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Supplementary Information

Experimental and Theoretical Insight into the Effect of Fluorine Substituents on the Properties of Azine Linked Covalent Organic Frameworks

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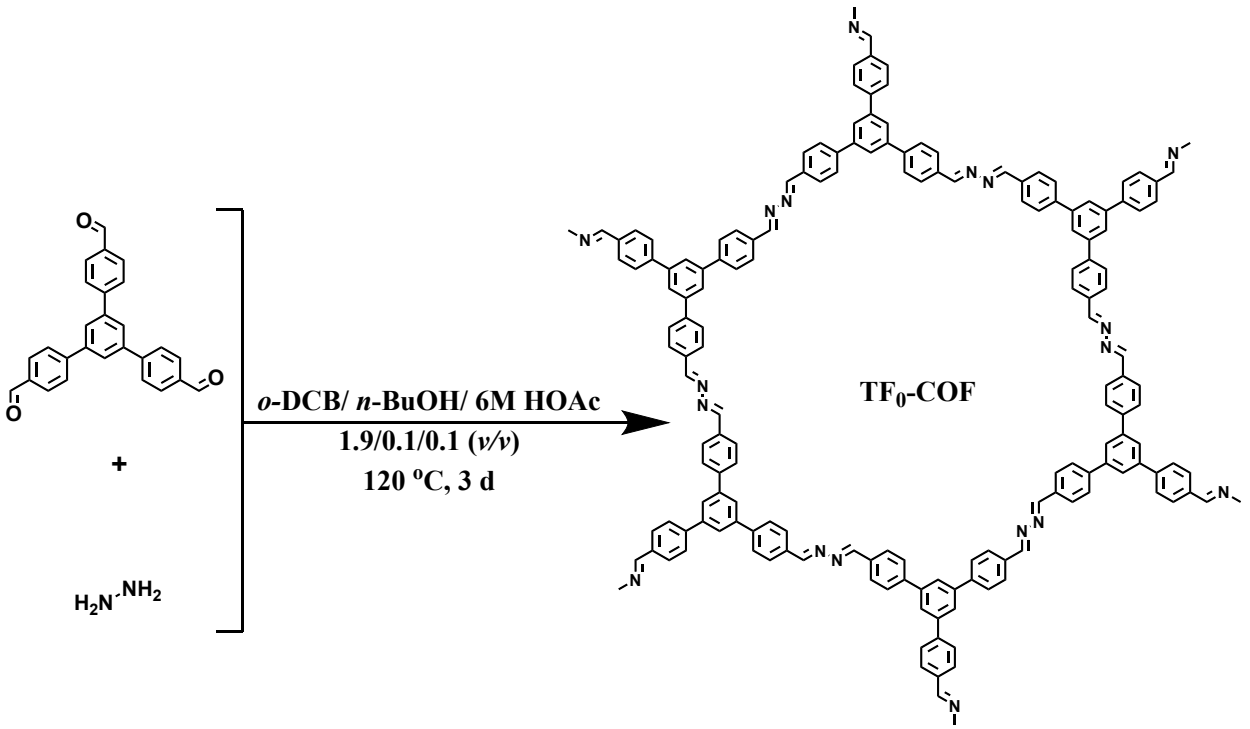
1. General Methods

Reagents were purchased from commercial suppliers (Sigma-Aldrich and Fisher Scientific) and used as received. Low-pressure gas adsorption experiments (up to 760 torr) were carried out on a Micromeritics ASAP 2020 surface area analyzer. Ultrahigh-purity-grade N_2 gas (obtained from Airgas Corporation) were used in all adsorption measurements. N_2 (77 K) isotherms were measured using a liquid nitrogen bath. Pore size distributions were determined using a density functional theory (DFT) cylindrical pores in an oxide surface model in the Micromeritics Software Package. Powder X-ray diffraction of all COFs were carried out on a Bruker D8 Advance diffractometer with a sealed tube radiation source (Cu Kα, λ = 1.54184 Å), a low background sample holder, and Lynxeye XE detector. Scanning electron microscope (SEM) images were acquired with Zeiss-LEO model 1530 SEM instrument. ^{1}H-NMR spectra were carried out on a Bruker 500 MHz Advance Spectrometer. ATR (attenuated total reflectance) FT-IR spectra were taken on a Cary 600 Series FT-IR spectrophotometer.

2. Synthesis, Characterization and Modeling of TF_x-COFs

Here the synthesis of TF_0-COF is shown in detail. Same procedure was followed for all the other COFs synthesized by varying the TF and NF monomer ratios.



