SUPPORTING INFORMATION

Tunable optical properties of OH-functionalised graphene quantum dots

K. R. Geethalakshmi,[*a] Ng Teng Yong,[a] and Rachel Crespo-Otero[*b]

a.) School of Mechanical and Aerospace Engineering, Nanyang Technological University Singapore, 50 Nanyang Avenue, Singapore-639798. E-mail: krgeetha@ntu.edu.sg

b.) School of Biological and Chemical Sciences, Materials Research Institute, Queen Mary University of London, Mile End Road, London E1 4NS, UK. E-mail: r.crespo-otero@qmul.ac.uk

SECTION S1. GO-QDS **3**

G2-QDs 3

G3-QDs 4

G4-QDs 5

SECTION S2. LEVEL OF THEORY **7**

SECTION S3. MOLECULAR ORBITALS **9**

G2 9

G2-2OH-C1 10

G2-12OH 11

G2-18OH 12

G3 13

G4 14

G4-18OH 15

G4-22OH 16

SECTION S4. VERTICAL EXCITATIONS **17**

G2 18

G2-2OH-C1 19

G2-2OH-C2 20

G2-4OH-C1 21

G2-4OH-C2 22

G2-12OH 23

G2-14OH 24

G3 25

G3-2OH-C1 26

G3-2OH-C2 27

G3-2OH-C3 28

G3-2OH-C4 29

G3-4OH-C1 30

G3-4OH-C2 31

G3-14OH 32

G3-16OH 33

G4 34

G4-2OH-C1 35

G4-2OH-C2 36

G4-4OH 37

G4-18OH 38

G4-22OH 39

SECTION S5. GEOMETRY DISTORTION **40**

SECTION S6. FLUORESCENSE WAVELENGHTS **42**

REFERENCES **43**

SECTION S1. GO-QDs

G2-QDs

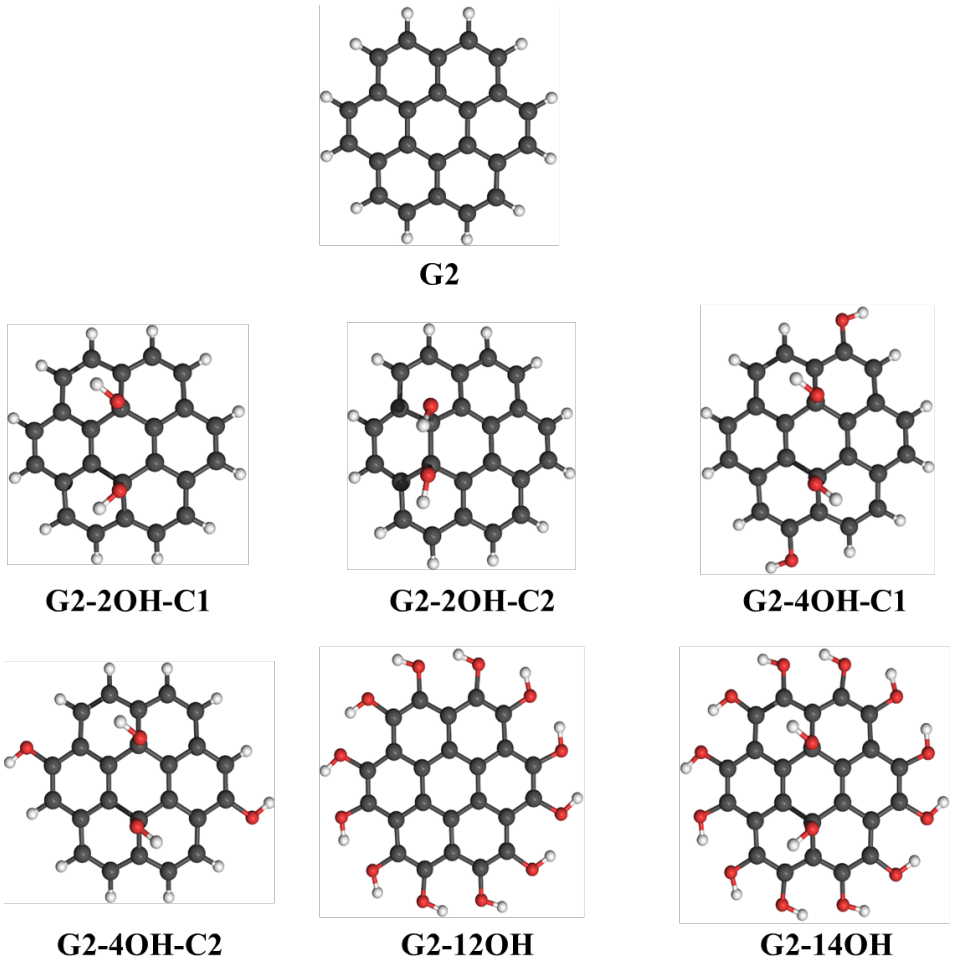


Figure 1S. Geometries of the **G2** based quantum dots.

G3-QDs

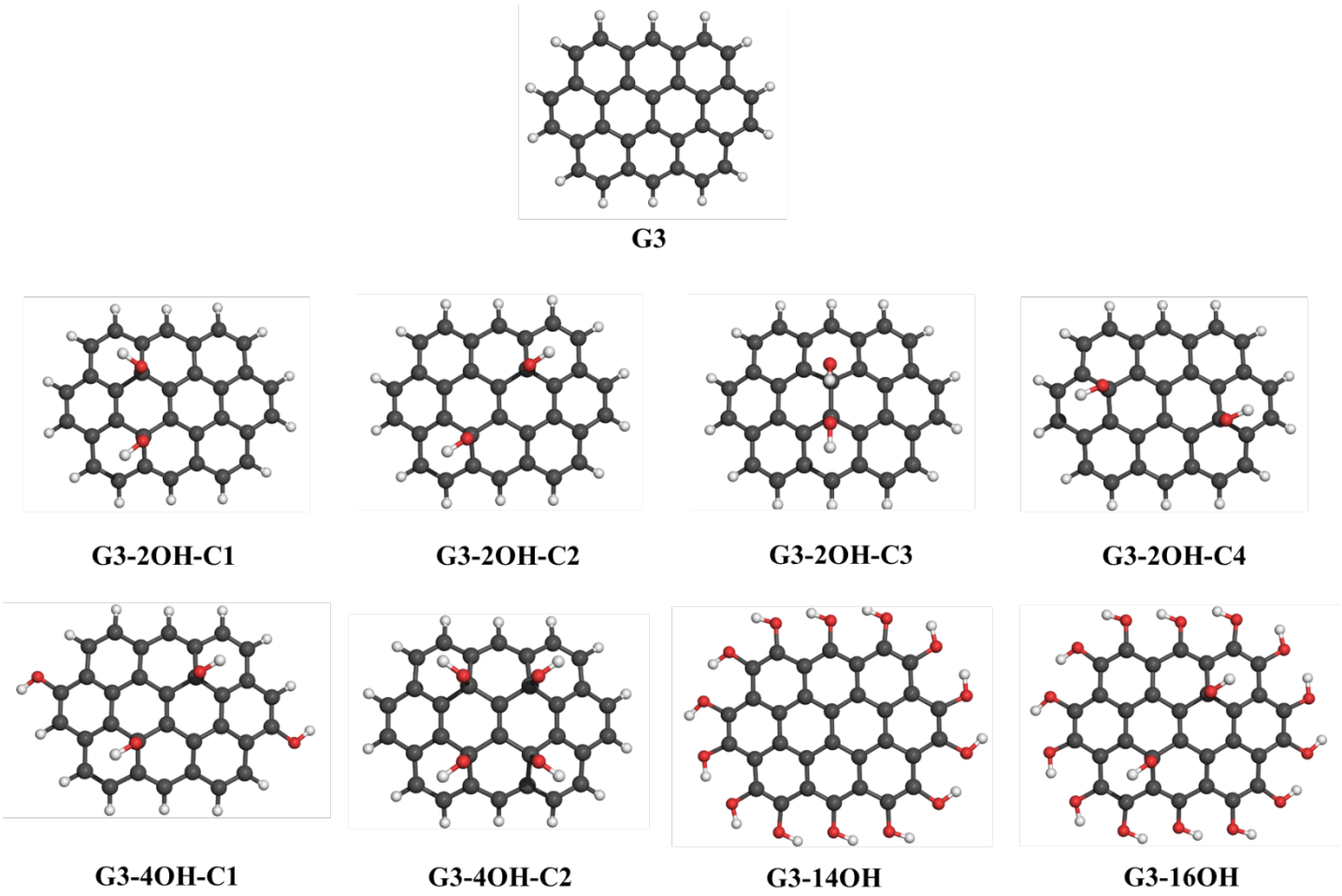
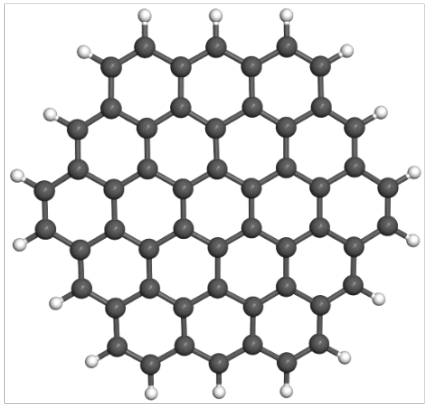
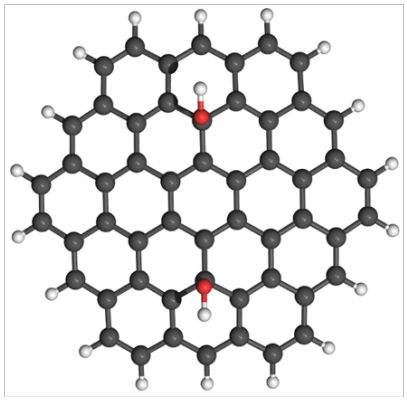


