Supporting Information

Orientation of a Water Molecule: Effects on Electronic Nature of the $C_{59}N$ Cage

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1. Relaxation Time T_2*

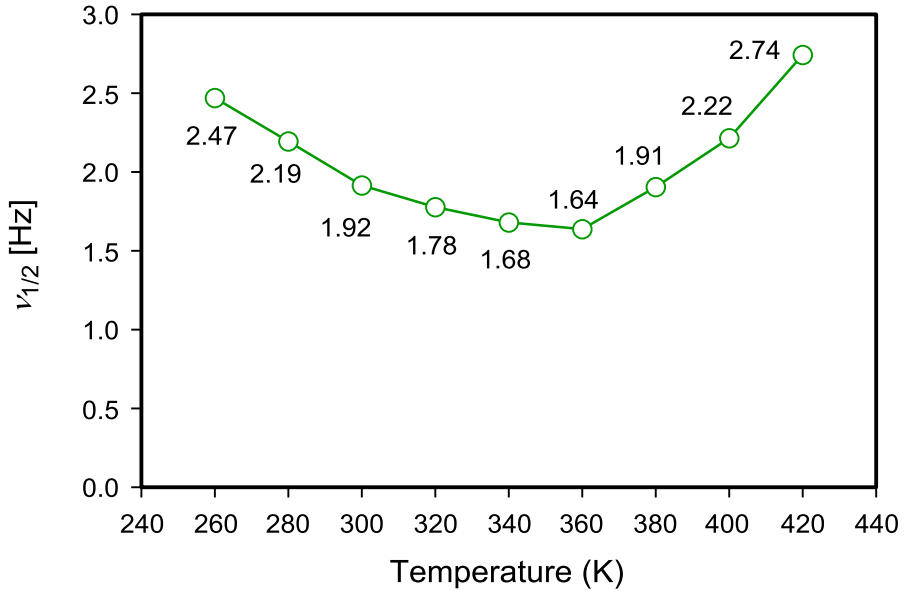


Figure S1. Temperature dependence of $\nu_{1/2}$ for $H_2O@C_{59}N$ dimer.

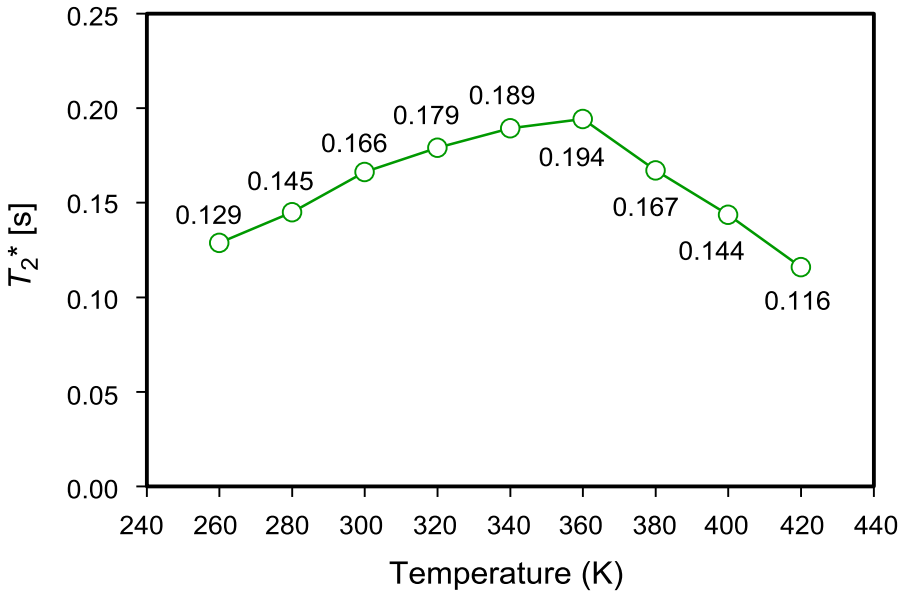


Figure S2. Temperature dependence of T_2* for $H_2O@C_{59}N$ dimer.

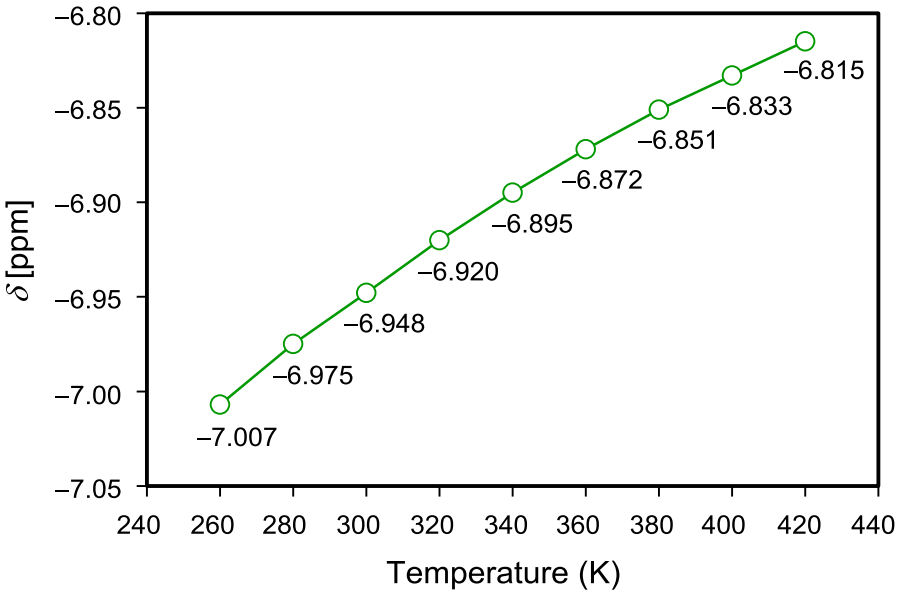
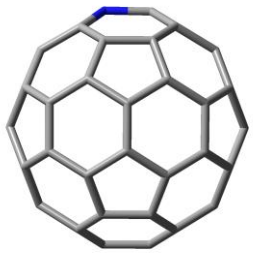


Figure S3. Temperature dependence of ^{1}H NMR chemical shifts for $H_2O@C_{59}N$ dimer.

