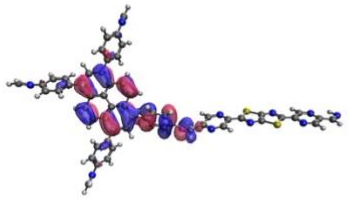
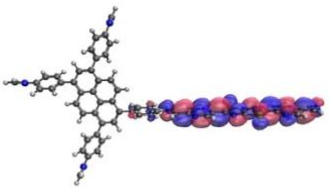
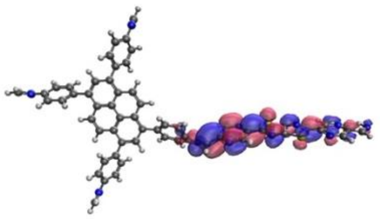
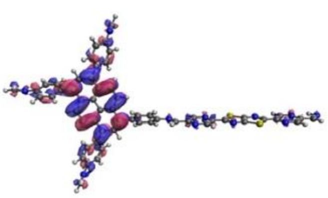
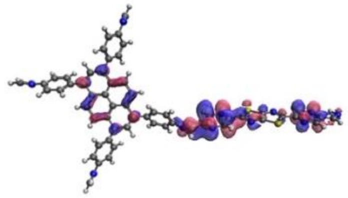
Mod6		
Geometry	**GS opt**	**ES opt**
Major contributions to S1	**258→ 260 0.42** **257 → 260 0.34**	**259 → 260 -0.52** **258 → 260 0.30**
MO 257		
MO 258		
MO 259		
MO 260		

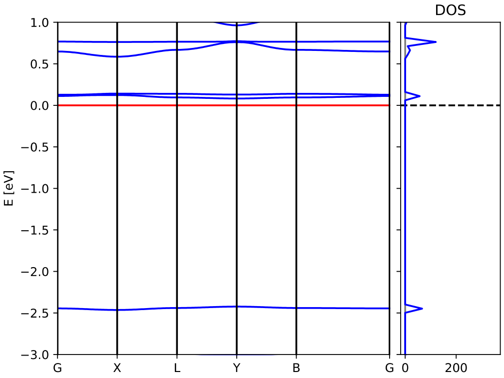
Figure S6. Major orbital contributions to the S_1 state of the D-A pair of Mod10 of COF-1 at the ground state geometry (GS opt) and at the optimized S_1 excited state geometry (ES opt).

Mod10		
Geometry	**GS opt**	**ES opt**
Major contributions to S1	**241→ 244 0.44** **243 → 246 0.25**	**243 → 244 -0.43** **242 → 244 0.41**
MO 241		
MO 242		
MO 243		
MO 244		
MO 246		

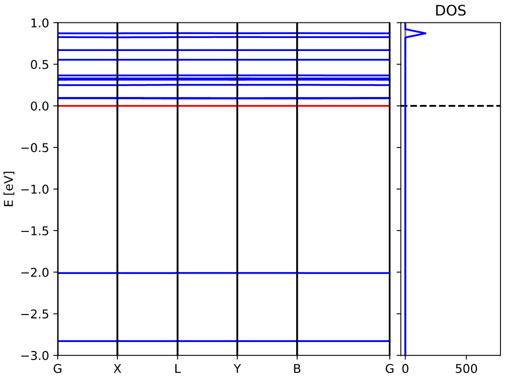
Section 8. Band structure of the 26 monolayers.

Figure S7. Band structures of COF-1 monolayers computed at PBE0 level.