Scheme S1 Synthesis of TF_0-COF

1,3,5-*tris*(4-formylphenyl)benzene (30.0 mg, 0.077 mmol) and hydrazine (3.60 μL, 0.115 mmol) were sonicated in a 4 mL ampoule with *o*-dichlorobenzene (1.9 mL) and *n*-butanol (0.1 mL) for about 10 mins. Then 6M acetic acid (0.1 mL) was added, the whole mixture was flash frozen in liquid N_2 and flame sealed. The ampoule was kept in an oven at 120 °C for 72 h. After that, it was cooled to rt, and the formed bright yellow solid was filtered. The filtered solid was soaked in neat THF (20 mL) for 2h and then dried under vacuum. Then it was degassed at 120 °C overnight to afford 24.7 mg (73% yield) of TF_0-COF.

IR Spectra of TF_x-COFs

FT-IR spectroscopy of the TF_x-COF exhibited a stretching vibration band at 1622 cm^{-1} which can be assigned to C=N bond based on previous literature[1,2]

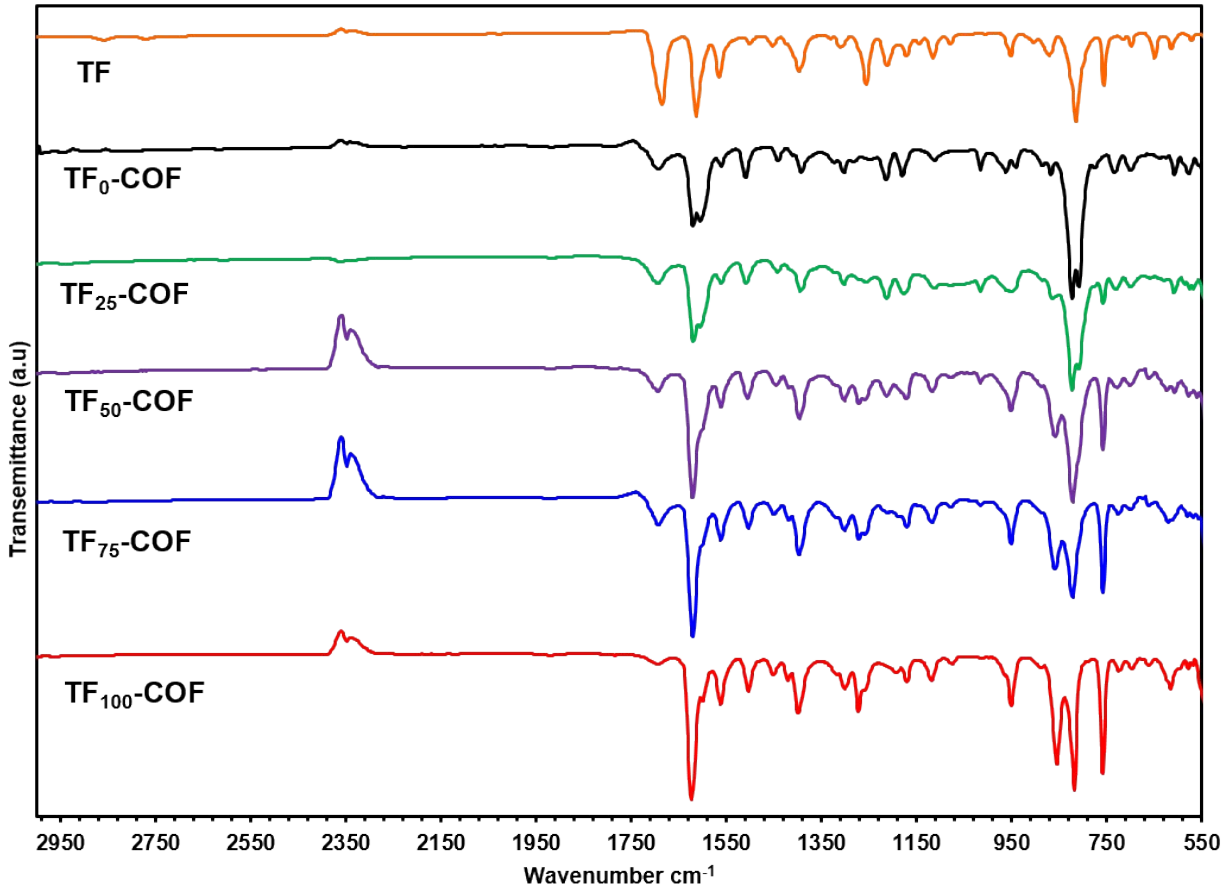


Fig. S1 FT-IR spectra of TF_0-COF (black), TF_{25}-COF (green), TF_{50}-COF (purple), TF_{75}-COF (blue) and TF_{100}-COF (red) compared with TF aldehyde monomer (orange)

Computational Modeling of TF_x-COFs

Computational modeling of the structure of TF_x-COFs were carried out using the Materials Studio (ver. 7.0) suite of programs. For clarity the modeling of TF_{100}-COF is shown here.

