A Ratiometric Near-Infrared Fluorescent Probe Based on a Novel Reactive Cyanine Platform for Mitochondrial pH Detection

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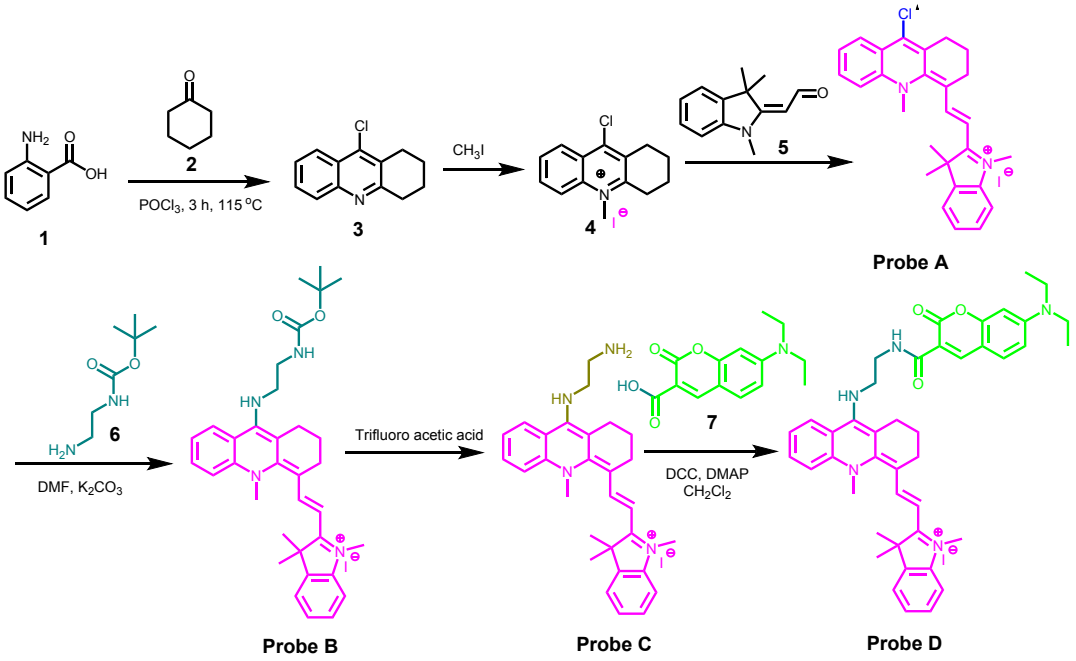
Synthetic route to near-infrared rhodamine dyes A, B, and C.



1.1 Synthesis of 9-chloro-1,2,3,4-tetrahydroacridine (compound 3).

2-aminobenzoic acid (580 mg, 2.7 mmol) was added to a solution of $POCl_3$ (6 mL) containing cyclohexanone (500 mg, 2.46 mmol). The suspension was stirred at 105 °C overnight under an argon atmosphere, allowed to cool down to room temperature, and poured into 200 mL ice-cold water. The pH was adjusted to 7.0 by adding saturated $NaHCO_3$. When the mixture was extracted with dichloromethane (2x100 mL), the organic layer was washed with a saturated NaCl solution. After the organic layer was collected, the solvent was evaporated under reduced pressure. The resulting yellow solid was further purified by column chromatography (packed with silica gel, particle size, 0.063-0.2 mm Merck) and eluted with a mixture of ethyl acetate and dichloromethane at a ratio of 1 to 1 to yield compound **3** as a slightly yellow solid. Yield (470 mg, 70%). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.10 (d, J = 7.0 Hz, 1H), 7.93 (d, J = 8.5 Hz, 1H), 7.64 – 7.57 (m, 1H), 7.47 (t, J = 7.0 Hz, 1H), 3.10 – 3.05 (t, 2H), 2.98 – 2.92 (t, 2H), 1.93 – 1.86 (t, 4H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 159.56, 146.77, 141.55, 129.37, 128.97, 128.76, 126.59, 125.51, 123.81, 34.46, 27.78, 22.96.

Synthesis of compound 4

CH3I (3.9 g, 27.5 mmol) was slowly added to sulfone containing compound **3** (1 g, 4.6 mmol) dissolved into 6 mL of sulfolane. The mixture was stirred at 50°C for 10 h, generating a yellow precipitate. The precipitate was collected by filtration under vacuum and washed with diethyl ether (2 x 50 mL) to obtain the pure compound 4 as a yellow solid. The obtained compound **4** was used for the next step without further purification.