Figure 2S. Geometries of the **G3** based quantum dots**.**

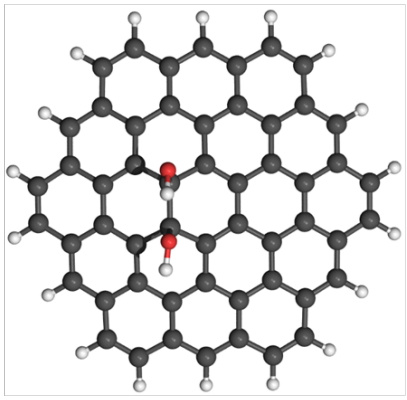
G4-QDs



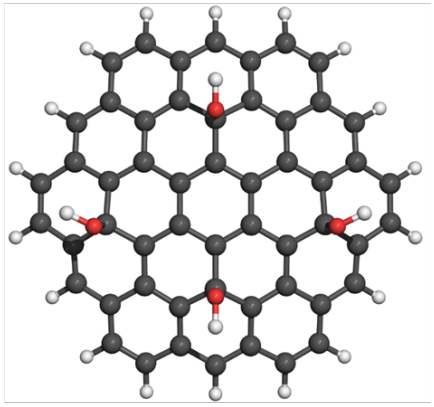
G4



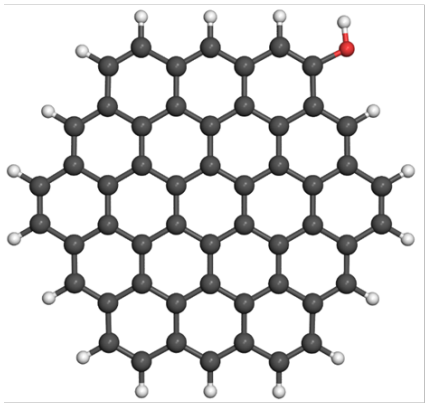
G4-2OH-C1



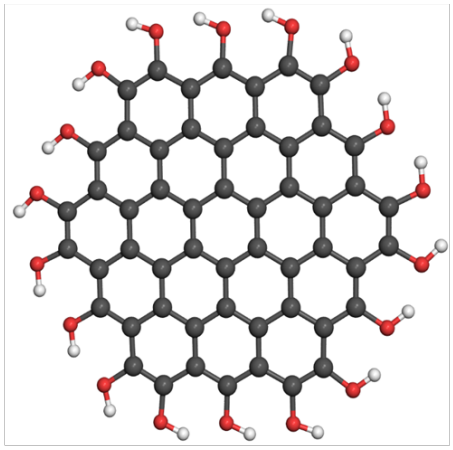
G4-2OH-C2



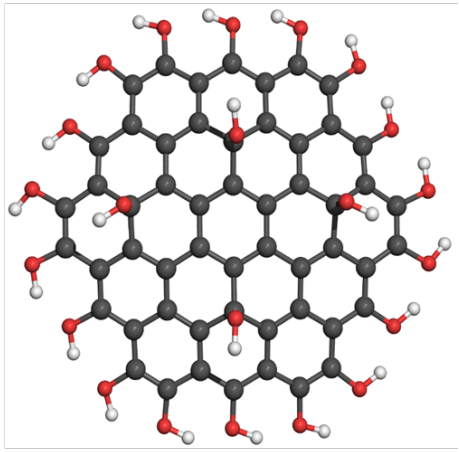
G4-4OH



G4-OH



G4-18OH



G4-22OH

Figure 3S. Geometries of the **G4** based quantum dots.

The total energies of the considered **GO-QDs** calculated at ωB97X-D/6-311+G(d) level of theory are reported in **Table 1S**.

Table 1S. Total ground states energies (Hartree) of the considered **GO-QDs** at ωB97X-D/6-311+G(d) level of theory.

GO-QDs	θ (°)	E(Hartree)
G2	0.	-921.7502520
G2-2OH-C1	20.7	-1073.2495761
G2-2OH-C2	17.9	-1073.2811985
G2-4OH-C1	21.2	-1223.6893289
G2-4OH-C2	20.9	-1223.6907574
G2-12OH	1.3	-1824.4306344
G2-18OH	21.8	-1975.9378379
G3	0.	-1227.811951
G3-2OH-C1	16.0	-1379.348966
G3-2OH-C2	18.6	-1379.324154
G3-2OH-C3	14.1	-1379.316511
G3-2OH-C4	17.6	-1379.282891
G3-4OH-C1	18.3	-1529.763216
G3-4OH-C2	25.9	-1530.858943
G3-14OH	1.3	-2280.943022
G3-16OH	19.2	-2432.454987
G4	0.	-2068.618795
G4-2OH-C1	9.8	-2220.073089
G4-2OH-C2	9.1	-2220.164048
G4-4OH	18.7	-2371.552079
G4-18OH	1.03	-3422.649148
G4-22OH	19.9	-3725.588343

SECTION S2. LEVEL OF THEORY

For **G2** and **G2-2OH-C1,** we perform calculations at RI-CC2/def2-TZVP level of theory to analyse the effect of the functional on the prediction of the excitation energies**.** The predicted values for the S_1-S_0 gaps are 3.31 eV, 3.21 eV and 3.57 eV at TDDFT-ωB97X-D/6-311+G(d), TDDFT-B3LYP/6-311+G(d) and CC2/def2-TZVP levels of theory respectively (**Table 2S**). The value predicted with TDDFT-ωB97X-D/6-311+G(d) is in good agreement with experimental value of 3.54 eV.[1]

The vertical excitation of first bright state corresponds to 4.51 eV, 4.10 eV and 4.71 eV for CC2, TD-B3LYP and TD-ωB97X-D methods respectively. Experimentally, the first intense absorption band in cyclohexane is observed at 4.10 eV,[1] which is closest the TDDFT-B3LYP/6-311+G(d) value.

In the case of **G2-2OH-C1**, both DFT functionals predicted a smallest S_1–S_2 than the obtained with CC2 method. The value of the optical gap with TDDFT-B3LYP is in very good agreement with the CC2 value (1.95 eV and 1.96 eV respectively), but the analysis of the electronic densities and the oscillator strengths show that they correspond to different states. In general, TDDFT-B3LYP/6-311+G(d) level of theory underestimates the excitation energies with respect to the CC2 values. TDDFT-ωB97X-D/6-311+G(d) level of theory provides a better agreement in terms of the ordering of the states and their intensities.

Table 2S. Excitation energies for **G2** and **G2-2OH-C1** calculated at RI-CC2/def2-TZVP, TD-B3LYP/6-311+G(d) and TD-ωB97X-D/6-311+G(d) levels of theory. Oscillator strengths are given in parentheses. (D6h symmetry for **G2**). The % of the contribution of the two most important electron to the excited states are reported. H represents the HOMO orbitals and L the LUMO orbitals.

	G2								
	CC2			**B3LYP**			**ωB97X-D**		
	E (eV) (Osc Str)	Contributions (%)		E (eV) (Osc Str)	Contributions (%)		E (eV) (Osc Str)	Contributions (%)	
S_1	3.31 (0.000)	H-1→L 48.2%	H→L+1 48.2%	3.21 (0.000)	H-1→L 49.8%	H→L+1 49.8%	3.57 (0.000)	H-1→L 47.9%	H→L+1 47.9%
S_2	3.84 (0.000)	H→L 49.2%	H-1→L+1 49.2%	3.41 (0.000)	H→L 49.9%	H-1→L+1 49.9%	3.87 (0.000)	H→L 48.5%	H-1→L+1 48.5%
S_3	4.37 (0.000)	H-1→L+1 46.5 %	H→L 46.5 %	4.10 (0.669)	H-1→L 46.7%	H→L+1 46.7%	4.71 (1.061)	H-1→L 48.5%	H→L+1 48.5%
S_4	4.37 (0.000)	H→L+1 46.4%	H-1→L 46.4%	4.10 (0.669)	H-1→L+1 46.6%	H→L 46.6%	4.71 (1.061)	H-1→L+1 48.5%	H→L 48.5%
S_5	4.51 (1.058)	H→L +2 34.0%	H-1→L +3 34.0%	4.24 (0.000)	H-1→L+2 51.3%	H→L+3 48.2 %	4.71 (0.000)	H-1→L+2 38.7%	H→L+3 39.2%

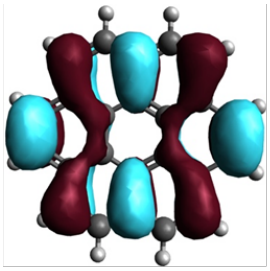
	G2-2OH-C1								
	CC2			**B3LYP**			**ωB97X-D**		
	E (eV) (Osc Str)	Contributions (%)		E (eV) (Osc Str)	Contributions (%)		E (eV) (Osc Str)	Contributions (%)	
S_1	1.96 (0.023)	H-1→L 79.2%	H→L+1 18.1%	1.95 (0.132)	H→L 93.6%	H→ L+2 6.3%	1.80 (0.013)	H-1→L 78.0%	H→L+1 9.6%
S_2	2.27 (0.177)	H→L 84.4%	H→ L+2 5.9%	1.96 (0.021)	H-1→L 82.9%	H→L+1 17.1%	1.87 (0.165)	H→L 92.2%	H→ L+2 7.1%
S_3	3.16 (0.429)	H→L+1 74.2 %	H-1→L 13.3%	3.10 (0.378)	H→L+1 78.2%	H-1→L 16.1%	3.19 (0.462)	H→L+1 74.5%	H-1→L 22.2%
S_4	3.31 (0.000)	H-2→L 46.2%	H-3→L 31.7%	3.11 (0.000)	H-2→L 98%	-	3.53 (0.000)	H-2→L 95.9%	-
S_5	3.45 (0.000)	H-2→L 43.3%	H-3→L 40.1%	3.41 0.005	H-3→L 90.3%	H-6→L 7.8%	3.73 (0.234)	H-1→L+1 66.5%	H→L+2 18.9%

Table 3S. The contributions of the two most important electronic transitions to S_1 for all considered **GQDs** at TD-ωB97X-D/6-311+G(d) level of theory.