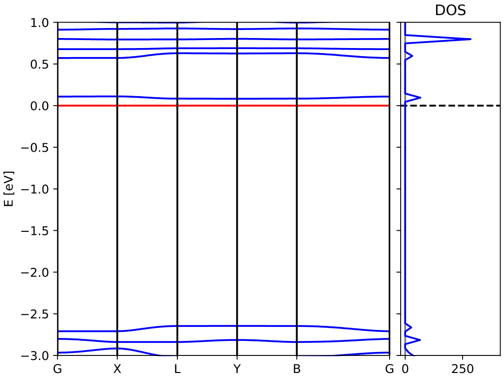
Mod0



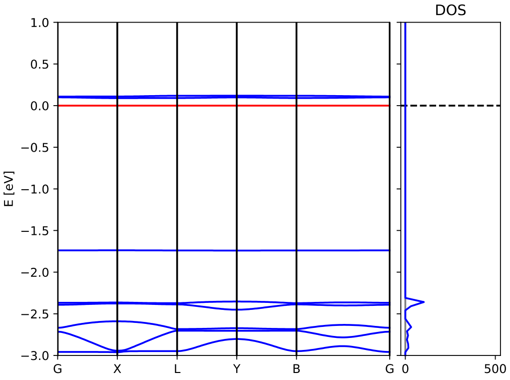
Mod1



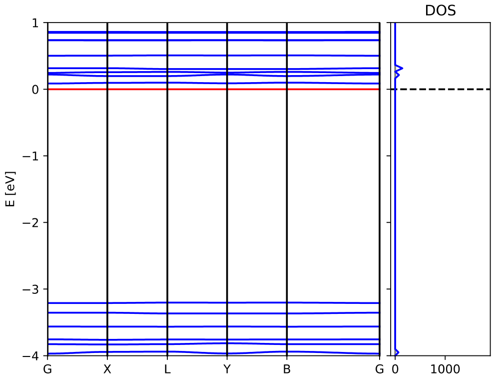
Mod2



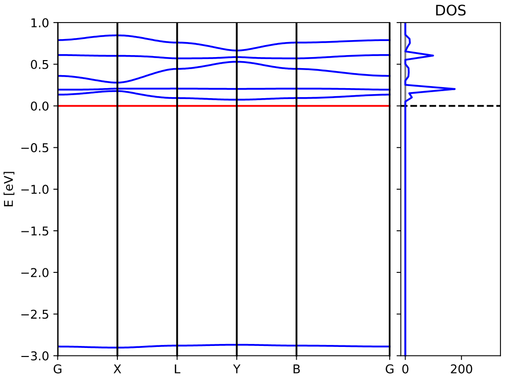
Mod3



Mod4

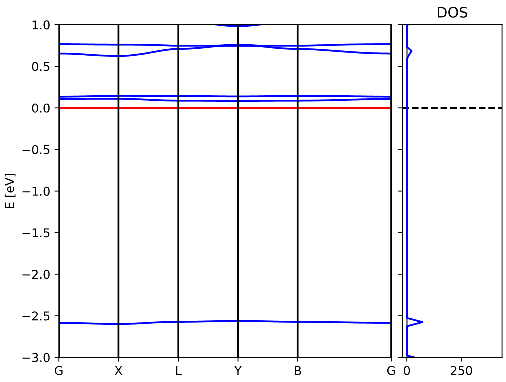


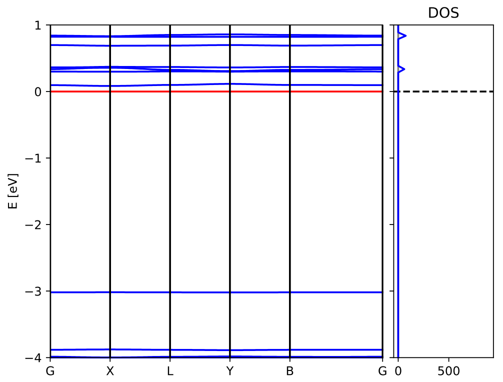
Mod5



Mod6