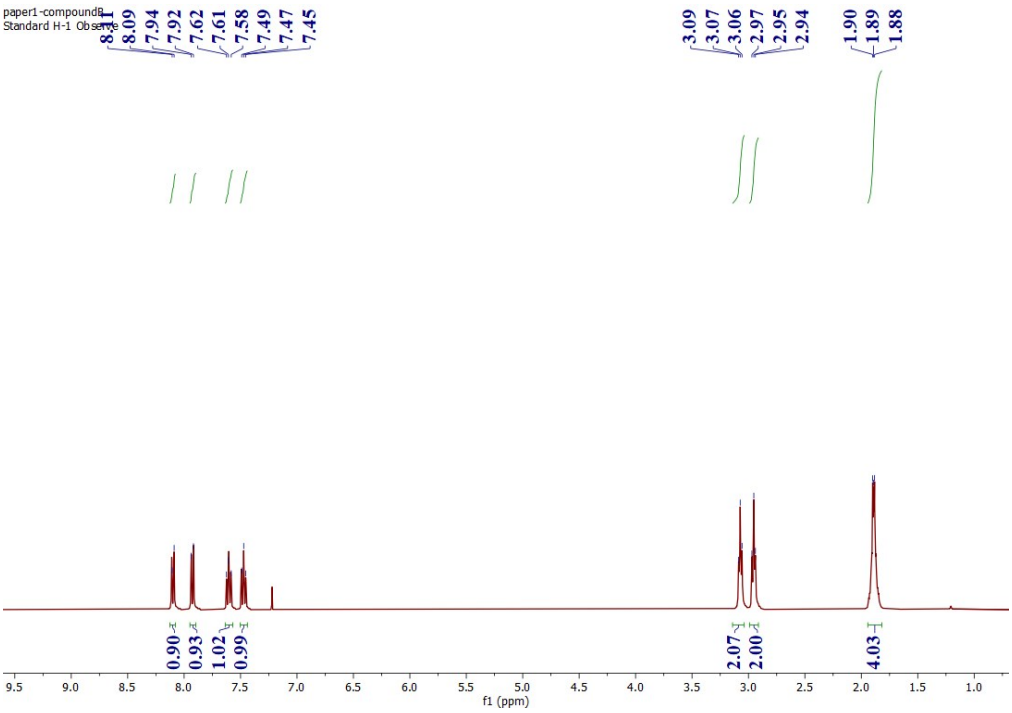


Figure S1. ^{1}H NMR spectrum of compound **3**.

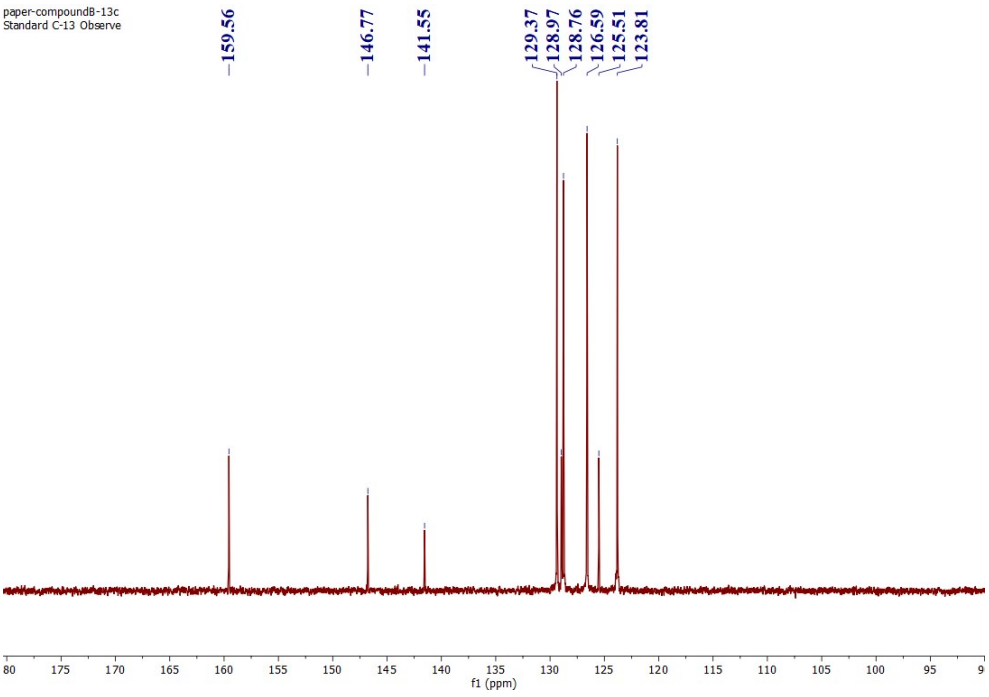


Figure S2. ^{13}C NMR spectrum of compound **3**.