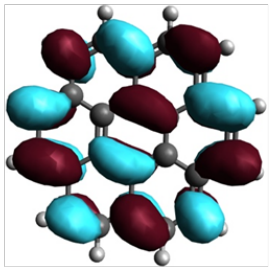
GQs	Contributions (%) to S_1	
G2	H-1→L 47.9%	H→L+1 47.9%
G2-2OH-C1	H-1→L 78.0%	H→L+1 9.6%
G2-2OH-C2	H→L 85.1%	H-1→L+1 6.0%
G2-4OH-C1	H-1→L 51.7%	H→L 37.7%
G2-4OH-C2	H-1→L 43.7%	H→L 42.0%
G2-12OH	H→L 34.0%	H-1→L+1 34.0%
G2-14OH	H-1→L 77.0%	H→L+1 20.0%
G3	H→L 90.0%	H-1→L+1 7%
G3-2OH-C1	H→L 78.9%	H-1→L+1 18.6%
G3-2OH-C2	H→L 94.5 %	H-1→L+1 2.6%
G3-2OH-C3	H→L 97.0%	H-1→L+1 -
G3-2OH-C4	H→L 80.0%	H-1→L+1 17.2%
G3-4OH-C1	H→L 94.5%	H-1→L+1 2.7%
G3-4OH-C2	H-1→L 65.9%	H-1→L+1 39.8%
G3-14OH	H→L 91.0%	H-1→L+1 6.5%
G3-16OH	H→L 94.1%	H-1→L+1 3.0%
G4	H-1→L 42.3%	H→L+1 42.3%
G4-2OH-C1	H→L 96.7%	H-1→L+1 1.0%
G4-2OH-C2	H→L 88.8%	H-1→L+1 3.4%
G4-4OH	H→L 95.6%	H-1→L+1 2.4%
G4-18OH	H→L 44.5%	H-1→L+1 44.5%
G4-22OH	H→L 87.4%	H-1→L 8.1%

SECTION S3. MOLECULAR ORBITALS

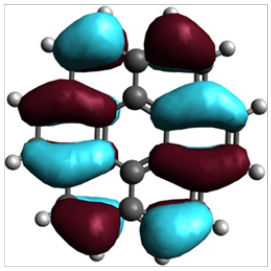
G2



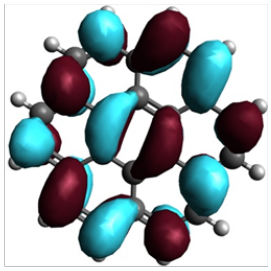
HOMO



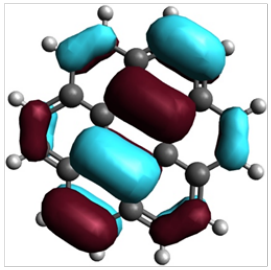
LUMO



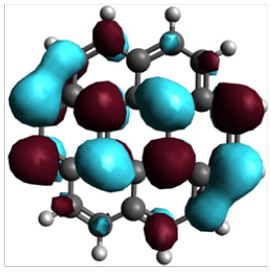
HOMO-1



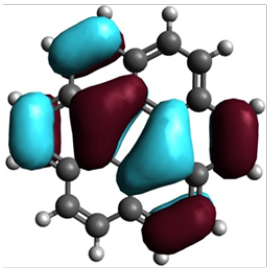
LUMO+1



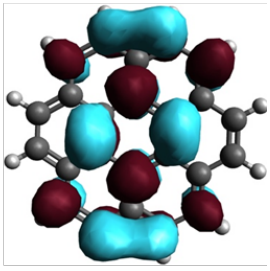
HOMO-2



LUMO+2



HOMO-3



LUMO+3

Figure 4S. Frontier molecular orbitals for **G2** obtained at ωB97X-D/6-311+G(d) level of theory.

G2-2OH-C1

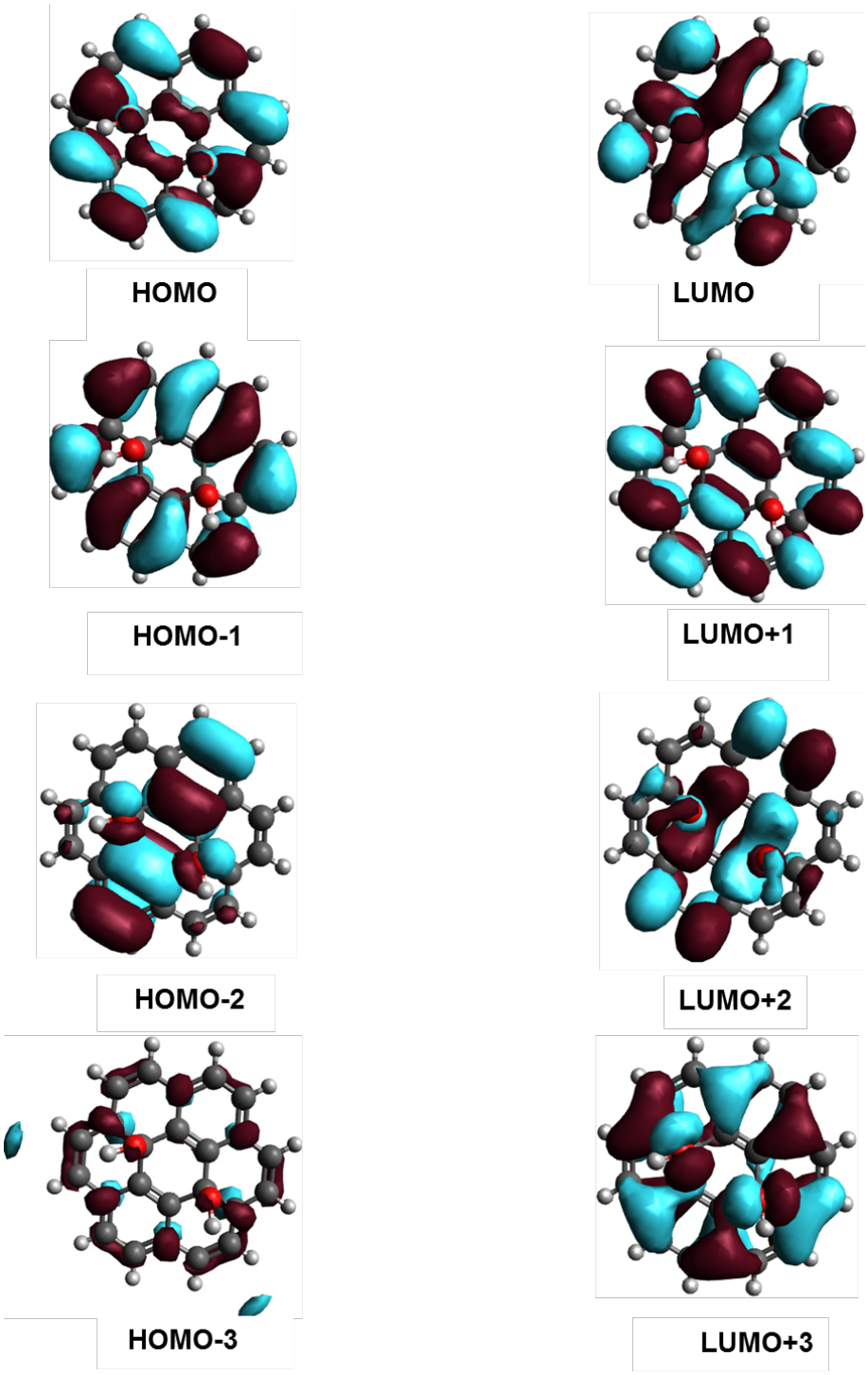


Figure 5S. Frontier molecular orbitals for **G2-2OH-C1** obtained at ωB97X-D/6-311+G(d) level of theory.