2. Cartesian Coordinates of Optimized Structures

*Cartesian coordinates used for **Figure 2** were shown below.*

Table S1. Optimized geometry of $C_{59}N^{\bullet}$ (UB3LYP/6-31G(d))



Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	-1.310200	-2.267574	2.393081
2	6	0	-1.437503	-3.028663	1.166645
3	6	0	-2.369964	-2.648082	0.198305
4	6	0	-3.210053	-1.481997	0.423353
5	6	0	-3.084833	-0.748000	1.602830
6	6	0	-2.117537	-1.147996	2.611059
7	6	0	-0.104101	-3.456403	0.767354
8	6	0	-2.011419	-2.679862	-1.206575
9	6	0	-3.364663	-0.790721	-0.845950
10	6	0	-3.115174	0.705265	1.564087
11	6	0	-1.553306	0.053609	3.196228
12	6	0	0.099452	-2.228819	2.755007
13	6	0	-3.269915	1.369265	0.347077
14	6	0	-3.394167	0.605036	-0.883442
15	6	0	-2.478378	2.555548	0.058934
16	6	0	-2.676622	1.329064	-1.921714
17	6	0	0.245981	-3.487212	-0.582594
18	6	0	-2.617738	-1.539352	-1.844981
19	6	0	-2.119553	2.526769	-1.346220
20	6	0	-1.911945	-0.834505	-2.832469
21	6	0	-0.722395	-3.087066	-1.591932
22	6	0	-2.166183	1.197992	2.548509
23	6	0	-0.197748	0.090863	3.543210
24	6	0	1.565183	-3.032969	-0.998698
25	6	0	0.640419	-1.072008	3.320223
26	6	0	0.843870	-2.969621	1.755737
27	6	0	0.592111	1.273815	3.257870
28	6	0	1.953904	-0.608628	2.900842
29	6	0	2.109568	-2.524303	1.352209
30	6	0	-1.941050	0.601874	-2.870163
31	6	0	-1.405537	2.336586	2.269998
32	6	0	-0.000723	-2.364020	-2.612683
33	6	0	1.924490	0.843441	2.862757
34	6	0	2.672650	-1.319093	1.940251
35	6	0	2.480809	-2.558568	-0.044392
36	6	0	3.265705	-1.370064	-0.331518
37	6	0	1.412369	-2.342017	-2.250057
38	6	0	-0.584468	-1.244311	-3.189439
39	7	0	0.183056	-0.090616	-3.518204
40	6	0	3.391668	-0.604132	0.898423
41	6	0	3.362246	0.790587	0.860979
42	6	0	3.205838	1.483498	-0.407866
43	6	0	2.614135	1.530276	1.864040
44	6	0	-1.562570	3.024423	1.004307
45	6	0	1.542062	-0.052657	-3.149604
46	6	0	0.004172	2.375312	2.632374
47	6	0	0.718346	3.090867	1.593464
48	6	0	2.150518	-1.189398	-2.522117
49	6	0	-0.631675	1.046502	-3.250372
50	6	0	-0.095576	2.218724	-2.735024
51	6	0	3.118460	-0.700896	-1.548138
52	6	0	3.088273	0.744626	-1.586652
53	6	0	2.001442	2.677317	1.212681
54	6	0	2.372220	2.651997	-0.183955
55	6	0	2.101984	1.140064	-2.584037
56	6	0	-0.247575	3.485034	0.581337
57	6	0	-0.847769	2.965920	-1.754111
58	6	0	1.316753	2.274381	-2.373583
59	6	0	0.102421	3.458425	-0.768751
60	6	0	1.439725	3.037577	-1.161560

The total electronic energy was calculated to be –2302.7913454 Hartree.

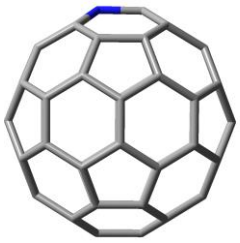
Table S2. Optimized geometry of $C_{60}^{\bullet-}$ (UB3LYP/6-31G(d))

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	-1.112941	-1.564747	-2.980787
2	6	0	-1.385749	-0.174273	-3.265337
3	6	0	-2.404487	0.502706	-2.576120
4	6	0	-3.183319	-0.207117	-1.574287
5	6	0	-2.920894	-1.550832	-1.301144
6	6	0	-1.862684	-2.251021	-2.017338
7	6	0	-0.109224	0.487108	-3.502631
8	6	0	-2.193932	1.854481	-2.108819
9	6	0	-3.454419	0.713704	-0.482745
10	6	0	-2.917547	-2.027552	0.069698
11	6	0	-1.217619	-3.162832	-1.097443
12	6	0	0.331795	-1.765208	-3.052952
13	6	0	-3.177055	-1.143926	1.118825
14	6	0	-3.451068	0.255204	0.835258
15	6	0	-2.393079	-1.208058	2.341632
16	6	0	-2.828649	1.048739	1.883590
17	6	0	0.094392	1.792101	-3.049155
18	6	0	-2.835216	1.987684	-0.814837
19	6	0	-2.182700	0.142236	2.813352
20	6	0	-2.237805	2.749406	0.194666
21	6	0	-0.962086	2.495272	-2.343136
22	6	0	-1.857783	-3.019910	0.192359
23	6	0	0.178889	-3.357245	-1.168236
24	6	0	1.372798	2.165511	-2.457913
25	6	0	0.955422	-2.642559	-2.159101
26	6	0	0.949133	-0.501094	-3.390006
27	6	0	0.960472	-3.410498	0.049096
28	6	0	2.234660	-2.273533	-1.562678
29	6	0	2.182700	-0.142242	-2.813344
30	6	0	-2.234658	2.273529	1.562681
31	6	0	-1.103132	-3.079040	1.370695
32	6	0	-0.341770	3.278627	-1.297119
33	6	0	2.237805	-2.749407	-0.194671
34	6	0	2.828637	-1.048738	-1.883582
35	6	0	2.393074	1.208059	-2.341634
36	6	0	3.177056	1.143929	-1.118826
37	6	0	1.103141	3.079052	-1.370705
38	6	0	-0.960468	3.410475	-0.049098
39	6	0	-0.178890	3.357229	1.168232
40	6	0	3.451058	-0.255207	-0.835258
41	6	0	3.454411	-0.713707	0.482744
42	6	0	3.183322	0.207120	1.574290
43	6	0	2.835204	-1.987679	0.814831
44	6	0	-1.372791	-2.165500	2.457895
45	6	0	1.217615	3.162832	1.097444
46	6	0	0.341775	-3.278615	1.297116
47	6	0	0.962081	-2.495260	2.343126
48	6	0	1.857785	3.019905	-0.192364
49	6	0	-0.955418	2.642540	2.159089
50	6	0	-0.331792	1.765214	3.052960
51	6	0	2.917552	2.027559	-0.069701
52	6	0	2.920900	1.550835	1.301152
53	6	0	2.193932	-1.854481	2.108810
54	6	0	2.404485	-0.502708	2.576123
55	6	0	1.862687	2.251012	2.017341
56	6	0	-0.094395	-1.792092	3.049143
57	6	0	-0.949138	0.501097	3.390020
58	6	0	1.112949	1.564749	2.980800
59	6	0	0.109221	-0.487108	3.502644
60	6	0	1.385756	0.174270	3.265356