The initial lattice was created by starting with the space group *P*3. The constructed model was geometry optimized using the Forcite module (Smart algorithm). Then the calculated PXRD pattern was generated with the Reflex Plus module using Pseudo-Voigt function. Finally, Pawley refinement was applied for profile fitting, producing the refined PXRD profile with the lattice parameters of a = b = 29.522454 Å and c = 3.637044 Å. Rwp and Rp values converged to 5.46% and 3.07%, respectively. The staggered arrangement for TF_{100}-COF was also constructed considering $P6_3/m$ as the space group. Comparison of the observed and the simulated PXRD patterns (Fig.S1) evident that the most preferred structure of TF_{100}-COF is the eclipsed layer arrangement.

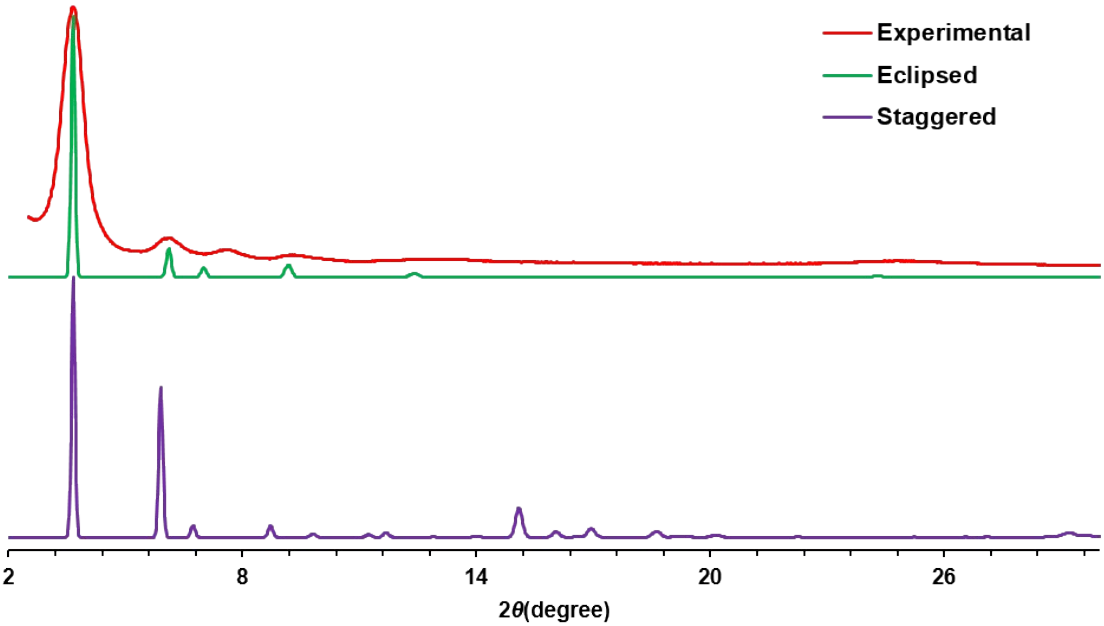


Fig. S2 Experimental powder X-ray diffraction pattern of TF_{100}-COF (red), compared with simulated PXRD patterns of eclipsed layers of COF (green) considering *P*3 symmetry and staggered layers of COF (purple) considering $P6_3/m$ symmetry.

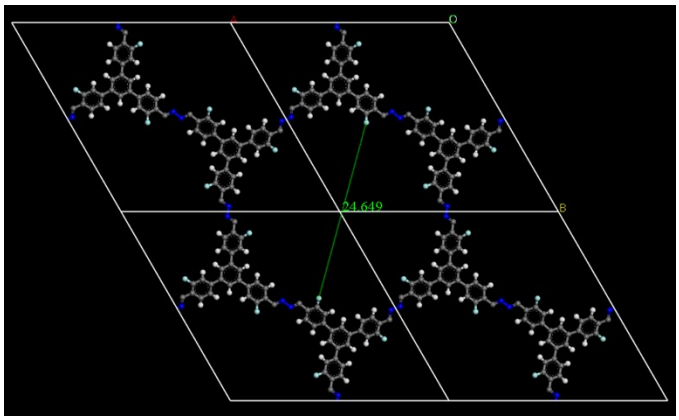


Fig. S3 View of the simulated structure of the eclipsed TF_{100}-COF along *c* axis