G2-12OH

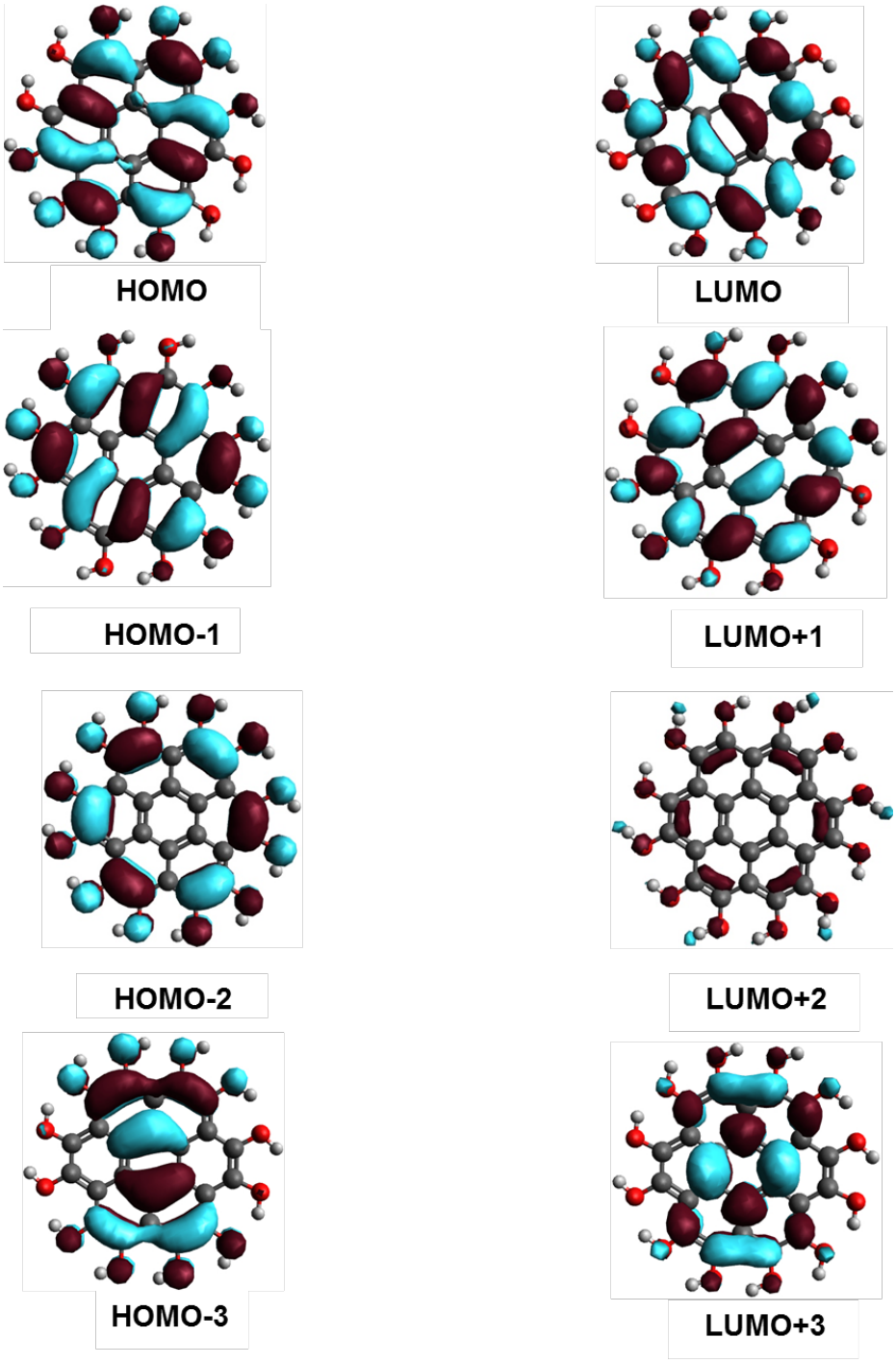


Figure 6S. Frontier molecular orbitals for **G2-12OH** obtained at ωB97X-D/6-311+G(d) level of theory.

G2-18OH

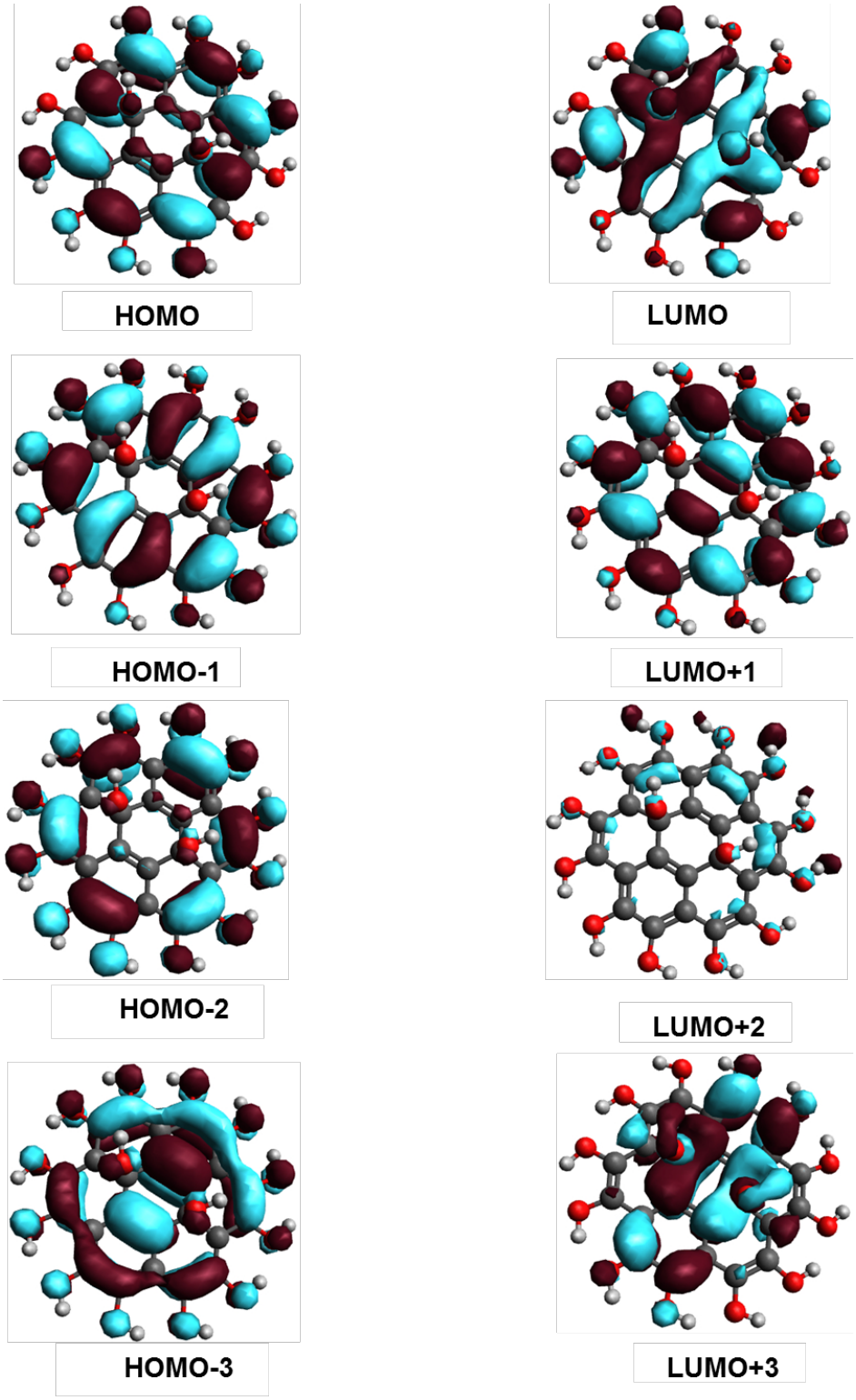


Figure 7S. Frontier molecular orbitals for **G2-12OH** obtained at ωB97X-D/6-311+G(d) level of theory.

G3

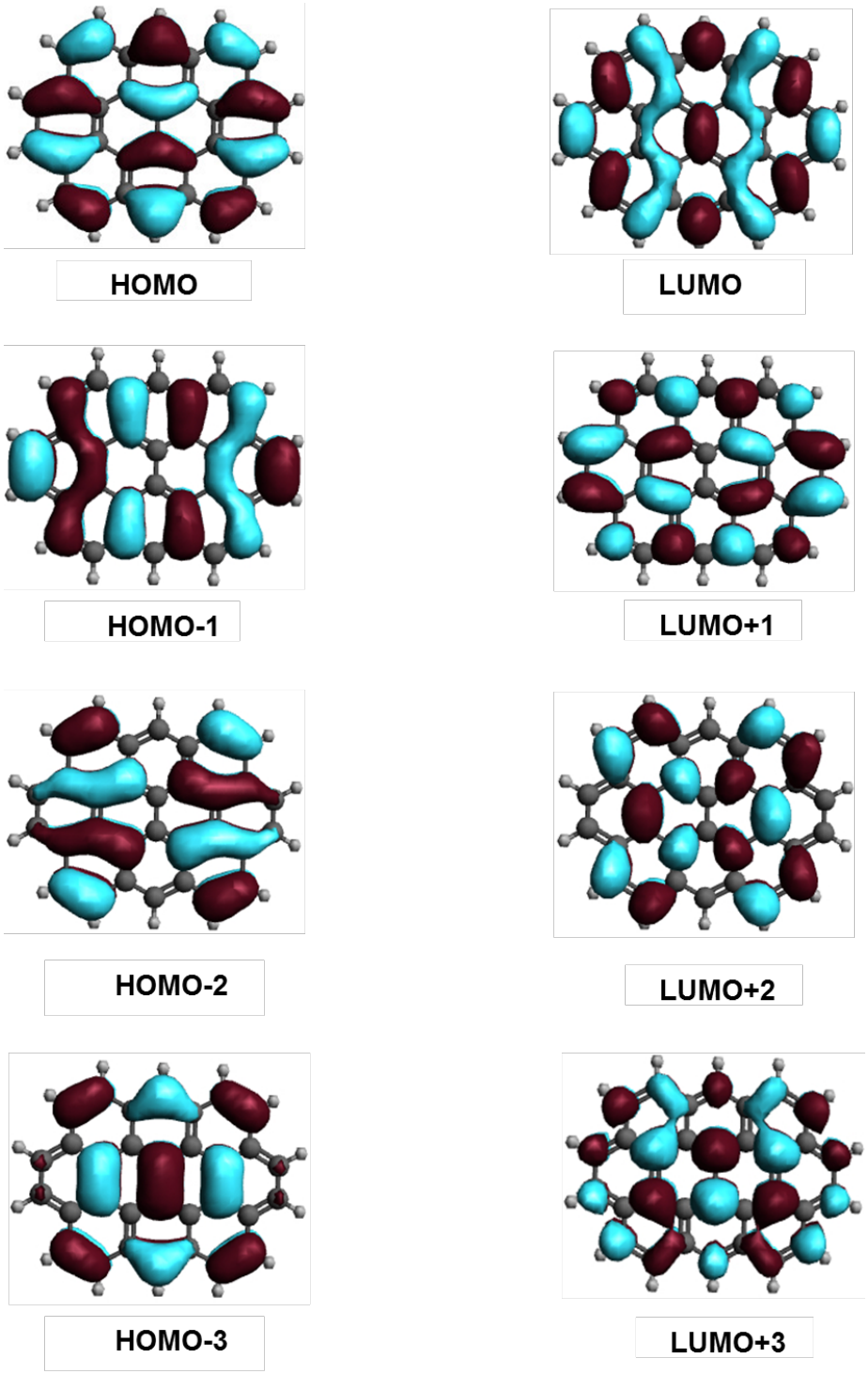


Figure 8S. Frontier molecular orbitals for **G3** obtained at ωB97X-D/6-311+G(d) level of theory.