The total electronic energy was calculated to be –2286.2498938 Hartree.

*Cartesian coordinates used for **Table 1 and Figure 3** were shown below.*

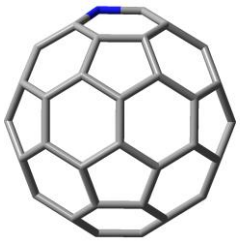
Table S3. Optimized geometry of $C_{59}N^{\bullet}$ (UB3LYP/6-31G(d,p))



Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	-1.310200	-2.267574	2.393081
2	6	0	-1.437503	-3.028663	1.166645
3	6	0	-2.369964	-2.648082	0.198305
4	6	0	-3.210053	-1.481997	0.423353
5	6	0	-3.084833	-0.748000	1.602830
6	6	0	-2.117537	-1.147996	2.611059
7	6	0	-0.104101	-3.456403	0.767354
8	6	0	-2.011419	-2.679862	-1.206575
9	6	0	-3.364663	-0.790721	-0.845950
10	6	0	-3.115174	0.705265	1.564087
11	6	0	-1.553306	0.053609	3.196228
12	6	0	0.099452	-2.228819	2.755007
13	6	0	-3.269915	1.369265	0.347077
14	6	0	-3.394167	0.605036	-0.883442
15	6	0	-2.478378	2.555548	0.058934
16	6	0	-2.676622	1.329064	-1.921714
17	6	0	0.245981	-3.487212	-0.582594
18	6	0	-2.617738	-1.539352	-1.844981
19	6	0	-2.119553	2.526769	-1.346220
20	6	0	-1.911945	-0.834505	-2.832469
21	6	0	-0.722395	-3.087066	-1.591932
22	6	0	-2.166183	1.197992	2.548509
23	6	0	-0.197748	0.090863	3.543210
24	6	0	1.565183	-3.032969	-0.998698
25	6	0	0.640419	-1.072008	3.320223
26	6	0	0.843870	-2.969621	1.755737
27	6	0	0.592111	1.273815	3.257870
28	6	0	1.953904	-0.608628	2.900842
29	6	0	2.109568	-2.524303	1.352209
30	6	0	-1.941050	0.601874	-2.870163
31	6	0	-1.405537	2.336586	2.269998
32	6	0	-0.000723	-2.364020	-2.612683
33	6	0	1.924490	0.843441	2.862757
34	6	0	2.672650	-1.319093	1.940251
35	6	0	2.480809	-2.558568	-0.044392
36	6	0	3.265705	-1.370064	-0.331518
37	6	0	1.412369	-2.342017	-2.250057
38	6	0	-0.584468	-1.244311	-3.189439
39	7	0	0.183056	-0.090616	-3.518204
40	6	0	3.391668	-0.604132	0.898423
41	6	0	3.362246	0.790587	0.860979
42	6	0	3.205838	1.483498	-0.407866
43	6	0	2.614135	1.530276	1.864040
44	6	0	-1.562570	3.024423	1.004307
45	6	0	1.542062	-0.052657	-3.149604
46	6	0	0.004172	2.375312	2.632374
47	6	0	0.718346	3.090867	1.593464
48	6	0	2.150518	-1.189398	-2.522117
49	6	0	-0.631675	1.046502	-3.250372
50	6	0	-0.095576	2.218724	-2.735024
51	6	0	3.118460	-0.700896	-1.548138
52	6	0	3.088273	0.744626	-1.586652
53	6	0	2.001442	2.677317	1.212681
54	6	0	2.372220	2.651997	-0.183955
55	6	0	2.101984	1.140064	-2.584037
56	6	0	-0.247575	3.485034	0.581337
57	6	0	-0.847769	2.965920	-1.754111
58	6	0	1.316753	2.274381	-2.373583
59	6	0	0.102421	3.458425	-0.768751
60	6	0	1.439725	3.037577	-1.161560

The total electronic energy was calculated to be –2302.7913454 Hartree.