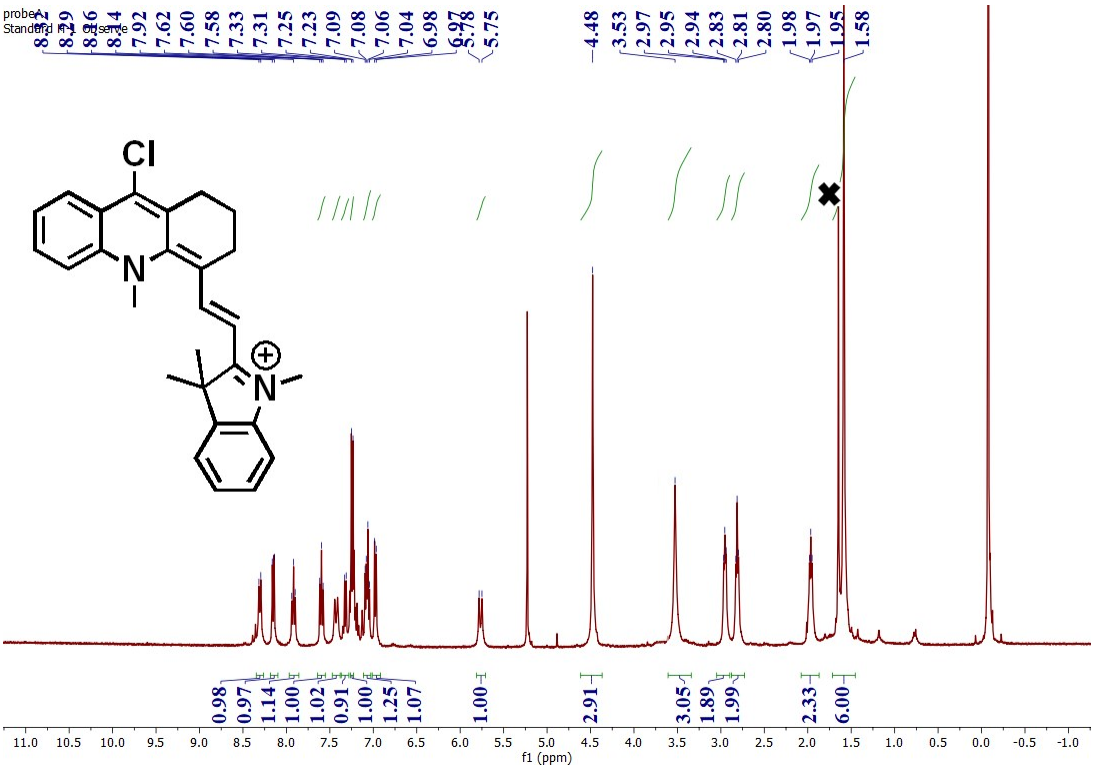


Figure S3. ^{1}H NMR spectrum of probe A in $CDCl_3$ solution.