G4

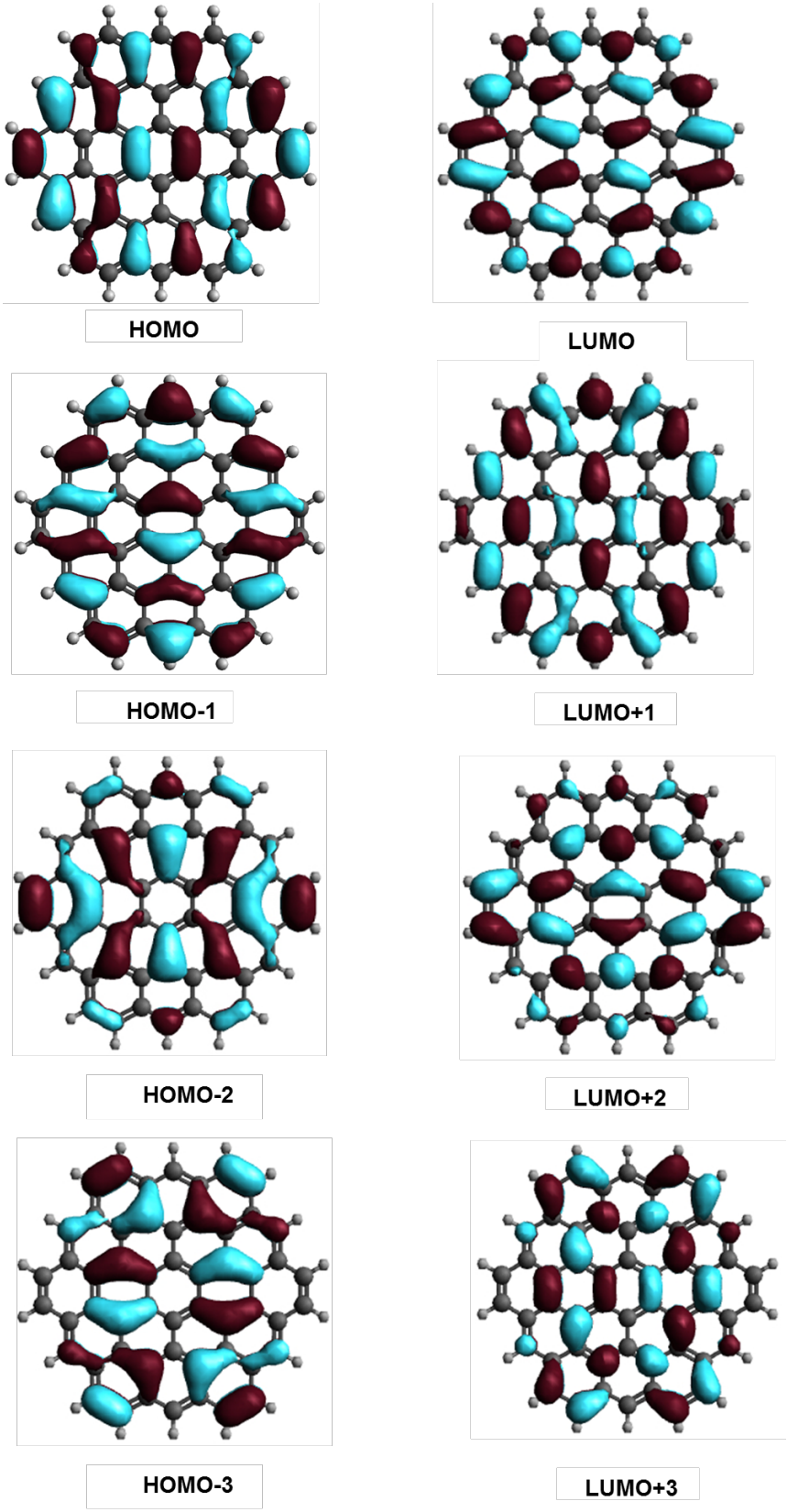
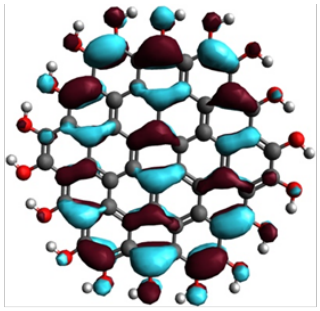
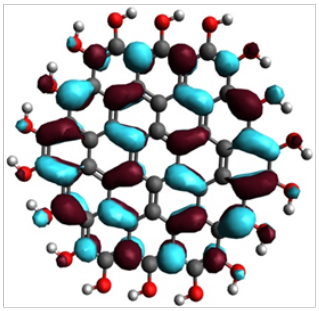


Figure 9S. Frontier molecular orbitals for **G4** obtained at ωB97X-D/6-311+G(d) level of theory.

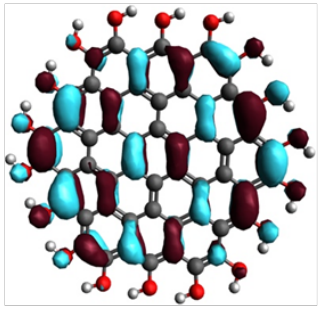
G4-18OH



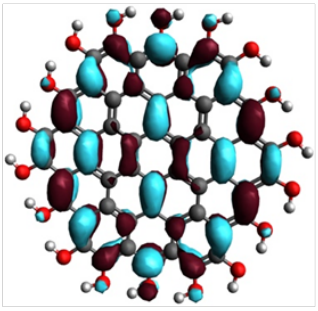
HOMO



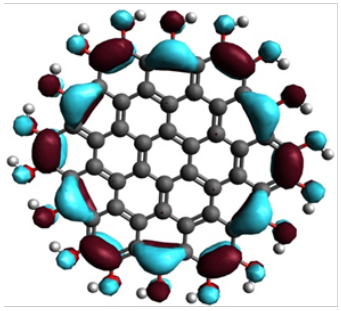
LUMO



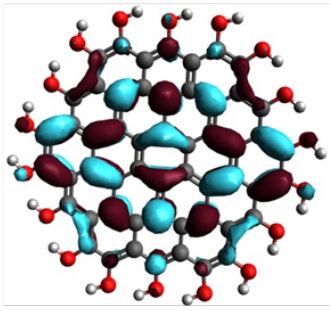
HOMO-1



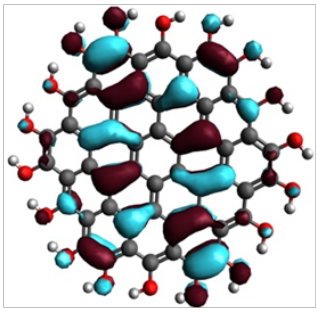
LUMO+1



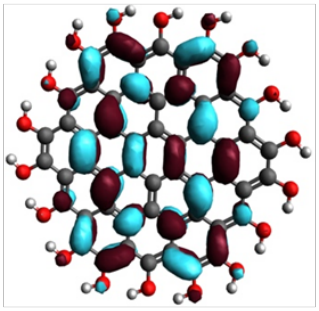
HOMO-2



LUMO+2



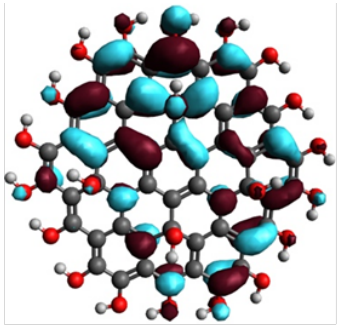
HOMO-3



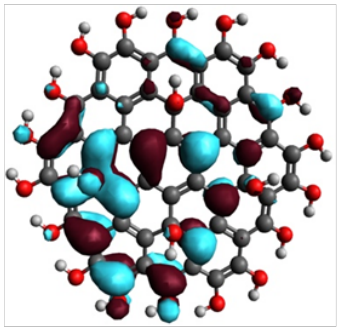
LUMO+3

Figure 10S. Frontier molecular orbitals for **G4-18OH** obtained at ωB97X-D/6-311+G(d) level of theory.

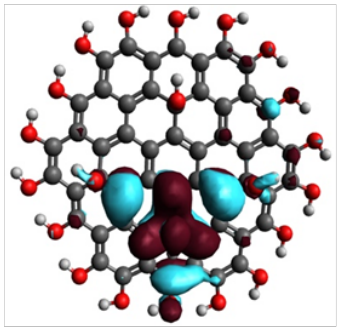
G4-22OH



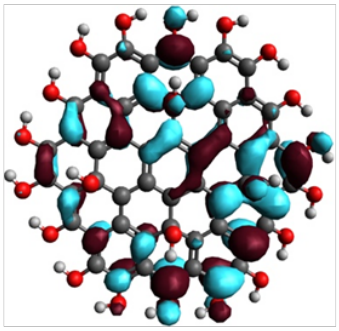
HOMO



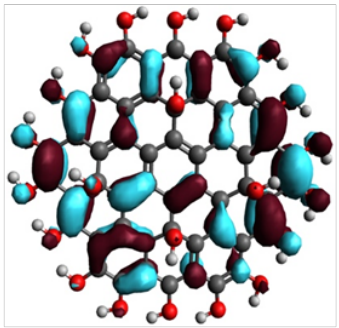
LUMO



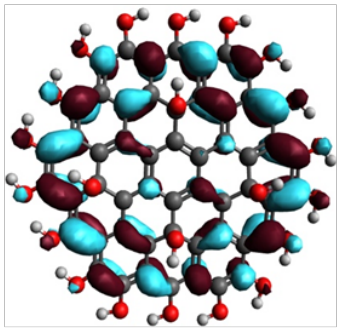
HOMO-1



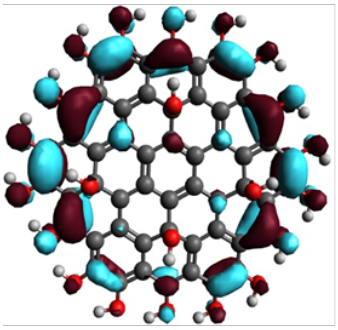
LUMO+1



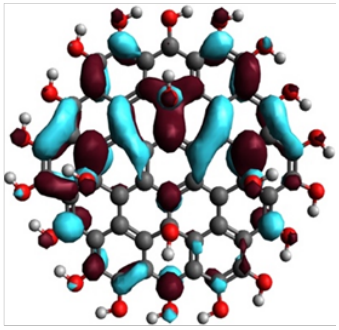
HOMO-2



LUMO+2



HOMO-3



LUMO+3

Figure 11S. Frontier molecular orbitals for **G4-22OH** obtained at ωB97X-D/6-311+G(d) level of theory.