Table S4. Optimized geometry of $C_{59}N^{\bullet}$ (UB3LYP/6-311G(d,p))



Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
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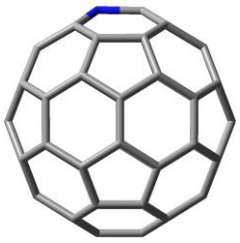
3	6	0	-2.489353	-2.537220	-0.011496
4	6	0	-3.269230	-1.360025	0.330991
5	6	0	-3.097007	-0.746021	1.567868
6	6	0	-2.141668	-1.281378	2.520887
7	6	0	-0.262671	-3.498277	0.447967
8	6	0	-2.146509	-2.455981	-1.416526
9	6	0	-3.402169	-0.548892	-0.866350
10	6	0	-3.057041	0.702688	1.662209
11	6	0	-1.515240	-0.167976	3.204454
12	6	0	0.018657	-2.471330	2.534718
13	6	0	-3.190568	1.480576	0.516399
14	6	0	-3.363380	0.840799	-0.775417
15	6	0	-2.346639	2.648227	0.327224
16	6	0	-2.624153	1.622429	-1.752095
17	6	0	0.071411	-3.421930	-0.900915
18	6	0	-2.702081	-1.235501	-1.938913
19	6	0	-2.004113	2.732020	-1.077459
20	6	0	-1.974216	-0.478168	-2.865160
21	6	0	-0.885371	-2.886856	-1.854813
22	6	0	-2.077152	1.057064	2.672855
23	6	0	-0.158867	-0.226535	3.533668
24	6	0	1.405124	-2.993891	-1.292188
25	6	0	0.619136	-1.400707	3.193973
26	6	0	0.715623	-3.149549	1.462310
27	6	0	0.683031	0.937691	3.346359
28	6	0	1.948451	-0.964022	2.801365
29	6	0	1.994328	-2.729758	1.083683
30	6	0	-1.935618	0.953960	-2.772338
31	6	0	-1.267239	2.177086	2.489689
32	6	0	-0.140632	-2.106876	-2.812975
33	6	0	1.987500	0.483703	2.895117
34	6	0	2.620778	-1.614138	1.771262
35	6	0	2.348464	-2.652040	-0.313079
36	6	0	3.186735	-1.482316	-0.501126
37	6	0	1.273234	-2.183821	-2.470109
38	6	0	-0.673772	-0.915639	-3.276731
39	7	0	0.144692	0.225237	-3.505442
40	6	0	3.361415	-0.841211	0.790233
41	6	0	3.400132	0.547414	0.881055
42	6	0	3.265442	1.359918	-0.315629
43	6	0	2.698268	1.224972	1.956783
44	6	0	-1.403298	2.984928	1.296906
45	6	0	1.504598	0.165254	-3.156864
46	6	0	0.144556	2.116114	2.833546
47	6	0	0.881306	2.889727	1.856611
48	6	0	2.062667	-1.050252	-2.646067
49	6	0	-0.611377	1.369413	-3.127684
50	6	0	-0.015275	2.459174	-2.515068
51	6	0	3.061720	-0.699998	-1.646511
52	6	0	3.101591	0.741074	-1.552741
53	6	0	2.136766	2.453536	1.422446
54	6	0	2.491434	2.539073	0.026008
55	6	0	2.127086	1.272090	-2.495453
56	6	0	-0.073332	3.419628	0.899838
57	6	0	-0.719846	3.146440	-1.460708
58	6	0	1.399601	2.414223	-2.170304
59	6	0	0.260882	3.500721	-0.448692
60	6	0	1.570884	3.055221	-0.896956

The total electronic energy was calculated to be –2303.2139912 Hartree.

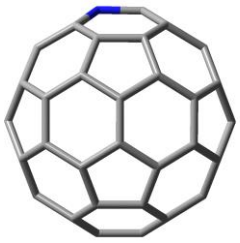
Table S5. Optimized geometry of $C_{59}N^{\bullet}$ (UM06-2X/6-31G(d))



Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	-1.381161	-1.958843	2.600692
2	6	0	-1.543512	-2.831093	1.456784
3	6	0	-2.461062	-2.515389	0.463470
4	6	0	-3.254185	-1.302976	0.573169
5	6	0	-3.094891	-0.465622	1.667260
6	6	0	-2.139868	-0.800361	2.707369
7	6	0	-0.231548	-3.346599	1.099568
8	6	0	-2.113371	-2.700647	-0.931533
9	6	0	-3.388507	-0.735400	-0.756189
10	6	0	-3.069201	0.975299	1.484637
11	6	0	-1.527287	0.429067	3.170249
12	6	0	0.028478	-1.939344	2.954452
13	6	0	-3.203662	1.517893	0.215500
14	6	0	-3.363730	0.641434	-0.930725
15	6	0	-2.368685	2.635928	-0.189249
16	6	0	-2.625150	1.231090	-2.035654
17	6	0	0.107979	-3.523511	-0.233679
18	6	0	-2.676509	-1.607815	-1.675980
19	6	0	-2.020880	2.454511	-1.584416
20	6	0	-1.953191	-1.033959	-2.722750
21	6	0	-0.848739	-3.187812	-1.274760
22	6	0	-2.098399	1.525105	2.412968
23	6	0	-0.177994	0.447528	3.506667
24	6	0	1.439607	-3.165047	-0.693929
25	6	0	0.613003	-0.762269	3.400303
26	6	0	0.738231	-2.802211	2.032340
27	6	0	0.655158	1.563842	3.105846
28	6	0	1.939801	-0.394074	2.936063
29	6	0	2.009323	-2.450828	1.588522
30	6	0	-1.927090	0.389842	-2.902879
31	6	0	-1.299863	2.592797	2.024272
32	6	0	-0.106638	-2.601280	-2.360990
33	6	0	1.966100	1.045715	2.753564
34	6	0	2.621816	-1.217761	2.054639
35	6	0	2.373009	-2.638241	0.203573
36	6	0	3.198916	-1.518162	-0.198775
37	6	0	1.308257	-2.598615	-2.004359
38	6	0	-0.644629	-1.523841	-3.037677
39	7	0	0.160063	-0.444221	-3.483095
40	6	0	3.360757	-0.638924	0.947030
41	6	0	3.385595	0.736989	0.772579
42	6	0	3.249422	1.307424	-0.557118
43	6	0	2.673012	1.599722	1.697673
44	6	0	-1.436724	3.157441	0.698059
45	6	0	1.516292	-0.422830	-3.123734
46	6	0	0.110041	2.612354	2.378087
47	6	0	0.845388	3.193093	1.273202
48	6	0	2.082429	-1.510915	-2.386752
49	6	0	-0.603779	0.740384	-3.323969
50	6	0	-0.025568	1.932758	-2.934853
51	6	0	3.071779	-0.968809	-1.468284
52	6	0	3.097329	0.463290	-1.649826
53	6	0	2.101695	2.698226	0.936197
54	6	0	2.465703	2.521189	-0.450075
55	6	0	2.123502	0.793600	-2.678757
56	6	0	-0.108768	3.521047	0.229838
57	6	0	-0.741888	2.797081	-2.032587
58	6	0	1.389676	1.969276	-2.582874
59	6	0	0.230818	3.348267	-1.103945
60	6	0	1.546623	2.839244	-1.454646

The total electronic energy was calculated to be –2302.0647337 Hartree.