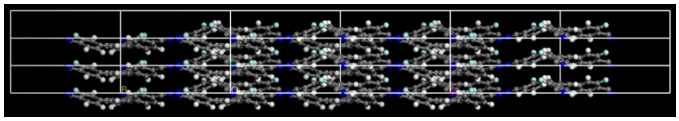


Fig. S4 View of the simulated structure of the eclipsed TF_{100}-COF along *a* axis

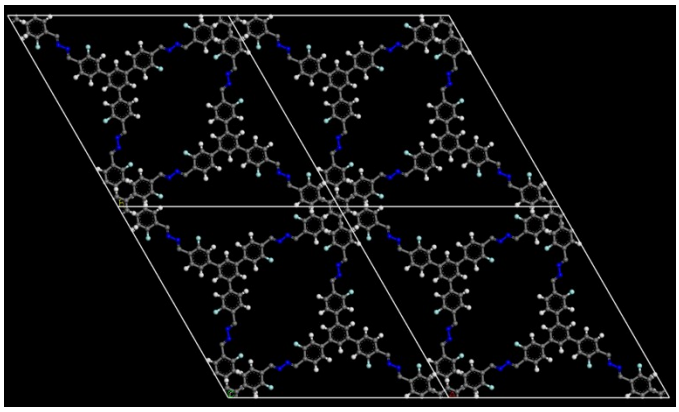


Fig. S5 View of the simulated structure of the staggered TF_{100}-COF along *c* axis

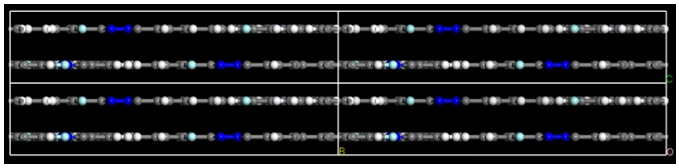


Fig. S6 View of the simulated structure of the staggered TF_{100}-COF along *a* axis

3. Atom Coordinates for Dimer Calculations

TF/TF Dimer

ATOM	ATOMIC CHARGE	COORDINATES (BOHR) X	Y	Z
C	6.0	-1.0981575660	-1.7606577028	7.1035744741
C	6.0	0.3388656641	0.4002061697	7.5075789931
C	6.0	-0.8892483580	2.6866234367	7.8970326219
C	6.0	-3.5116211112	2.8258017557	7.9186132927
C	6.0	-4.9040090135	0.6282394046	7.5697320808
C	6.0	-3.7239885177	-1.6822718688	7.1499861444
C	6.0	-5.2405692119	-4.0197117375	6.7295221121
C	6.0	-7.5516474031	-3.9379999858	5.4561680498
C	6.0	-8.9108328198	-6.1120919402	5.0186019974
C	6.0	-8.0341889341	-8.4500609322	5.8426359116
C	6.0	-5.7634752900	-8.4899530478	7.1306920421
C	6.0	-4.3644922440	-6.3433377093	7.5889505941
H	1.0	-2.6400983829	-6.5195168631	8.6405831063
F	9.0	-4.8660066239	-10.6815060703	7.9584676137
C	6.0	-9.4308665143	-10.8058689375	5.2452368351
O	8.0	-11.2488396063	-10.8467626079	3.9157012192
H	1.0	-8.6856530711	-12.5352516752	6.0709337082
H	1.0	-10.6485870436	-6.0504301812	3.9741882385
H	1.0	-8.2428524776	-2.1788918584	4.7296439966
H	1.0	-6.9299086559	0.7268453067	7.6217940318
C	6.0	-4.8042314813	5.2948232450	8.3002434559
C	6.0	-3.9621695812	7.4489407957	7.0302909003
C	6.0	-5.1940063636	9.7216764466	7.3276392845
C	6.0	-7.2905817579	9.9319651545	8.9048423884
C	6.0	-8.0769534332	7.7774507614	10.1551606909
C	6.0	-6.8855567869	5.4790337343	9.8859692239
H	1.0	-7.5731713821	3.8876954800	10.9379229895
F	9.0	-10.0606176997	7.9086166422	11.6862355834
C	6.0	-8.6300195380	12.3821271784	9.1860336154
O	8.0	-8.0656150772	14.2239297151	8.0222080713
H	1.0	-10.1899694436	12.4145737736	10.5279091420
H	1.0	-4.5843051709	11.3712371585	6.3165036003
H	1.0	-2.3979111003	7.3149781204	5.7469023930
H	1.0	0.2016715574	4.3581238674	8.2685527511
C	6.0	3.1495307147	0.2644104602	7.5598677111
C	6.0	4.3697267850	-1.7037958478	8.8208629655
C	6.0	6.9696117989	-1.8222816672	8.8887419230
C	6.0	8.4441649871	-0.0074266231	7.6869895783
C	6.0	7.1919002668	1.9168624529	6.4448159948
C	6.0	4.5979111980	2.0960651683	6.3666947225