SECTION S4. VERTICAL EXCITATIONS

In this section, we report the energy of the excited states (in eV) for all considered **GO-QDs** calculated at TD-ωB97X-D/6-311+G(d) and TD-B3LYP/6-311+G(d) levels of theory.

G2

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	3.570	0.0000
2	3.867	0.0000
3	4.709	1.0611
4	4.709	1.0612
5	4.711	0.0000
6	4.719	0.0000
7	4.720	0.0000
8	5.103	0.0000
9	5.116	0.0000
10	5.116	0.0000
11	5.301	0.0000
12	5.698	0.0000
13	5.800	0.0000
14	5.800	0.0000
15	6.094	0.0000
16	6.094	0.0000
17	6.125	0.0000
18	6.125	0.0000
19	6.180	0.0000
20	6.328	0.0000

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	3.209	0.0000
2	3.419	0.0000
3	4.095	0.6688
4	4.095	0.6691
5	4.232	0.0000
6	4.234	0.0000
7	4.235	0.0000
8	4.345	0.0000
9	4.543	0.0000
10	4.614	0.0000
11	4.614	0.0000
12	4.832	0.0000
13	4.833	0.0000
14	4.979	0.0000
15	5.158	0.0000
16	5.158	0.0000
17	5.200	0.0000
18	5.201	0.0000
19	5.382	0.0000
20	5.384	0.0000

G2-2OH-C1

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.803	0.0129
2	1.880	0.1657
3	3.174	0.4635
4	3.469	0.0001
5	3.721	0.2522
6	3.870	0.0019
7	3.990	0.0146
8	4.288	0.5357
9	4.348	0.0001
10	4.400	0.0136
11	4.464	0.0120
12	4.603	0.0009
13	4.790	0.0213
14	4.838	0.1852
15	4.928	0.0290
16	5.112	0.0287
17	5.184	0.0025
18	5.286	0.0183
19	5.372	0.0002
20	5.440	0.0141

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.945	0.1319
2	1.966	0.0211
3	3.100	0.3796
4	3.106	0.0001
5	3.407	0.0052
6	3.439	0.0816
7	3.617	0.0272
8	3.859	0.0001
9	3.882	0.0032
10	3.952	0.5443
11	4.023	0.1005
12	4.068	0.0000
13	4.301	0.0725
14	4.347	0.0391
15	4.371	0.1410
16	4.530	0.0010
17	4.669	0.0021
18	4.690	0.0048
19	4.708	0.0458
20	4.711	0.0000

G2-2OH-C2

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	2.859	0.0592
2	3.310	0.0532
3	3.852	0.0242
4	3.945	0.0459
5	4.201	0.1859
6	4.380	0.1270
7	4.431	0.1708
8	4.680	0.1748
9	4.880	0.0593
10	5.042	0.4185
11	5.120	0.6080
12	5.302	0.0481
13	5.436	0.0600
14	5.539	0.0282
15	5.637	0.0033
16	5.702	0.0124
17	5.758	0.0900
18	5.832	0.0440
19	5.896	0.0041
20	5.922	0.0186

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	2.359	0.0593
2	2.459	0.0275
3	3.030	0.0014
4	3.380	0.1261
5	3.524	0.0546
6	3.636	0.0038
7	3.814	0.1024
8	3.829	0.1506
9	4.032	0.1273
10	4.237	0.0440
11	4.374	0.2217
12	4.597	0.0862
13	4.600	0.1034
14	4.737	0.2928
15	4.752	0.0115
16	4.835	0.0858
17	4.888	0.0209
18	4.902	0.1753
19	5.013	0.0151
20	5.060	0.0050

G2-4OH-C1

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.562	0.0567
2	1.902	0.1351
3	3.253	0.4045
4	3.394	0.0000
5	3.599	0.2476
6	3.663	0.0010
7	3.837	0.1838
8	4.233	0.1969
9	4.316	0.2651
10	4.338	0.0009
11	4.426	0.0000
12	4.490	0.0002
13	4.629	0.3297
14	4.829	0.0002
15	4.954	0.0003
16	4.965	0.0207
17	5.136	0.0033
18	5.165	0.1027
19	5.195	0.0000
20	5.291	0.0016

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.712	0.0607
2	1.967	0.1035
3	3.052	0.0001
4	3.139	0.0006
5	3.157	0.3193
6	3.292	0.0567
7	3.500	0.1767
8	3.736	0.0005
9	3.845	0.1321
10	3.914	0.4306
11	3.929	0.0002
12	4.031	0.0003
13	4.120	0.3165
14	4.292	0.0009
15	4.297	0.0000
16	4.304	0.0000
17	4.420	0.0002
18	4.457	0.1080
19	4.502	0.0002
20	4.521	0.0014

G2-4OH-C2

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.702	0.0622
2	1.887	0.1032
3	3.111	0.5004
4	3.247	0.0000
5	3.602	0.1199
6	3.818	0.2556
7	3.925	0.0010
8	4.203	0.0039
9	4.342	0.0002
10	4.354	0.4331
11	4.482	0.0001
12	4.577	0.0008
13	4.671	0.0016
14	4.833	0.0917
15	4.872	0.0000
16	5.068	0.0002
17	5.080	0.0177
18	5.222	0.0023
19	5.251	0.0023
20	5.300	0.5109

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.808	0.0862
2	1.982	0.0520
3	2.796	0.0000
4	3.032	0.4230
5	3.203	0.0100
6	3.413	0.0008
7	3.535	0.2203
8	3.752	0.0014
9	3.790	0.0001
10	3.965	0.4758
11	4.007	0.0007
12	4.019	0.0000
13	4.118	0.0010
14	4.212	0.0000
15	4.235	0.0002
16	4.330	0.1061
17	4.391	0.0004
18	4.585	0.2999
19	4.597	0.0001
20	4.643	0.0318

G2-12OH

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	3.208	0.0000
2	3.485	0.0000
3	4.229	1.0940
4	4.229	1.0940
5	4.366	0.0000
6	4.366	0.0000
7	4.417	0.0000
8	4.782	0.0000
9	4.782	0.0000
10	4.830	0.0000
11	4.930	0.0000
12	4.954	0.0000
13	4.955	0.0000
14	5.014	0.0000
15	5.189	0.0000
16	5.279	0.0000
17	5.279	0.0000
18	5.303	0.0000
19	5.303	0.0000
20	5.594	0.0000

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	2.832	0.0000
2	3.021	0.0000
3	3.616	0.7108
4	3.616	0.7110
5	3.889	0.0000
6	3.889	0.0000
7	3.928	0.0000
8	3.972	0.0000
9	3.973	0.0000
10	4.000	0.0000
11	4.143	0.0000
12	4.169	0.0000
13	4.181	0.0000
14	4.181	0.0000
15	4.248	0.0000
16	4.248	0.0000
17	4.252	0.0000
18	4.252	0.0000
19	4.329	0.0000
20	4.463	0.0000

G2-14OH

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.635	0.0191
2	1.718	0.1537
3	2.918	0.5161
4	3.350	0.0135
5	3.400	0.3229
6	3.740	0.0004
7	3.770	0.0033
8	4.043	0.5931
9	4.106	0.0023
10	4.262	0.0000
11	4.369	0.0008
12	4.483	0.0316
13	4.560	0.2367
14	4.624	0.0068
15	4.681	0.0043
16	4.729	0.0056
17	4.755	0.0000
18	4.826	0.0261
19	4.828	0.0011
20	4.878	0.0049

Level of theory: TD-DFT B3LYP /6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.748	0.1182
2	1.778	0.0289
3	2.816	0.4179
4	2.818	0.0004
5	3.101	0.1289
6	3.168	0.0021
7	3.348	0.0461
8	3.505	0.0003
9	3.577	0.0015
10	3.664	0.6293
11	3.778	0.0312
12	3.787	0.0001
13	3.872	0.0007
14	3.896	0.0002
15	3.952	0.1358
16	3.984	0.1538
17	4.088	0.0107
18	4.114	0.0127
19	4.124	0.0067
20	4.178	0.0404

G3

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	3.155	0.2217
2	3.217	0.0000
3	3.957	0.0000
4	4.139	0.0000
5	4.233	0.0000
6	4.319	1.6946
7	4.516	0.4290
8	4.519	0.0000
9	4.689	0.0000
10	4.727	0.1855
11	4.777	0.0000
12	4.906	0.0000
13	5.246	0.0526
14	5.258	0.0000
15	5.277	0.0277
16	5.424	0.0000
17	5.447	0.0000
18	5.509	0.0000
19	5.659	0.0000
20	5.710	0.0524

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	2.689	0.1623
2	2.835	0.0000
3	3.383	0.0000
4	3.529	0.0000
5	3.623	0.0000
6	3.682	1.0511
7	3.894	0.2963
8	3.937	0.0000
9	3.997	0.0000
10	4.070	0.0008
11	4.096	0.0000
12	4.245	0.0000
13	4.305	0.0000
14	4.460	0.0000
15	4.463	0.0000
16	4.512	0.0117
17	4.539	0.0000
18	4.662	0.0000
19	4.679	0.0149
20	4.750	0.0000

G3-2OH-C1

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	2.695	0.0552
2	3.103	0.1016
3	3.540	0.3545
4	3.714	0.4289
5	3.904	0.3132
6	4.200	0.0018
7	4.532	0.0904
8	4.562	0.0396
9	4.617	0.0579
10	4.644	0.0378
11	4.703	0.1898
12	4.864	0.0212
13	4.971	0.0017
14	5.028	0.0283
15	5.094	0.0310
16	5.124	0.0002
17	5.138	0.0131
18	5.221	0.0932
19	5.270	0.0770
20	5.321	0.3315