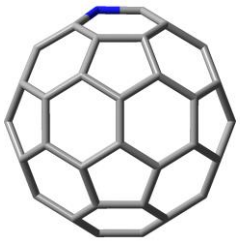
Table S6. Optimized geometry of $C_{59}N^{\bullet}$ (UM06-2X/6-31G(d,p))



Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	1.299293	2.606964	2.006588
2	6	0	1.439561	3.160430	0.676355
3	6	0	2.372477	2.630347	-0.204841
4	6	0	3.204710	1.514537	0.210730
5	6	0	3.066873	0.982257	1.483870
6	6	0	2.094929	1.540788	2.405483
7	6	0	0.113531	3.521968	0.201948
8	6	0	2.027921	2.439419	-1.599418
9	6	0	3.365961	0.629104	-0.928041
10	6	0	3.090196	-0.456739	1.678082
11	6	0	1.520687	0.451841	3.170165
12	6	0	-0.111065	2.631901	2.356776
13	6	0	3.251218	-1.303033	0.590836
14	6	0	3.389173	-0.746349	-0.743039
15	6	0	2.456731	-2.514970	0.488445
16	6	0	2.677411	-1.624275	-1.657631
17	6	0	-0.223428	3.339004	-1.131102
18	6	0	2.631522	1.211492	-2.039048
19	6	0	2.111265	-2.709959	-0.905367
20	6	0	1.934394	0.364146	-2.901845
21	6	0	0.750211	2.779774	-2.053611
22	6	0	2.132522	-0.782004	2.718491
23	6	0	0.170647	0.475146	3.503367
24	6	0	-1.539424	2.829792	-1.479969
25	6	0	-0.659191	1.589714	3.091708
26	6	0	-0.843294	3.203836	1.245461
27	6	0	-0.622088	-0.733923	3.404409
28	6	0	-1.970023	1.070158	2.741242
29	6	0	-2.099657	2.707820	0.910307
30	6	0	1.957034	-1.058109	-2.710383
31	6	0	1.372206	-1.940040	2.618528
32	6	0	0.034834	1.908885	-2.950387
33	6	0	-1.947097	-0.368125	2.934713
34	6	0	-2.673760	1.616529	1.679354
35	6	0	-2.461128	2.520841	-0.475096
36	6	0	-3.246536	1.307671	-0.574650
37	6	0	-1.380799	1.950453	-2.600963
38	6	0	0.612646	0.712552	-3.328456
39	7	0	-0.152759	-0.471687	-3.480026
40	6	0	-3.386238	0.748181	0.759488
41	6	0	-3.363077	-0.626339	0.944351
42	6	0	-3.200030	-1.514495	-0.193985
43	6	0	-2.628260	-1.198088	2.057906
44	6	0	1.536266	-2.821444	1.482082
45	6	0	-1.509727	-0.445436	-3.123568
46	6	0	-0.038008	-1.915370	2.969493
47	6	0	-0.746689	-2.785282	2.053493
48	6	0	-2.116359	0.775673	-2.689620
49	6	0	0.648863	-1.549590	-3.024088
50	6	0	0.107657	-2.620443	-2.339468
51	6	0	-3.092680	0.454662	-1.660522
52	6	0	-3.069573	-0.975594	-1.467479
53	6	0	-2.016397	-2.435580	1.603346
54	6	0	-2.376940	-2.632696	0.219267
55	6	0	-2.079023	-1.526412	-2.379315
56	6	0	0.224163	-3.337591	1.126768
57	6	0	0.846734	-3.198488	-1.246746
58	6	0	-1.307664	-2.612598	-1.986557
59	6	0	-0.112741	-3.524633	-0.205705
60	6	0	-1.442520	-3.167979	-0.672120

The total electronic energy was calculated to be –2302.0647137 Hartree.

Table S7. Optimized geometry of $C_{59}N^{\bullet}$ (UM06-2X/6-311G(d,p))