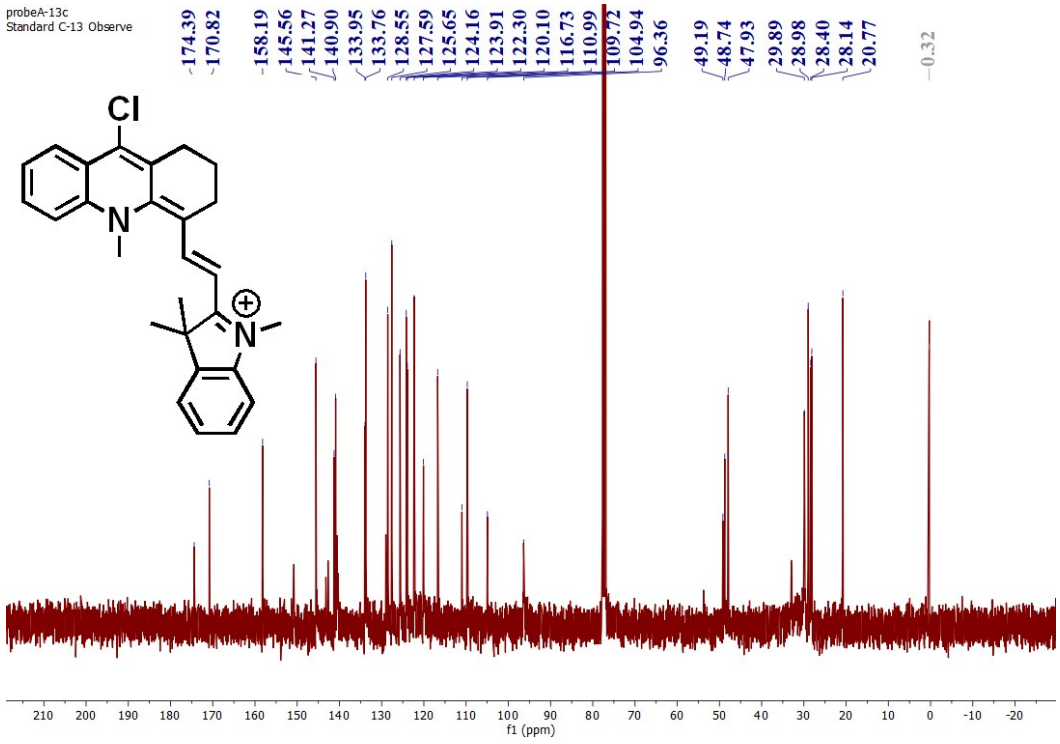


Figure S4. ^{13}C NMR spectrum of probe **A** in $CDCl_3$ solution.

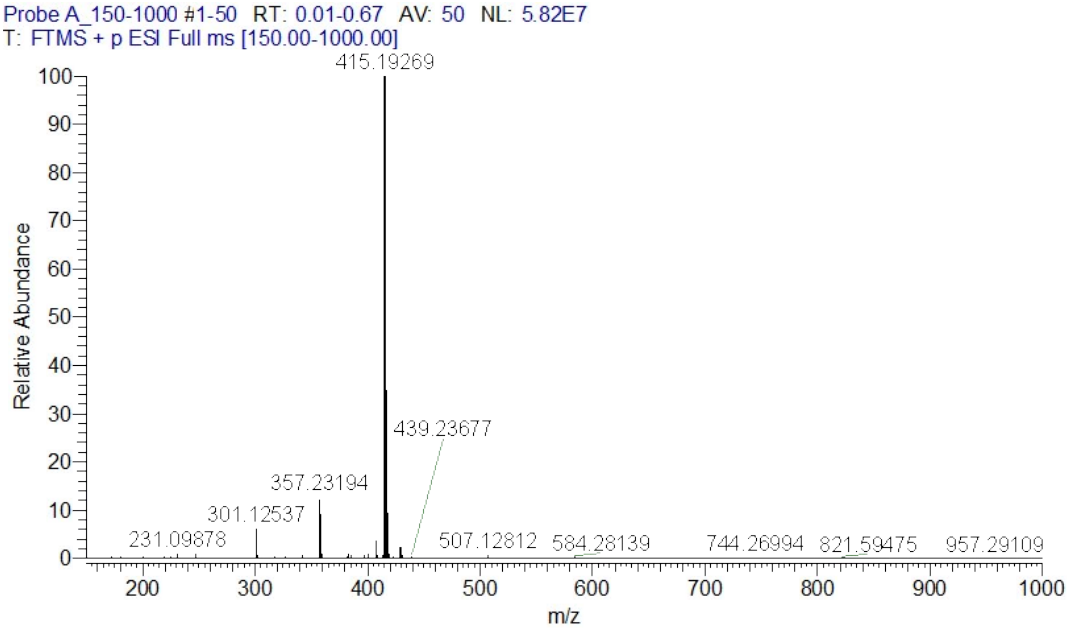


Figure S5. High-resolution ESI mass spectrum of probe **A**.