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	2.213	0.0295
2	2.270	0.0512
3	2.965	0.4239
4	3.011	0.2748
5	3.388	0.1508
6	3.450	0.0007
7	3.711	0.0621
8	3.742	0.0088
9	3.891	0.0008
10	3.910	0.0035
11	3.941	0.0270
12	3.962	0.0096
13	4.004	0.0490
14	4.023	0.0251
15	4.078	0.0817
16	4.156	0.0612
17	4.181	0.0454
18	4.231	0.0267
19	4.306	0.0045
20	4.322	0.4076

G3-2OH-C2

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.969	0.0198
2	2.533	0.0004
3	3.236	0.0010
4	3.545	0.0028
5	3.654	0.0354
6	3.920	0.0949
7	4.220	0.0011
8	4.231	0.3310
9	4.369	0.6431
10	4.472	0.0000
11	4.506	0.2991
12	4.616	0.2077
13	4.786	0.3763
14	4.842	0.0007
15	4.972	0.6375
16	5.060	0.0225
17	5.181	0.0009
18	5.210	0.0001
19	5.216	0.1559
20	5.340	0.3773

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.391	0.0137
2	1.848	0.0002
3	2.435	0.0003
4	2.659	0.0013
5	2.928	0.0041
6	3.170	0.0348
7	3.269	0.0753
8	3.390	0.0021
9	3.434	0.1827
10	3.600	0.0004
11	3.648	0.0222
12	3.694	0.0045
13	3.796	0.0527
14	4.051	0.0003
15	4.136	0.0002
16	4.136	0.6875
17	4.288	0.0009
18	4.297	0.5923
19	4.351	0.0002
20	4.376	0.0829

G3-2OH-C3

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.371	0.1251
2	1.820	0.0261
3	2.791	0.0033
4	3.122	0.6678
5	3.171	0.0261
6	3.288	0.0002
7	3.608	0.0138
8	3.698	0.0163
9	3.845	0.0012
10	4.039	0.3264
11	4.111	0.0273
12	4.307	0.3505
13	4.365	0.0024
14	4.462	0.0025
15	4.648	0.1782
16	4.707	0.0002
17	4.787	0.0017
18	4.809	0.0006
19	4.885	0.0351
20	4.938	0.0002

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.399	0.0904
2	1.866	0.0344
3	2.372	0.0000
4	2.704	0.0298
5	2.824	0.0020
6	2.913	0.4397
7	3.115	0.0142
8	3.240	0.0024
9	3.318	0.0002
10	3.367	0.0104
11	3.640	0.2247
12	3.715	0.3320
13	3.815	0.0014
14	3.908	0.0016
15	4.094	0.0017
16	4.101	0.2633
17	4.189	0.0038
18	4.206	0.0001
19	4.215	0.0008
20	4.288	0.0016

G3-2OH-C4

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.096	0.0572
2	1.332	0.2078
3	2.112	0.0751
4	2.728	0.1346
5	2.814	0.0001
6	3.021	0.1124
7	3.518	0.7533
8	3.606	0.0002
9	3.655	0.0000
10	3.756	0.0000
11	3.768	0.0076
12	3.908	0.0926
13	3.921	0.0003
14	4.234	0.0005
15	4.246	0.0838
16	4.381	0.0002
17	4.491	0.0010
18	4.513	1.0321
19	4.595	0.0002
20	4.646	0.0229

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.250	0.0427
2	1.482	0.1375
3	2.150	0.0454
4	2.425	0.0001
5	2.549	0.1335
6	2.779	0.1053
7	3.070	0.0000
8	3.143	0.0001
9	3.229	0.2516
10	3.245	0.2686
11	3.361	0.0000
12	3.433	0.0003
13	3.478	0.1682
14	3.636	0.0003
15	3.705	0.0034
16	3.905	0.0002
17	3.959	0.0006
18	4.031	0.0006
19	4.047	0.8075
20	4.143	0.1491

G3-4OH-C1

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.827	0.0189
2	2.470	0.0007
3	3.024	0.0015
4	3.234	0.0027
5	3.450	0.0213
6	3.743	0.0584
7	4.050	0.4074
8	4.183	0.0016
9	4.237	0.7938
10	4.378	0.2288
11	4.392	0.0001
12	4.538	0.2036
13	4.559	0.4122
14	4.630	0.0011
15	4.910	0.4303
16	4.945	0.0001
17	5.048	0.0065
18	5.137	0.0016
19	5.157	0.4382
20	5.172	0.2227

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.391	0.0142
2	1.931	0.0003
3	2.344	0.0011
4	2.422	0.0009
5	2.733	0.0149
6	3.067	0.0225
7	3.255	0.0427
8	3.434	0.2173
9	3.439	0.0019
10	3.490	0.0678
11	3.618	0.0006
12	3.650	0.0075
13	3.766	0.1314
14	3.835	0.0014
15	4.037	0.0001
16	4.141	0.9144
17	4.183	0.0000
18	4.255	0.0002
19	4.259	0.3972
20	4.286	0.1614

G3-4OH-C2

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.387	0.0073
2	1.427	0.1281
3	2.952	1.2188
4	3.245	0.3481
5	3.277	0.0000
6	3.421	0.0000
7	3.449	0.0009
8	3.616	0.1712
9	3.838	0.0000
10	3.889	0.0000
11	4.035	0.0156
12	4.048	0.0000
13	4.213	0.0036
14	4.323	0.0000
15	4.333	0.0022
16	4.391	0.0000
17	4.410	0.0762
18	4.439	0.0016
19	4.677	0.0040
20	4.680	0.0002

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.628	0.1129
2	1.714	0.0137
3	2.855	0.0000
4	2.876	1.0984
5	2.951	0.0938
6	2.956	0.0010
7	3.039	0.0000
8	3.174	0.3441
9	3.279	0.0000
10	3.404	0.0000
11	3.442	0.0284
12	3.497	0.0021
13	3.612	0.0000
14	3.788	0.0000
15	3.847	0.0070
16	3.857	0.0000
17	3.901	0.0176
18	3.901	0.0604
19	3.999	0.0065
20	4.024	0.0024

G3-14OH

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	2.845	0.2520
2	2.873	0.0498
3	3.563	0.0000
4	3.805	0.0000
5	3.836	0.0000
6	3.937	1.6545
7	4.085	0.5447
8	4.221	0.0000
9	4.332	0.0000
10	4.409	0.0000
11	4.497	0.1211
12	4.580	0.0000
13	4.626	0.0000
14	4.801	0.0003
15	4.819	0.0000
16	4.880	0.0000
17	4.911	0.0006
18	4.947	0.0151
19	4.973	0.0534
20	4.986	0.0000

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	2.374	0.1764
2	2.488	0.0289
3	3.013	0.0000
4	3.173	0.0000
5	3.260	0.0000
6	3.261	0.9874
7	3.387	0.0000
8	3.435	0.3204
9	3.599	0.0000
10	3.631	0.0000
11	3.688	0.0000
12	3.749	0.0000
13	3.774	0.0000
14	3.838	0.0000
15	3.848	0.0011
16	3.859	0.0047
17	3.992	0.0000
18	4.007	0.0000
19	4.104	0.0000
20	4.187	0.0103

G3-16OH

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.633	0.0084
2	2.020	0.0006
3	2.964	0.0023
4	3.090	0.0007
5	3.337	0.2232
6	3.462	0.3226
7	3.681	0.0782
8	3.875	0.6314
9	3.902	0.0009
10	4.146	0.3442
11	4.304	0.0001
12	4.390	0.2718
13	4.413	0.0019
14	4.495	0.7134
15	4.582	0.1569
16	4.672	0.0011
17	4.718	0.0012
18	4.731	0.0221
19	4.800	0.0000
20	4.828	0.0541

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.167	0.0065
2	1.433	0.0002
3	2.246	0.0014
4	2.338	0.0000
5	2.469	0.0714
6	2.728	0.0049
7	3.030	0.4534
8	3.052	0.0222
9	3.142	0.0018
10	3.263	0.0122
11	3.380	0.0004
12	3.493	0.0115
13	3.619	0.0002
14	3.689	0.0004
15	3.702	0.0006
16	3.730	0.1111
17	3.788	0.1688
18	3.869	0.5226
19	3.897	0.0002
20	3.904	0.2517

G4

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	2.672	0.0000
2	2.873	0.0000
3	3.579	1.5787
4	3.579	1.5778
5	3.651	0.0000
6	3.651	0.0000
7	3.655	0.0000
8	3.883	0.0000
9	3.883	0.0000
10	4.085	0.0000
11	4.096	0.0000
12	4.354	0.0001
13	4.355	0.0000
14	4.424	0.2252
15	4.424	0.2224
16	4.438	0.0000
17	4.460	0.0128
18	4.461	0.0158
19	4.764	0.0000
20	4.764	0.0000

Level of theory: TD-DFT B3LYP /6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	2.229	0.0000
2	2.373	0.0000
3	2.878	0.9297
4	2.878	0.9297
5	3.079	0.0000
6	3.079	0.0000
7	3.084	0.0000
8	3.167	0.0000
9	3.202	0.0000
10	3.254	0.0000
11	3.254	0.0000
12	3.518	0.0000
13	3.518	0.0000
14	3.570	0.0000
15	3.681	0.0000
16	3.681	0.0000
17	3.688	0.0000
18	3.689	0.0000
19	3.714	0.0108
20	3.714	0.0109

G4-2OH-C1

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	0.569	0.1536
2	0.644	0.0122
3	1.640	0.0990
4	1.659	0.0003
5	2.209	0.0000
6	2.243	0.0239
7	2.732	0.0000
8	2.936	0.5408
9	2.955	0.1242
10	3.016	0.0000
11	3.050	0.1944
12	3.098	0.0000
13	3.147	0.0001
14	3.267	0.0059
15	3.546	0.0000
16	3.579	0.0873
17	3.633	0.0017
18	3.710	0.0364
19	3.719	0.0000
20	3.755	0.0000

Level of theory: TD-DFT B3LYP /6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	0.805	0.0926
2	0.892	0.0202
3	1.501	0.0015
4	1.748	0.0887
5	1.850	0.0000
6	2.037	0.0279
7	2.151	0.0000
8	2.554	0.0000
9	2.577	0.0377
10	2.584	0.4462
11	2.615	0.0653
12	2.641	0.0103
13	2.708	0.0000
14	2.752	0.0028
15	2.962	0.0002
16	3.029	0.2317
17	3.076	0.0011
18	3.160	0.0000
19	3.168	0.0000
20	3.249	0.1750