H	1.0	3.7548666403	3.6048034997	5.3068796968
F	9.0	8.5271428552	3.6604559300	5.2168909453
C	6.0	11.2513151473	-0.1728910306	7.7829120695
O	8.0	12.3298195630	-1.6908323275	9.0429624608
H	1.0	12.2821417764	1.1896014065	6.6418388263
H	1.0	7.9145881763	-3.3180375810	9.8802811487
H	1.0	3.2751030066	-3.1074843115	9.7957159108
H	1.0	-0.1514615379	-3.5081062209	6.6954503526
C	6.0	-1.7541948399	-2.6024550413	-0.4161932515
C	6.0	-0.4353361758	-0.4055729915	0.1537670036
C	6.0	-1.7863768735	1.8046316265	0.5918810766
C	6.0	-4.4082582979	1.8483220913	0.4341078539
C	6.0	-5.6781163671	-0.3780585811	-0.1307690384
C	6.0	-4.3742054356	-2.6187444793	-0.5491354748
C	6.0	-5.7690121871	-5.0009519566	-1.0888223196
C	6.0	-7.8915713138	-5.0272947369	-2.6517013005
C	6.0	-9.2449552717	-7.2244035538	-3.0033982041
C	6.0	-8.5446795124	-9.4592124041	-1.8120771469
C	6.0	-6.4105174681	-9.3918436727	-0.3148283496
C	6.0	-5.0101360249	-7.2337010057	0.0559925810
H	1.0	-3.4456507770	-7.2905439634	1.3457116704
F	9.0	-5.7030607501	-11.4739437660	0.9099597549
C	6.0	-10.0955020415	-11.7843501567	-2.1229181746
O	8.0	-11.8371679981	-11.8973746680	-3.5420079051
H	1.0	-9.5553049706	-13.3969478283	-0.9705254728
H	1.0	-10.8862389838	-7.2507085395	-4.1950594119
H	1.0	-8.4701676167	-3.3315302246	-3.6039531230
H	1.0	-7.7059435300	-0.3694036361	-0.2447762072
C	6.0	-5.8267811105	4.2482174040	0.8447075165
C	6.0	-4.9833019159	6.4979550896	-0.2443604675
C	6.0	-6.2981165664	8.7165500910	0.1098119771
C	6.0	-8.4835468767	8.7829739594	1.5684536725
C	6.0	-9.2809734490	6.5361842463	2.6291946640
C	6.0	-8.0133074619	4.2896024031	2.2921431368
H	1.0	-8.7263010771	2.6262088969	3.2055421930
F	9.0	-11.3540784465	6.5168145550	4.0484544698
C	6.0	-9.8662593819	11.2020310936	1.9492334591
O	8.0	-9.3026297088	13.1011868140	0.8861303102
H	1.0	-11.4426876981	11.1546556631	3.2704353834
H	1.0	-5.6650772578	10.4417754315	-0.7478779569
H	1.0	-3.3256531768	6.4893946309	-1.4156126347
H	1.0	-0.7977100312	3.4976749334	1.1209287641
C	6.0	2.3774642652	-0.4288355184	0.2824006516
C	6.0	3.6715675188	-2.4596106538	1.3595633619
C	6.0	6.2678998479	-2.4476297911	1.5051478520
C	6.0	7.6681490102	-0.4300449430	0.5651036594

C	6.0	6.3528808255	1.5502367140	-0.5132306810
C	6.0	3.7566429828	1.5894107338	-0.6699645544
H	1.0	2.8569255428	3.1690704814	-1.5683402890
F	9.0	7.6189216483	3.5088621113	-1.4386294972
C	6.0	10.4604270270	-0.3882819987	0.8141128528
O	8.0	11.6083032837	-1.9165600967	2.0067756154
H	1.0	11.4367161640	1.1447204143	-0.1489293051
H	1.0	7.2620279982	-3.9790259370	2.3875742992
H	1.0	2.6329363214	-4.0160078746	2.1433083180
H	1.0	-0.7299822518	-4.3042666768	-0.8348431469