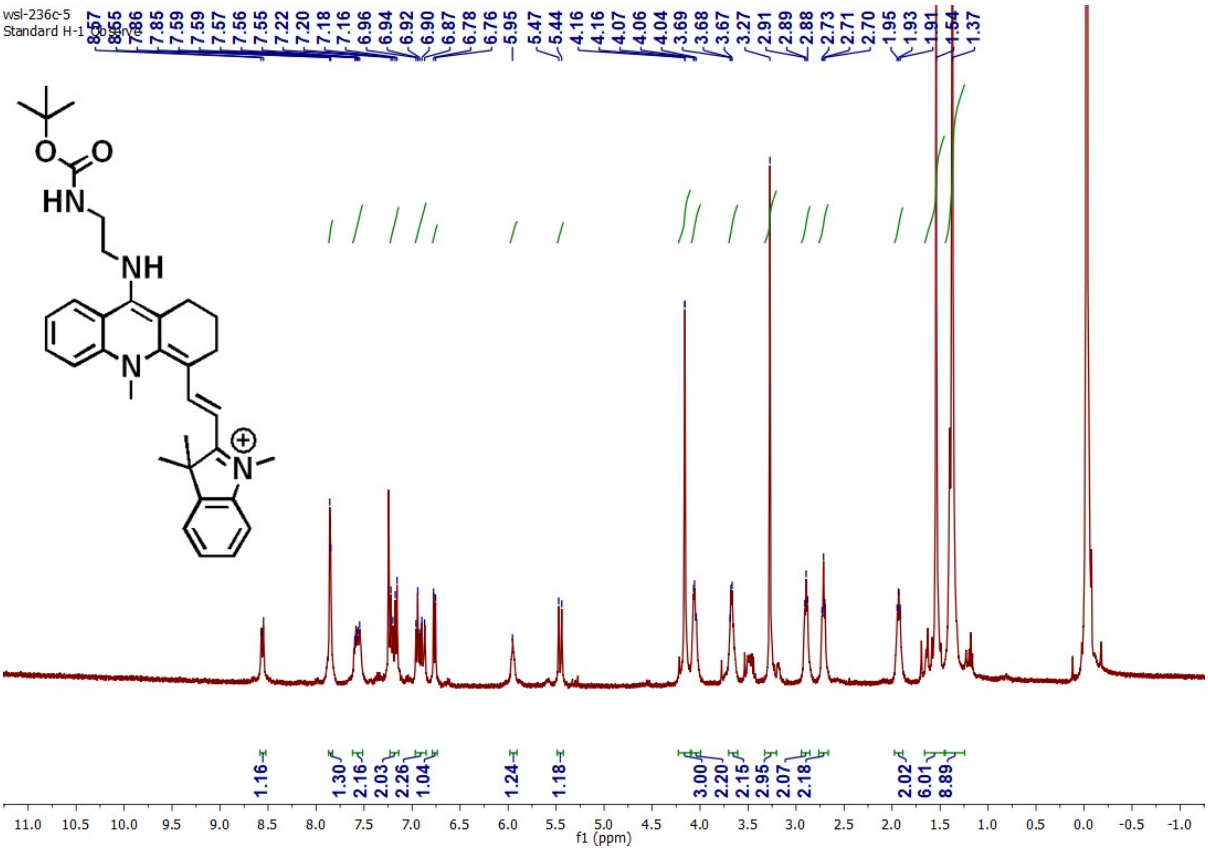


Figure S6. ^{1}H NMR spectrum of probe **B** in $CDCl_3$ solution.