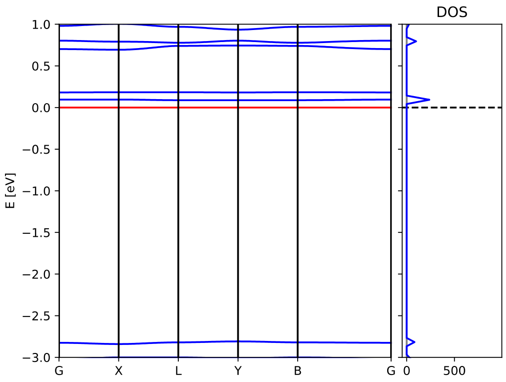
Mod7

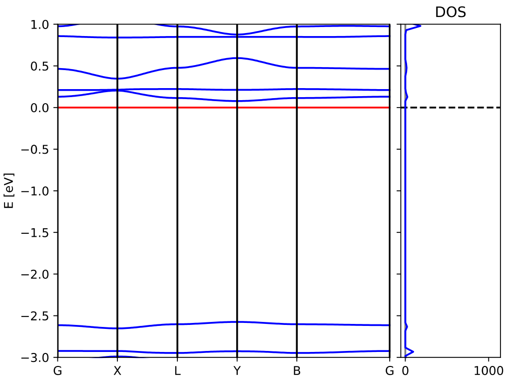




Mod8

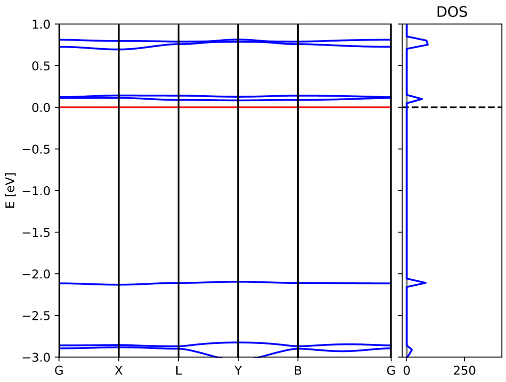
Mod9

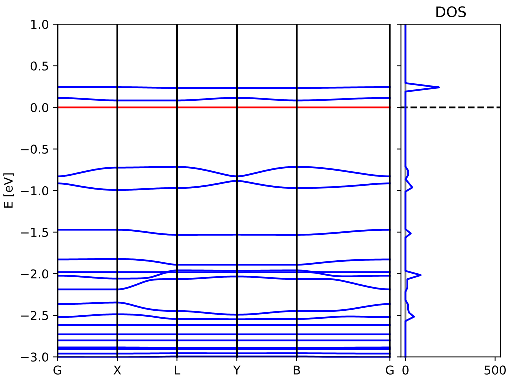




Mod10

Mod11





Mod12

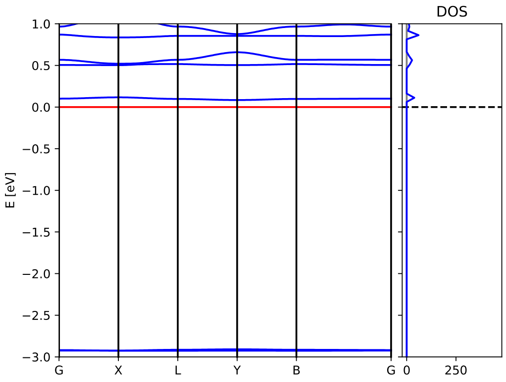
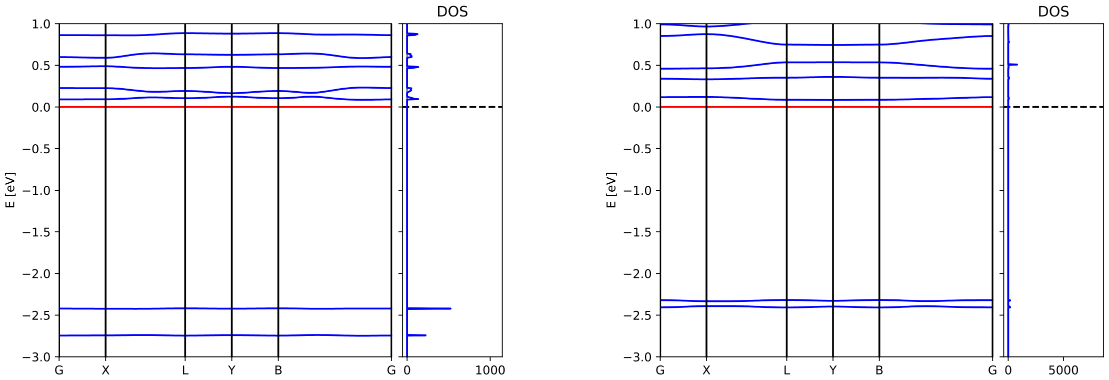
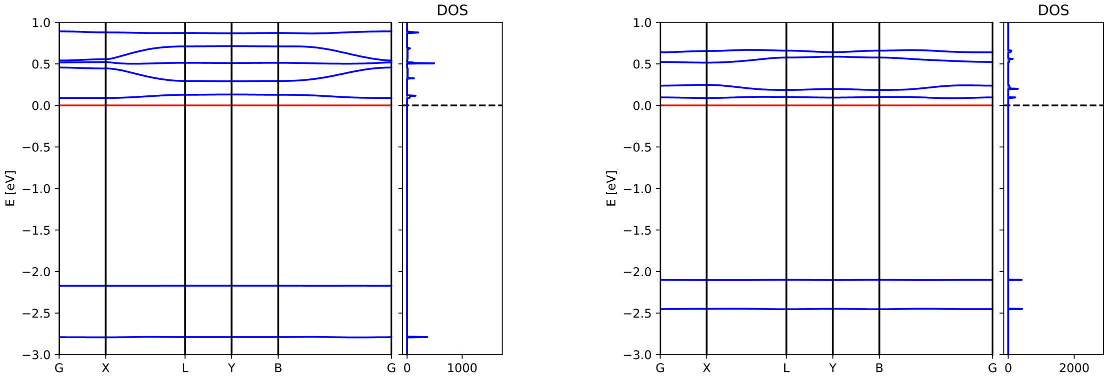