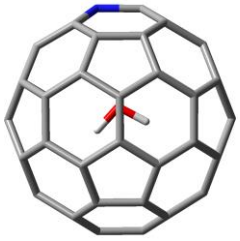


Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	1.365380	2.293684	2.314379
2	6	0	1.539581	2.998355	1.063617
3	6	0	2.462165	2.546597	0.132536
4	6	0	3.249717	1.360727	0.415129
5	6	0	3.079464	0.684224	1.611113
6	6	0	2.118022	1.161060	2.586008
7	6	0	0.233494	3.461839	0.627901
8	6	0	2.127355	2.538300	-1.276078
9	6	0	3.392541	0.615333	-0.820814
10	6	0	3.050092	-0.766552	1.628701
11	6	0	1.498115	0.009775	3.207984
12	6	0	-0.045961	2.326039	2.655008
13	6	0	3.193221	-1.478616	0.449998
14	6	0	3.365054	-0.770134	-0.803880
15	6	0	2.358564	-2.638901	0.196522
16	6	0	2.634463	-1.504423	-1.821968
17	6	0	-0.093938	3.453919	-0.717740
18	6	0	2.691979	1.352909	-1.857430
19	6	0	2.022855	-2.651303	-1.211741
20	6	0	1.976487	0.643035	-2.819900
21	6	0	0.869196	2.975897	-1.693310
22	6	0	2.070854	-1.180283	2.614579
23	6	0	0.148019	0.040820	3.531708
24	6	0	-1.421423	3.037611	-1.134746
25	6	0	-0.637275	1.224753	3.253002
26	6	0	-0.744432	3.054076	1.617664
27	6	0	-0.684498	-1.117223	3.282156
28	6	0	-1.960314	0.799405	2.833326
29	6	0	-2.011142	2.647891	1.216246
30	6	0	1.947363	-0.790758	-2.801836
31	6	0	1.273297	-2.287965	2.370502
32	6	0	0.134837	2.246755	-2.693228
33	6	0	-1.989633	-0.650258	2.851403
34	6	0	-2.631465	1.494030	1.842549
35	6	0	-2.362630	2.642847	-0.182636
36	6	0	-3.188734	1.480624	-0.433444
37	6	0	-1.281256	2.295831	-2.351583
38	6	0	0.673856	1.087619	-3.209843
39	7	0	-0.130297	-0.040948	-3.507892
40	6	0	-3.362714	0.769850	0.820437
41	6	0	-3.390250	-0.614607	0.837400
42	6	0	-3.245332	-1.362416	-0.398494
43	6	0	-2.688681	-1.342068	1.877828
44	6	0	1.418981	-3.029855	1.137871
45	6	0	-1.487766	-0.009457	-3.160043
46	6	0	-0.138030	-2.255645	2.711595
47	6	0	-0.865660	-2.981233	1.692743
48	6	0	-2.055749	1.169955	-2.586780
49	6	0	0.627811	-1.192946	-3.181086
50	6	0	0.043077	-2.316543	-2.636453
51	6	0	-3.053922	0.762498	-1.611972
52	6	0	-3.083098	-0.679616	-1.594446
53	6	0	-2.115578	-2.535133	1.280467
54	6	0	-2.466500	-2.550038	-0.118495
55	6	0	-2.102477	-1.150678	-2.558458
56	6	0	0.095046	-3.451765	0.713883
57	6	0	0.748215	-3.048808	-1.618167
58	6	0	-1.373751	-2.300937	-2.295247
59	6	0	-0.232480	-3.464360	-0.631682
60	6	0	-1.542398	-3.005967	-1.060210

The total electronic energy was calculated to be –2302.5041656 Hartree.

*Cartesian coordinates used for **Tables 2–5** were shown below. Geometries of $HC_{59}N$,*

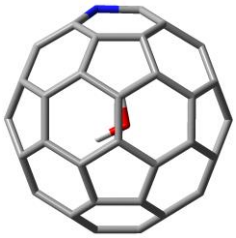
Table S8. Optimized geometry of H_2O-**I**@$C_{59}N^{\bullet}$ (UM06-2X/6-31G(d,p))



Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	1.391181	2.969593	1.336923
2	6	0	1.577874	3.167706	-0.084435
3	6	0	2.498284	2.397173	-0.782540
4	6	0	3.269921	1.385582	-0.080452
5	6	0	3.089155	1.194288	1.282265
6	6	0	2.130320	2.005563	2.011108
7	6	0	0.278409	3.455458	-0.667430
8	6	0	2.166314	1.880316	-2.094090
9	6	0	3.406759	0.238924	-0.960791
10	6	0	3.040071	-0.151190	1.827217
11	6	0	1.493334	1.165140	3.005223
12	6	0	-0.021858	3.138752	1.632165
13	6	0	3.175422	-1.247522	0.986589
14	6	0	3.360689	-1.047417	-0.439866
15	6	0	2.325381	-2.411806	1.165158
16	6	0	2.622290	-2.094234	-1.125872
17	6	0	-0.046040	2.959529	-1.920815
18	6	0	2.717215	0.557316	-2.199587
19	6	0	1.994187	-2.933821	-0.144077
20	6	0	1.992784	-0.447546	-2.840637
21	6	0	0.913434	2.149927	-2.651046
22	6	0	2.051166	-0.167158	2.889475
23	6	0	0.139869	1.327209	3.286074
24	6	0	-1.378233	2.436189	-2.168146
25	6	0	-0.629594	2.337178	2.588861
26	6	0	-0.708497	3.447412	0.395736
27	6	0	-0.706993	0.165638	3.467655
28	6	0	-1.958273	1.802887	2.343513
29	6	0	-1.981608	2.936892	0.159528
30	6	0	1.945442	-1.775635	-2.303268
31	6	0	1.238342	-1.281326	3.057546
32	6	0	0.171301	1.116656	-3.323138
33	6	0	-2.006405	0.459310	2.887874
34	6	0	-2.618123	2.096968	1.159392
35	6	0	-2.329082	2.426065	-1.144043
36	6	0	-3.170920	1.261610	-0.961943
37	6	0	-1.243597	1.301305	-3.030842
38	6	0	0.695213	-0.159491	-3.370818
39	7	0	-0.121219	-1.307724	-3.240719
40	6	0	-3.358043	1.058261	0.464859
41	6	0	-3.404127	-0.227626	0.985549
42	6	0	-3.264640	-1.376778	0.107046
43	6	0	-2.712831	-0.534281	2.225399
44	6	0	1.376820	-2.422341	2.179367
45	6	0	-1.475927	-1.137822	-2.941321
46	6	0	-0.174358	-1.111223	3.353269
47	6	0	-0.908582	-2.150217	2.662357
48	6	0	-2.032086	0.174572	-2.846160
49	6	0	0.620079	-2.271061	-2.516258
50	6	0	0.019962	-3.118836	-1.607567
51	6	0	-3.037877	0.159619	-1.796784
52	6	0	-3.086094	-1.176814	-1.255481
53	6	0	-2.153877	-1.871060	2.107023
54	6	0	-2.501941	-2.390803	0.806374
55	6	0	-2.109894	-1.978547	-1.975115
56	6	0	0.048854	-2.955462	1.927740
57	6	0	0.713966	-3.437466	-0.388019
58	6	0	-1.396176	-2.961707	-1.304848
59	6	0	-0.275396	-3.452552	0.674728
60	6	0	-1.579044	-3.167523	0.100768
61	8	0	-0.039335	-0.161558	-0.330396
62	1	0	0.763556	0.085802	0.152792
63	1	0	-0.763876	0.078085	0.267043

The total electronic energy was calculated to be –2378.4823406 Hartree.