TF/NF Dimer

ATOM	ATOMIC CHARGE	COORDINATES (BOHR) X	Y	Z
C	6.0	3.4558930918	19.9880286146	12.6746567613
C	6.0	2.8024636398	17.8244624340	13.9984854047
C	6.0	2.8452659334	17.7589456340	16.6229557537
C	6.0	3.4394335785	19.9649172658	17.9258462332
C	6.0	3.4437610510	19.9893325255	9.8608925601
C	6.0	4.3617521413	17.9187408635	8.5341537386
C	6.0	4.3669677850	17.9701792049	5.9341175466
F	9.0	5.3101489228	15.9842850617	4.7122207230
O	8.0	2.2616996511	21.5167602469	0.5556550294
C	6.0	3.4561009617	19.9949450117	4.5591718152
C	6.0	3.4400004963	20.0144847784	1.7450674634
C	6.0	1.0934710455	15.3186290825	20.3060694983
C	6.0	0.5911440835	13.0567971505	21.4935544117
F	9.0	-0.4962609416	13.1462000870	23.7324072757
C	6.0	1.1776961328	10.7439048224	20.4035037703
C	6.0	2.3370241290	10.8017682321	18.0507382238
C	6.0	2.8869910832	13.0404888153	16.8390459204
O	8.0	-0.6490830823	7.9311232787	23.4121743098
C	6.0	0.6243654663	8.2597466280	21.5975082383
C	6.0	4.0861356060	22.1463791515	14.0203117399
C	6.0	4.0158000047	22.1890302670	16.6466718149
C	6.0	2.5264124675	22.0605666944	8.5132533692
C	6.0	2.5253731182	22.0488692905	5.9088519102
C	6.0	4.5538049934	24.5987143603	18.0129814985
C	6.0	5.7061410035	24.6577871946	20.3872143322
C	6.0	6.2380232800	26.9257607361	21.5525705543
C	6.0	5.6244292518	29.2169779071	20.4167507494
C	6.0	4.4596399475	29.1181263407	18.0838840176
C	6.0	3.9140193630	26.8827883671	16.8789191388
F	9.0	3.8632235285	31.2642125559	16.9186600763
O	8.0	7.5700155397	31.8163526950	23.4663527539

C	6.0	6.3090202853	31.6598077942	21.6061253888
C	6.0	2.2517974870	15.3434033902	17.9478237465
C	6.0	10.5145298820	19.9623094439	12.6784173160
C	6.0	9.8756324228	17.8047903865	14.0222392604
C	6.0	9.9453633117	17.7559976615	16.6483914655
C	6.0	10.5270587653	19.9798649983	17.9273391168
C	6.0	10.5297988680	19.9682053890	9.8652200326
C	6.0	11.4523064034	17.9043222542	8.5294105264
C	6.0	11.4667250127	17.9162653225	5.9166942730
O	8.0	9.2461646963	21.3483478669	0.5494378309
C	6.0	10.5380758678	19.9778240943	4.5996119514
C	6.0	10.4620899858	19.9222094584	1.8019104211
C	6.0	9.4085488504	15.3397940136	18.0085217452
C	6.0	8.2456492721	15.2827053915	20.3764617914
C	6.0	7.7130866942	13.0225364184	21.5595436432
C	6.0	8.3411182290	10.7483456784	20.4067540990
C	6.0	9.5311731698	10.7800930750	18.0779691752
C	6.0	10.0636412613	13.0453643083	16.8960400562
O	8.0	6.3957587082	8.0772557893	23.4269141725
C	6.0	7.6679600376	8.2998277162	21.5844691290
C	6.0	11.1693388340	22.1219827890	14.0057608498
C	6.0	11.1183729241	22.1904286642	16.6297398700
C	6.0	9.6364687018	22.0531022767	8.5206421978
C	6.0	9.6278704485	22.0587903519	5.9195288620
C	6.0	11.7109720966	24.6078795313	17.9581794449
C	6.0	12.8831880240	24.6534219276	20.3188629433
C	6.0	13.3928471229	26.9112098459	21.5180074660
C	6.0	12.7640219032	29.1905028460	20.3781814420
C	6.0	11.6080009275	29.1646891890	18.0325023680
C	6.0	11.0703171922	26.9033107913	16.8448473792
O	8.0	14.5612458039	31.8071119349	23.5285436361
C	6.0	13.3462842746	31.6271533291	21.6461119907
H	1.0	12.6001826601	33.3083291568	20.7034032845
H	1.0	5.6103696904	33.3524920532	20.6643993401
H	1.0	14.2953046656	26.9364565851	23.3353380512
H	1.0	13.4290920674	22.9168393339	21.2179945682
H	1.0	7.1920325476	26.9696779680	23.3411395099
H	1.0	6.2781421627	22.9323539843	21.2845507175
H	1.0	2.9560416708	26.9614198654	15.0942430187
H	1.0	11.0771202058	30.9138006660	17.1378493936
H	1.0	10.0874140142	26.9340566331	15.0701868069
H	1.0	10.5421387787	19.9814145736	19.9564323961
H	1.0	9.2741515382	16.1548328321	13.0077209666
H	1.0	11.7594624654	23.7650050490	12.9768428440
H	1.0	11.6089079960	18.4498294551	0.9099219603
H	1.0	4.5701511232	18.5691467540	0.8158325034