Figure S8. Band structures of COF-2 monolayers computed at PBE0 level.

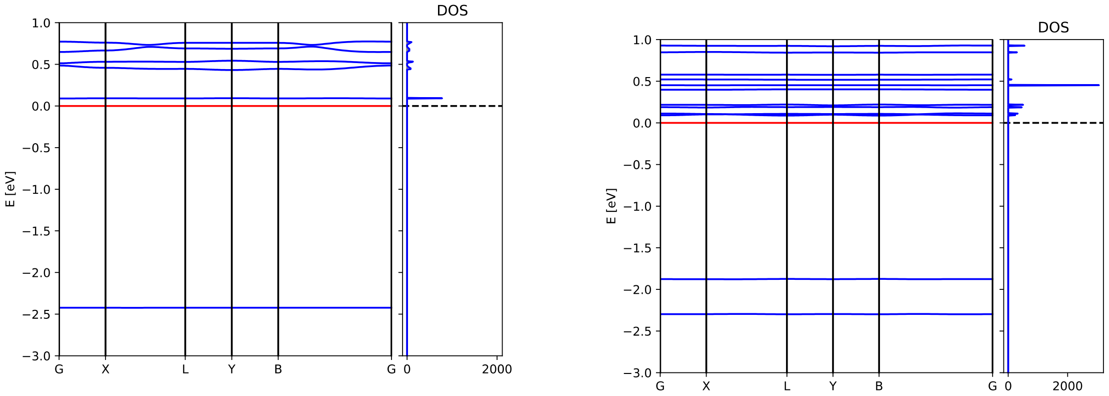
Mod0 **Mod1**



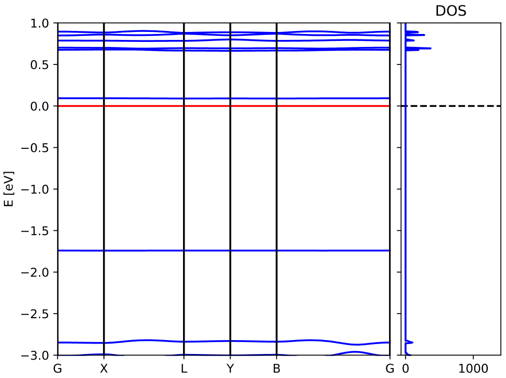
Mod2 **Mod3**

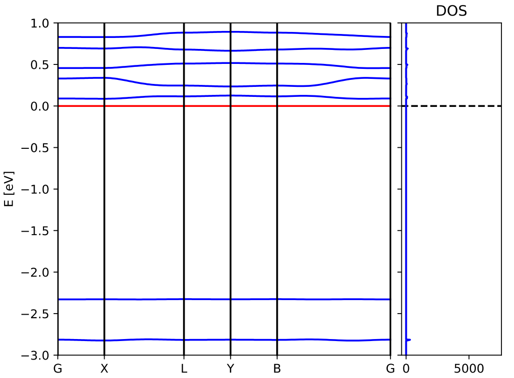


Mod4 **Mod5**



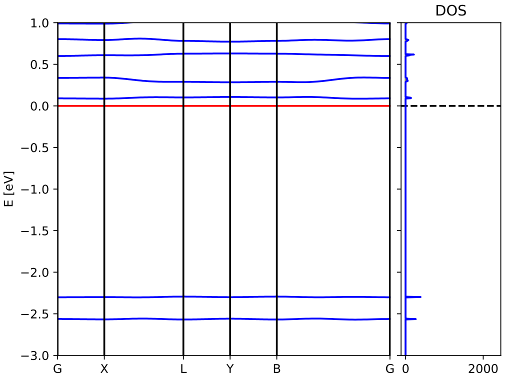
Mod6 **Mod7**

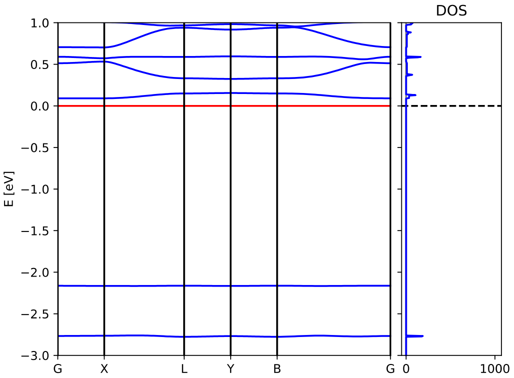




Mod8

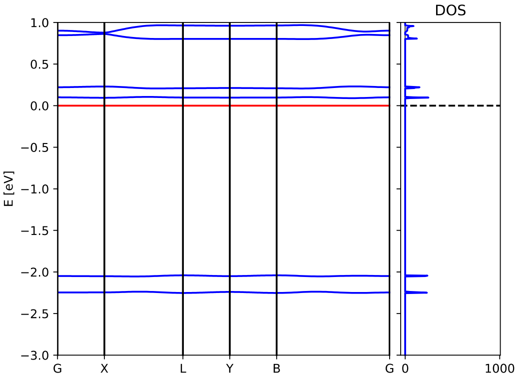
Mod9

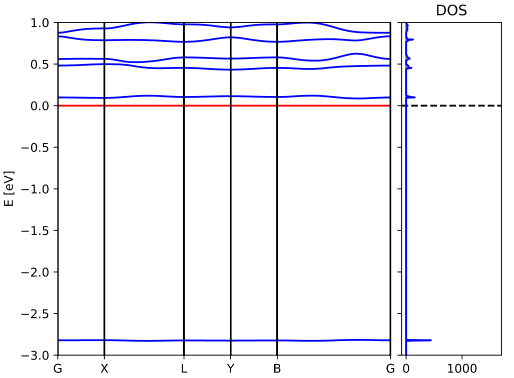




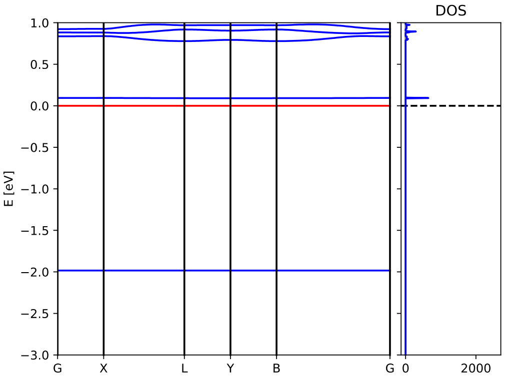
Mod10

Mod11



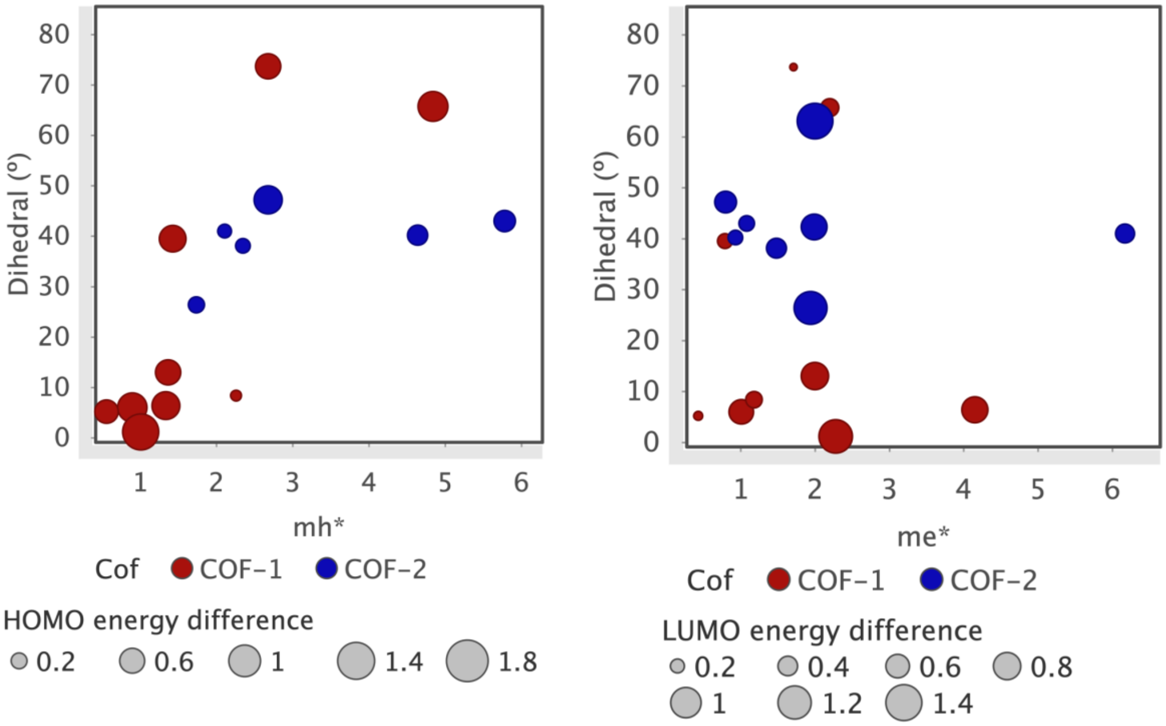


Mod12

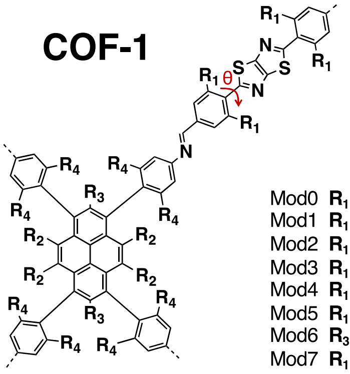