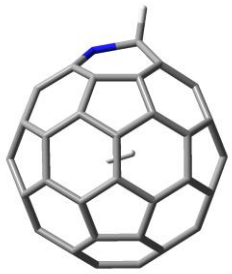
Table S9. Optimized geometry of H_2O-**II**@$C_{59}N^{\bullet}$ (UM06-2X/6-31G(d,p))



Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	1.492604	2.256185	-2.272237
2	6	0	1.547327	1.013159	-3.010662
3	6	0	2.406134	-0.000950	-2.610247
4	6	0	3.244161	0.185516	-1.438435
5	6	0	3.188691	1.376540	-0.727071
6	6	0	2.296569	2.438829	-1.154185
7	6	0	0.198432	0.697903	-3.451801
8	6	0	1.956971	-1.379060	-2.627121
9	6	0	3.304708	-1.079033	-0.728283
10	6	0	3.197304	1.357593	0.725822
11	6	0	1.756878	3.076334	0.030259
12	6	0	0.113044	2.710262	-2.259925
13	6	0	3.261333	0.148290	1.404421
14	6	0	3.312764	-1.097056	0.660241
15	6	0	2.437596	-0.069263	2.580946
16	6	0	2.519878	-2.073937	1.389719
17	6	0	-0.236419	-0.618343	-3.468937
18	6	0	2.502952	-2.036630	-1.473253
19	6	0	1.988286	-1.447342	2.567091
20	6	0	1.727445	-2.957773	-0.766521
21	6	0	0.656421	-1.680935	-3.040626
22	6	0	2.310168	2.407875	1.191136
23	6	0	0.434158	3.503436	0.043720
24	6	0	-1.583147	-0.939015	-3.028819
25	6	0	-0.400899	3.321272	-1.125600
26	6	0	-0.685104	1.752743	-2.995630
27	6	0	-0.386973	3.290354	1.217615
28	6	0	-1.741489	2.998451	-0.675162
29	6	0	-1.969828	1.442552	-2.563088
30	6	0	1.735882	-2.976619	0.668568
31	6	0	1.519875	2.195981	2.313823
32	6	0	-0.138656	-2.639728	-2.318262
33	6	0	-1.733023	2.979193	0.774330
34	6	0	-2.507310	2.083543	-1.376971
35	6	0	-2.433116	0.076077	-2.583286
36	6	0	-3.245585	-0.139738	-1.404144
37	6	0	-1.525056	-2.185818	-2.324113
38	6	0	0.386852	-3.234270	-1.186692
39	7	0	-0.411368	-3.504801	-0.043571
40	6	0	-3.299508	1.104435	-0.658537
41	6	0	-3.291934	1.086458	0.727205
42	6	0	-3.229476	-0.177024	1.439202
43	6	0	-2.490696	2.046310	1.461141
44	6	0	1.583559	0.933934	3.018353
45	6	0	-1.739010	-3.041590	-0.029669
46	6	0	0.140219	2.650225	2.329288
47	6	0	-0.649144	1.673100	3.048383
48	6	0	-2.286284	-2.388152	-1.179891
49	6	0	0.400558	-3.264455	1.096881
50	6	0	-0.111446	-2.699655	2.249769
51	6	0	-3.191411	-1.346940	-0.720309
52	6	0	-3.182960	-1.365759	0.722549
53	6	0	-1.938702	1.373987	2.623266
54	6	0	-2.402442	0.007487	2.613698
55	6	0	-2.272874	-2.419039	1.143727
56	6	0	0.239668	0.606690	3.466357
57	6	0	0.692451	-1.760367	2.988072
58	6	0	-1.497759	-2.246765	2.283652
59	6	0	-0.195135	-0.709607	3.453825
60	6	0	-1.546706	-1.018566	3.020865
61	8	0	-0.179911	0.255525	-0.000802
62	1	0	0.778284	0.383987	-0.018520
63	1	0	-0.304200	-0.702300	0.003242

The total electronic energy was calculated to be –2378.4774279 Hartree.

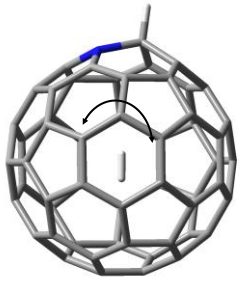
Table S10. Optimized geometry of $H_2@HC_{59}N$ (M06-2X/6-31G(d,p))



Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	1.812129	2.970515	-0.741612
2	6	0	1.810623	2.979105	0.709694
3	6	0	0.628126	3.205499	1.400962
4	6	0	-0.606147	3.427648	0.671684
5	6	0	-0.604682	3.419034	-0.713290
6	6	0	0.630970	3.188256	-1.437540
7	6	0	2.732373	1.956646	1.164552
8	6	0	0.314479	2.416285	2.578342
9	6	0	-1.677588	2.769014	1.403984
10	6	0	-1.674611	2.751727	-1.439796
11	6	0	0.319519	2.385301	-2.606246
12	6	0	2.734781	1.942737	-1.182595
13	6	0	-2.688210	2.102778	-0.740009
14	6	0	-2.689798	2.111634	0.709606
15	6	0	-3.208100	0.842977	-1.187836
16	6	0	-3.210574	0.857154	1.171113
17	6	0	2.431308	1.202046	2.290340
18	6	0	-1.110267	2.154067	2.574392
19	6	0	-3.779668	0.049701	-0.004318
20	6	0	-1.588534	0.917179	2.996603
21	6	0	1.195416	1.432539	3.007115
22	6	0	-1.105029	2.123160	-2.601961
23	6	0	1.201344	1.396803	-3.021778
24	6	0	2.696108	-0.226374	2.299079
25	6	0	2.435833	1.174884	-2.299983
26	6	0	3.308169	1.319320	-0.004782
27	6	0	0.700197	0.103538	-3.457662
28	6	0	2.700727	-0.253566	-2.290911
29	6	0	3.558921	-0.044891	0.003803
30	6	0	-2.655919	0.270647	2.276964
31	6	0	-1.582421	0.881468	-3.010532
32	6	0	0.693193	0.144496	3.457130
33	6	0	1.634777	-0.915553	-3.013453
34	6	0	3.255414	-0.848258	-1.165911
35	6	0	3.252905	-0.834251	1.182295
36	6	0	2.759203	-2.124658	0.741738
37	6	0	1.628657	-0.879680	3.027190
38	6	0	-0.670059	-0.107364	3.448272
39	6	0	-1.172994	-1.401628	3.008120
40	6	0	2.760772	-2.133318	-0.711073
41	6	0	1.740436	-2.769646	-1.403362
42	6	0	0.669684	-3.423016	-0.673015
43	6	0	1.159928	-2.145224	-2.580321
44	6	0	-2.651197	0.243485	-2.285514
45	6	0	-0.268561	-2.391116	2.612464
46	6	0	-0.663089	-0.148262	-3.448098
47	6	0	-1.166989	-1.437243	-2.993808
48	6	0	1.154565	-2.114211	2.607706
49	6	0	-2.404297	-1.178021	2.299501
50	6	0	-2.629865	-1.888163	1.138167
51	6	0	1.737445	-2.752673	1.439636
52	6	0	0.668228	-3.414643	0.715050
53	6	0	-0.263380	-2.422263	-2.584712
54	6	0	-0.561493	-3.192278	-1.410757
55	6	0	-0.564392	-3.174887	1.447236
56	6	0	-2.399628	-1.205283	-2.290133
57	7	0	-3.268207	-1.355958	0.004747
58	6	0	-1.733798	-2.930546	0.731152
59	6	0	-2.627510	-1.901683	-1.120966
60	6	0	-1.732385	-2.939186	-0.699807
61	1	0	-4.874950	0.018174	-0.005293
62	1	0	-0.196120	0.443898	0.050298
63	1	0	0.138255	-0.220913	-0.030864

The total electronic energy was calculated to be –2303.8759722 Hartree.

Table S11. Transition state for the rotation of entrapped H_2 inside the $HC_{59}N$ cage (M06-2X/6-31G(d,p))



Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	-1.852329	2.949671	0.725820
2	6	0	-1.852337	2.949737	-0.725534
3	6	0	-0.673413	3.187597	-1.418935
4	6	0	0.557946	3.430639	-0.692067
5	6	0	0.557956	3.430583	0.692345
6	6	0	-0.673394	3.187475	1.419213
7	6	0	-2.760154	1.911757	-1.173474
8	6	0	-0.349891	2.396234	-2.591861
9	6	0	1.637950	2.783491	-1.421353
10	6	0	1.637972	2.783379	1.421558
11	6	0	-0.349851	2.396023	2.592072
12	6	0	-2.760140	1.911658	1.173707
13	6	0	2.659992	2.144947	0.724746
14	6	0	2.659986	2.145001	-0.724617
15	6	0	3.198209	0.895278	1.179517
16	6	0	3.198199	0.895379	-1.179490
17	6	0	-2.449020	1.154538	-2.295201
18	6	0	1.078351	2.154286	-2.587771
19	6	0	3.780619	0.103348	-0.000009
20	6	0	1.573609	0.922218	-3.003645
21	6	0	-1.217261	1.398082	-3.014747
22	6	0	1.078389	2.154072	2.587943
23	6	0	-1.217215	1.397842	3.014896
24	6	0	-2.693466	-0.277680	-2.294956
25	6	0	-2.448985	1.154367	2.295348
26	6	0	-3.327215	1.273649	0.000110
27	6	0	-0.697286	0.114393	3.458039
28	6	0	-2.693424	-0.277847	2.294988
29	6	0	-3.558436	-0.094710	0.000032
30	6	0	2.650765	0.294802	-2.281267
31	6	0	1.573650	0.921960	3.003715
32	6	0	-0.697337	0.114676	-3.458003
33	6	0	-1.617666	-0.920433	3.020692
34	6	0	-3.241154	-0.886837	1.174143
35	6	0	-3.241179	-0.886743	-1.174164
36	6	0	-2.728460	-2.167087	-0.726368
37	6	0	-1.617711	-0.920189	-3.020731
38	6	0	0.669466	-0.117790	-3.448808
39	6	0	1.191218	-1.402204	-3.001682
40	6	0	-2.728444	-2.167150	0.726218
41	6	0	-1.698180	-2.784168	1.421050
42	6	0	-0.619186	-3.425690	0.693689
43	6	0	-1.125701	-2.145258	2.593763
44	6	0	2.650793	0.294596	2.281265
45	6	0	0.301185	-2.401645	-2.598482
46	6	0	0.669515	-0.118080	3.448799
47	6	0	1.191261	-1.402459	3.001552
48	6	0	-1.125740	-2.145044	-2.593917
49	6	0	2.420033	-1.157567	-2.295267
50	6	0	2.657415	-1.858235	-1.130203
51	6	0	-1.698206	-2.784050	-1.421252
52	6	0	-0.619198	-3.425635	-0.693956
53	6	0	0.301223	-2.401862	2.598291
54	6	0	0.609025	-3.174060	1.428627
55	6	0	0.609006	-3.173937	-1.428883
56	6	0	2.420060	-1.157772	2.295136
57	7	0	3.289384	-1.309913	-0.000034
58	6	0	1.775959	-2.910001	-0.715674
59	6	0	2.657431	-1.858331	1.130030
60	6	0	1.775968	-2.910062	0.715429
61	1	0	4.876209	0.087700	-0.000008
62	1	0	0.185377	-0.012516	-0.001092
63	1	0	-0.554317	0.099275	-0.001175

An imaginary frequency was found at 1004.16 cm^{-1}.

The total electronic energy was calculated to be –2303.8756488 Hartree.