H	1.0	12.1873342236	16.2979606784	9.5306818410
H	1.0	12.1833846963	16.3033652947	4.9058609449
H	1.0	8.8760429643	23.6448184762	9.5248614850
H	1.0	8.8753248685	23.6375808256	4.8895904042
H	1.0	1.7797061407	23.6632999963	9.5088366086
H	1.0	1.7921216405	23.6267904902	4.8665357471
H	1.0	5.1411318304	16.3063321645	9.4850260611
H	1.0	2.2140029672	16.1812134068	12.9683201798
H	1.0	4.6780544771	23.7985287880	13.0051509393
H	1.0	3.4333108663	19.9605331015	19.9545237728
H	1.0	0.5334885436	17.0292846356	21.2406712801
H	1.0	2.8627081043	9.0432459197	17.1757006051
H	1.0	3.8733335625	12.9921685217	15.0691852521
H	1.0	1.4763295306	6.6613407985	20.6008478551
H	1.0	8.4030823441	6.6263997650	20.6125452590
H	1.0	10.0432889124	9.0314162350	17.1718455641
H	1.0	11.0314455287	13.0208356650	15.1121576211
H	1.0	7.6799597977	17.0152250743	21.2684124576
H	1.0	6.7611372279	12.9936425080	23.3518164618

NF/NF Dimer

ATOM	ATOMIC CHARGE	COORDINATES (BOHR) X	Y	Z
C	6.0	-0.6599490067	-1.9813965954	8.0395368586
C	6.0	0.7227257040	0.2345527896	8.3258870375
C	6.0	-0.5645745361	2.5175874471	8.5026142119
C	6.0	-3.1874575153	2.5953307743	8.4233968985
C	6.0	-4.5269708844	0.3556275336	8.1410718359
C	6.0	-3.2838902324	-1.9472492468	7.9334854362
C	6.0	-4.7278298596	-4.3308551214	7.5349422254
C	6.0	-6.6900646335	-4.4287051330	5.7761931459
C	6.0	-7.9893835309	-6.6478670522	5.3518929698
C	6.0	-7.3568733455	-8.8227526915	6.6836017706
C	6.0	-5.4320740435	-8.7306852414	8.4620984867
C	6.0	-4.1259143380	-6.5067612127	8.8848301902
H	1.0	-2.6577862182	-6.4496725906	10.2871769484
H	1.0	-4.9377595196	-10.4006738912	9.5138632797
C	6.0	-8.6542458252	-11.2464207570	6.1494896175
O	8.0	-10.1213345957	-11.5618727162	4.4698255705
H	1.0	-8.1505015686	-12.8107359297	7.4061951938
H	1.0	-9.4578895959	-6.7353046737	3.9561035608
H	1.0	-7.1462444869	-2.7772357979	4.6879188468
H	1.0	-6.5566877733	0.4098815667	8.0817722344
C	6.0	-4.5787493765	5.0355339422	8.6254464011