Section 9. Correlation between the effective mass values and the dihedral and orbital energy differences of the D and A building blocks

Figure S9. *Left:* correlation between the m_h^* values and the dihedral and HOMO energy difference (given in eV) between the D and A building blocks. *Right*: correlation between the m_e^* values and the dihedral and LUMO energy difference (given in eV) between the D and A building blocks. The dihedral angle taken is depicted in red in the figure below.

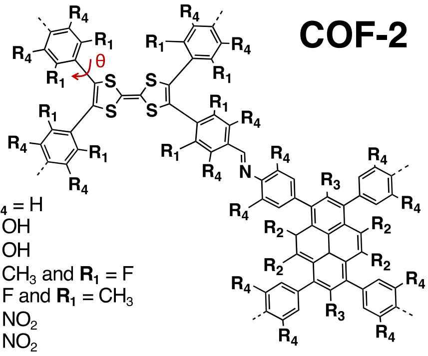


COF-1



Mod0 $\mathbf{R_1}$-$\mathbf{R_4}$ = H
Mod1 $\mathbf{R_1}$ = NO_2 and $\mathbf{R_2}$ = OH
Mod2 $\mathbf{R_1}$ = OH and $\mathbf{R_2}$ = NO_2
Mod3 $\mathbf{R_1}$ = CH_3 and $\mathbf{R_2}$ = F
Mod4 $\mathbf{R_1}$ = COOH and $\mathbf{R_2}$ = OCOH
Mod5 $\mathbf{R_1}$ = OCOH and $\mathbf{R_2}$ = COOH
Mod6 $\mathbf{R_3}$ = OCH_3
Mod7 $\mathbf{R_1}$ = $C(CH_3)_3$
Mod8 $\mathbf{R_3}$ = OCH_3 and $\mathbf{R_4}$ = CH_3

COF-2



Mod0 $\mathbf{R_1}$-$\mathbf{R_4}$ = H
Mod1 $\mathbf{R_2}$ = OH
Mod2 $\mathbf{R_1}$ = OH
Mod3 $\mathbf{R_2}$ = CH_3 and $\mathbf{R_1}$ = F
Mod4 $\mathbf{R_2}$ = F and $\mathbf{R_1}$ = CH_3
Mod5 $\mathbf{R_1}$ = NO_2
Mod6 $\mathbf{R_2}$ = NO_2
Mod7 $\mathbf{R_4}$ = OH
Mod8 $\mathbf{R_4}$ = F