G4-2OH-C2

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	2.432	0.1220
2	3.016	0.0005
3	3.194	0.1450
4	3.239	0.1089
5	3.604	0.2106
6	3.622	0.0906
7	3.678	0.1235
8	3.710	0.2060
9	3.804	0.0319
10	3.902	0.3036
11	3.976	0.0584
12	4.129	0.4897
13	4.160	0.4352
14	4.187	0.2180
15	4.301	0.5122
16	4.339	0.3238
17	4.441	0.0686
18	4.510	0.1246
19	4.564	0.0191
20	4.668	0.0132

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.877	0.0807
2	2.224	0.0144
3	2.471	0.0162
4	2.557	0.1384
5	2.610	0.0560
6	2.744	0.0895
7	2.960	0.1338
8	3.018	0.1823
9	3.101	0.1114
10	3.129	0.0108
11	3.224	0.0372
12	3.234	0.1179
13	3.248	0.0293
14	3.317	0.2099
15	3.385	0.0767
16	3.411	0.0366
17	3.464	0.0017
18	3.510	0.1301
19	3.536	0.0074
20	3.602	0.0934

G4-4OH

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	0.199	0.0010
2	0.454	0.0028
3	0.720	0.0056
4	0.849	0.0726
5	1.246	0.0141
6	1.256	0.1205
7	2.061	0.1087
8	2.298	0.0613
9	2.427	0.1098
10	2.473	0.0255
11	2.644	0.3868
12	2.785	0.0027
13	2.894	0.5610
14	2.919	0.0011
15	3.029	0.2785
16	3.082	0.0566
17	3.169	0.0128
18	3.216	0.1602
19	3.321	0.1715
20	3.375	0.0688

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	0.094	0.0000
2	0.289	0.0032
3	0.584	0.0011
4	0.818	0.0042
5	1.087	0.1324
6	1.356	0.0220
7	1.610	0.0052
8	1.828	0.0643
9	1.855	0.0300
10	1.909	0.0015
11	2.107	0.0312
12	2.305	0.0023
13	2.373	0.0022
14	2.393	0.0000
15	2.396	0.0002
16	2.450	0.6274
17	2.453	0.0030
18	2.483	0.0011
19	2.558	0.0207
20	2.655	0.0247

G4-18OH

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	2.410	0.0000
2	2.605	0.0000
3	3.222	1.6570
4	3.222	1.6571
5	3.265	0.0000
6	3.265	0.0000
7	3.321	0.0000
8	3.587	0.0000
9	3.587	0.0000
10	3.800	0.0000
11	3.804	0.0000
12	3.989	0.0000
13	4.038	0.0633
14	4.038	0.0641
15	4.079	0.0000
16	4.079	0.0000
17	4.092	0.0000
18	4.115	0.0000
19	4.246	0.1186
20	4.246	0.1183

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	1.955	0.0000
2	2.085	0.0000
3	2.519	0.9262
4	2.519	0.9265
5	2.717	0.0000
6	2.717	0.0000
7	2.741	0.0000
8	2.837	0.0000
9	2.841	0.0000
10	2.842	0.0000
11	2.886	0.0000
12	2.949	0.0000
13	2.950	0.0000
14	3.147	0.0000
15	3.307	0.0000
16	3.307	0.0000
17	3.357	0.0443
18	3.358	0.0445
19	3.398	0.0000
20	3.410	0.0000

G4-22OH

Level of theory: TD-DFT ωB97X-D/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	0.182	0.0017
2	0.453	0.0026
3	0.741	0.0659
4	0.825	0.0229
5	1.125	0.0605
6	1.278	0.0626
7	1.915	0.0490
8	2.166	0.3656
9	2.181	0.0654
10	2.336	0.0697
11	2.409	0.0446
12	2.530	0.0966
13	2.583	0.5594
14	2.768	0.1163
15	2.820	0.1350
16	2.880	0.0180
17	3.044	0.0083
18	3.087	0.2601
19	3.183	0.4796
20	3.233	0.0958

Level of theory: TD-DFT B3LYP/6-311+G(d)

State	Energy (eV)	Oscillator Strength
1	0.065	0.0000
2	0.299	0.0013
3	0.620	0.0010
4	0.651	0.0011
5	0.937	0.1070
6	1.180	0.0299
7	1.344	0.0456
8	1.605	0.0192
9	1.613	0.0669
10	1.754	0.0247
11	1.823	0.0271
12	1.920	0.0271
13	2.076	0.0358
14	2.161	0.3486
15	2.211	0.0694
16	2.298	0.0171
17	2.394	0.1402
18	2.440	0.0911
19	2.506	0.0292
20	2.540	0.1283

SECTION S5. GEOMETRY DISTORTION

The distortion from the planarity was measured calculating the deviation of each ring from the plane. Considering the six possible dihedrals (θ_i) formed by the carbons connected by bonds, the deviation from the plane was estimated as the average of the absolute value of the larger of these angles for each ring. The angle θ, reported here, is the average of these values. The shifts of the optical gap (E_{opt}) with respect to the pristine **QD**s (**Gm** with m=2,3,4) are also shown. The number of aromatic rings is also reported (N) and the number of rings (K) which significantly deviates from the plane (θ_i>10°).

Table 4S. Geometry distortion (θ), number of aromatic rings (N) and number of rings with significant deviations from the plane (K). Shift in the optical gaps with respect to the corresponding pristine **QDs**.

GO-QDs	N	K	θ (°)	ΔE(eV)= E_{opt}(**Gm**)-E_{opt}(**GO-QD**) (eV)
G2	7	0	0.	0.
G2-2OH-C1	7	5	20.7	1.767
G2-2OH-C2	7	4	17.9	0.711
G2-4OH-C1	7	5	21.2	2.008
G2-4OH-C2	7	5	20.9	1.868
G2-12OH	7	0	1.3	0.362
G2-14OH	7	5	21.8	1.935
G3	10	*0*	0.	0.
G3-2OH-C1	10	5	16.0	0.460
G3-2OH-C2	10	6	18.6	1.186
G3-2OH-C3	10	4	14.1	1.784
G3-2OH-C4	10	6	17.6	2.059
G3-4OH-C1	10	6	18.3	1.328
G3-4OH-C2	10	8	25.9	1.768
G3-14OH	10	0	1.3	0.310
G3-16OH	10	6	19.2	1.522
G4	19	0	0.	0.
G4-2OH-C1	19	6	9.8	2.103
G4-2OH-C2	19	4	9.1	0.240
G4-4OH	19	12	18.7	2.473
G4-18OH	19	0	1.03	0.262
G4-22OH	19	12	19.9	2.490

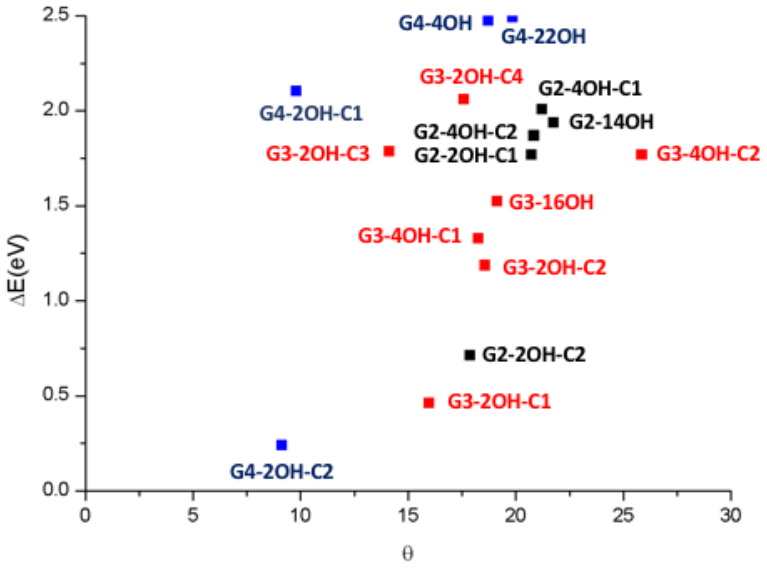


Figure 12S. Relationship between $\Delta E(eV)=E_{opt}$(**Gm**)$-E_{opt}$(**GO-QD**) and the distortion from planarity (angle θ). Black colour was used for **G2**, red for **G3** and blue for **G4** based **GO-QDs**.

SECTION S6. FLUORESCENSE WAVELENGHTS

Emission energies reported in eV and nm and the corresponding oscillator strengths obtained at TDDFT-ωB97X-D/6-311+G(d) level of theory.

Table 5S. Emission energies and oscillator strengths calculated at ωB97X-D/6-311+G(d) level of theory.

GO-QDs	E(eV)	Emission Wavelength (nm)	Osc. Str.
G2	3.35	371	0.000
G2-2OH-C1	1.72	721	0.018
G2-2OH-C2	2.07	599	0.104
G2-4OH-C1	1.43	867	0.060
G2-4OH-C2	1.53	810	0.097
G2-12OH	3.01	412	0.000
G2-14OH	1.55	800	0.027
G3	2.79	444	0.273
G3-2OH-C1	2.23	556	0.097
G3-2OH-C2	1.19	1042	0.019
G3-2OH-C3	1.17	1060	0.114
G3-2OH-C4	0.96	1292	0.039
G3-4OH-C1	1.21	1025	0.019
G3-4OH-C2	1.34	925	0.009
G3-14OH	2.49	498	0.303
G3-16OH	1.04	1192	0.011
G4	2.50	496	0.000
G4-2OH-C1	0.52	2431	0.144
G4-2OH-C2	2.01	617	0.161
G4-18OH	2.24	554	0.000

REFERENCES

1 R. Rieger, M. Kastler, V. Enkelmann and K. Müllen, *Chemistry*, 2008, **14**, 6322–5.