C	6.0	-4.0277997648	7.0278153593	6.9922885107
C	6.0	-5.4074887084	9.2360168677	7.0804631253
C	6.0	-7.3472735375	9.5075704922	8.8330894926
C	6.0	-7.8564979994	7.5605669098	10.5112795533
C	6.0	-6.4966511787	5.3314650319	10.3981605557
H	1.0	-6.9101043275	3.8265061525	11.6978573982
H	1.0	-9.3263079754	7.7749374258	11.9021178802
C	6.0	-8.9161807443	11.8261508955	8.8676525809
O	8.0	-8.8465821362	13.4249913620	7.2850826553
H	1.0	-10.2175972375	11.9884783579	10.4673245268
H	1.0	-5.0316977985	10.7437347470	5.7755128445
H	1.0	-2.5750918089	6.8029568640	5.5914724306
H	1.0	0.4786864899	4.2394679726	8.7736954048
C	6.0	3.5344301039	0.1408412779	8.4339226722
C	6.0	4.7494672222	-1.6636391706	9.9245196341
C	6.0	7.3489931882	-1.8162723386	9.9793216877
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C	6.0	7.6034069979	1.6512614653	7.0789513445
C	6.0	4.9932229774	1.8130787017	7.0316892976
H	1.0	4.0877040786	3.1722074265	5.8235874736
H	1.0	8.7102950979	2.9087040348	5.9283538824
C	6.0	11.5977397154	-0.3683642868	8.5402386563
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H	1.0	3.6445255399	-2.9332137809	11.0622291650
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C	6.0	-2.1610339478	-2.4148241479	-0.9328254393
C	6.0	-0.8087082364	-0.2285056664	-0.4057241696
C	6.0	-2.1105404694	2.0237453548	-0.0517218003
C	6.0	-4.7316282089	2.1116932022	-0.1966826808
C	6.0	-6.0439106265	-0.1034624978	-0.7113684508
C	6.0	-4.7828397831	-2.3777477241	-1.1013701002
C	6.0	-6.2244172529	-4.7290770788	-1.6784924168
C	6.0	-8.2605214156	-4.6841204975	-3.3559076898
C	6.0	-9.6476558796	-6.8380679729	-3.8245408375
C	6.0	-9.0387105774	-9.0902245078	-2.6146626711
C	6.0	-7.0018883187	-9.1573853694	-0.9728498357
C	6.0	-5.5944204031	-6.9995450585	-0.5198069274
H	1.0	-4.0649706749	-7.0591659134	0.8143018254
H	1.0	-6.5465021502	-10.8811934154	0.0029668698
C	6.0	-10.5681603055	-11.3994696648	-3.0626033193
O	8.0	-12.2896061940	-11.5094139227	-4.5069586862
H	1.0	-10.0127887350	-13.0520161438	-1.9549971233
H	1.0	-11.2082860866	-6.8033537065	-5.1217621390
H	1.0	-8.7331607824	-2.9606337050	-4.3200458888

H	1.0	-8.0722102209	-0.0524209989	-0.8278322634
C	6.0	-6.0956513241	4.5507058428	0.1897095919
C	6.0	-5.3686737366	6.6897244828	-1.1710254001
C	6.0	-6.5894367247	8.9651813392	-0.8322542223
C	6.0	-8.5590225326	9.1635269788	0.8957301182
C	6.0	-9.2939558665	7.0466748247	2.2491518706
C	6.0	-8.0782762414	4.7555143454	1.8993068985
H	1.0	-8.6286400380	3.1458457490	3.0098421697
H	1.0	-10.8006154993	7.1885365546	3.6092254585
C	6.0	-9.8688105120	11.6235533724	1.2610519461
O	8.0	-9.4314334321	13.4583072311	0.0372276020
H	1.0	-11.3039062216	11.6596471388	2.7430884493
H	1.0	-6.0442885717	10.6007013870	-1.9025383299
H	1.0	-3.8560614670	6.5489965885	-2.5211590293
H	1.0	-1.0762367445	3.7104391824	0.4126405667
C	6.0	2.0053961154	-0.2785078161	-0.2064714614
C	6.0	3.2016682547	-1.9091901654	1.4812806127
C	6.0	5.7920479469	-1.8741168511	1.7569538398
C	6.0	7.2455117931	-0.2286190500	0.3152440893
C	6.0	6.0735226328	1.3519288689	-1.4137418059
C	6.0	3.4725226805	1.3435195882	-1.6619006226
H	1.0	2.5776429390	2.5883009936	-2.9951212042
H	1.0	7.1881397121	2.6118658767	-2.5589346518
C	6.0	10.0199507965	-0.0894596283	0.6549223356
O	8.0	11.1584540123	-1.0957576140	2.3172009034
H	1.0	11.0178961933	1.0627063065	-0.7446087309
H	1.0	6.7009683525	-3.0691795657	3.1221296879
H	1.0	2.0808717714	-3.1452599340	2.6389645473
H	1.0	-1.1593468935	-4.1472682417	-1.2825570279

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