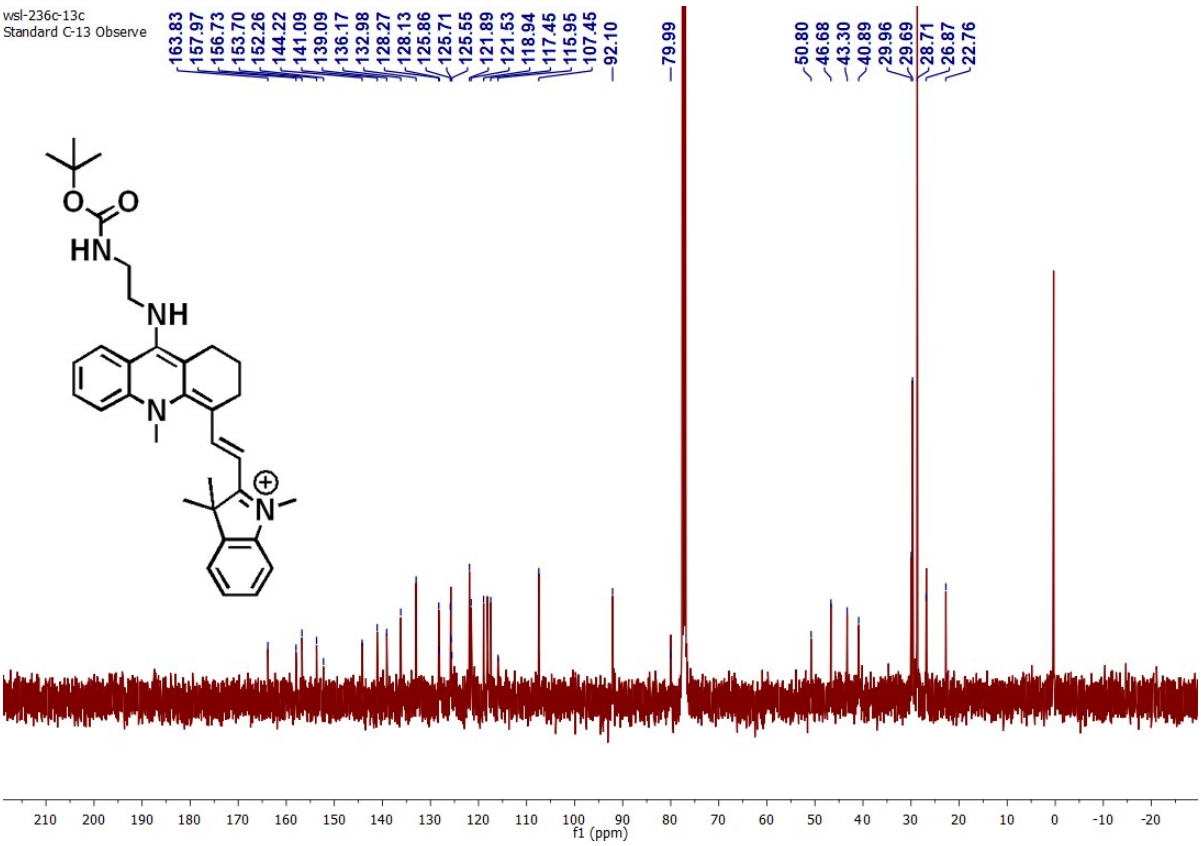


Figure S7. ^{13}C NMR spectrum of probe **B** in $CDCl_3$ solution.

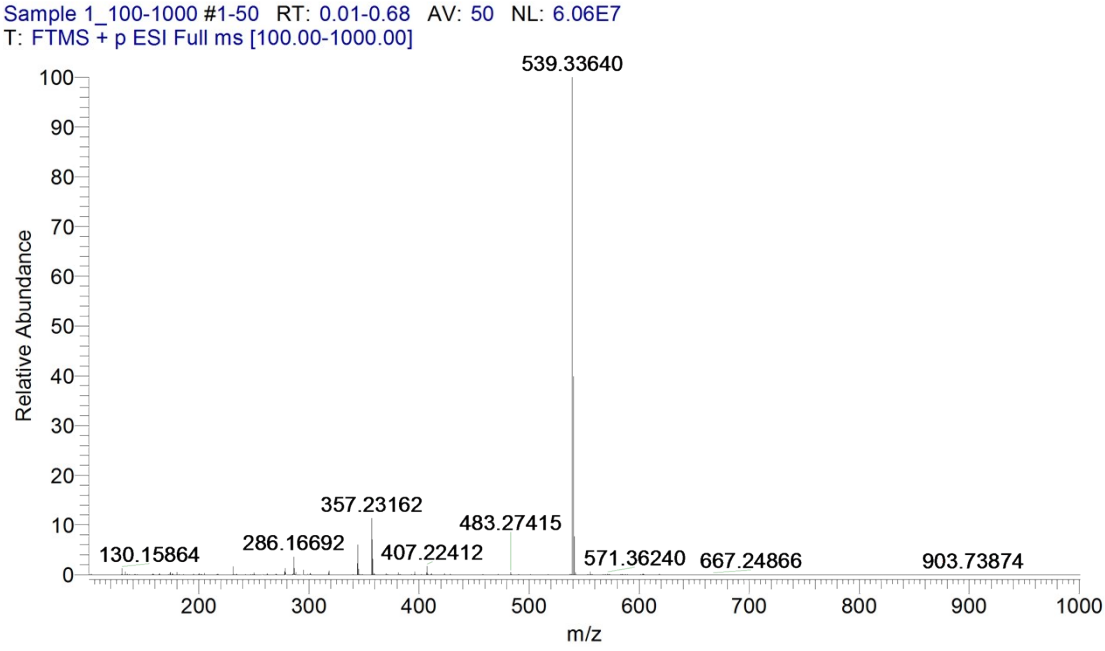


Figure S8. High-resolution ESI mass spectrum of probe **B**.

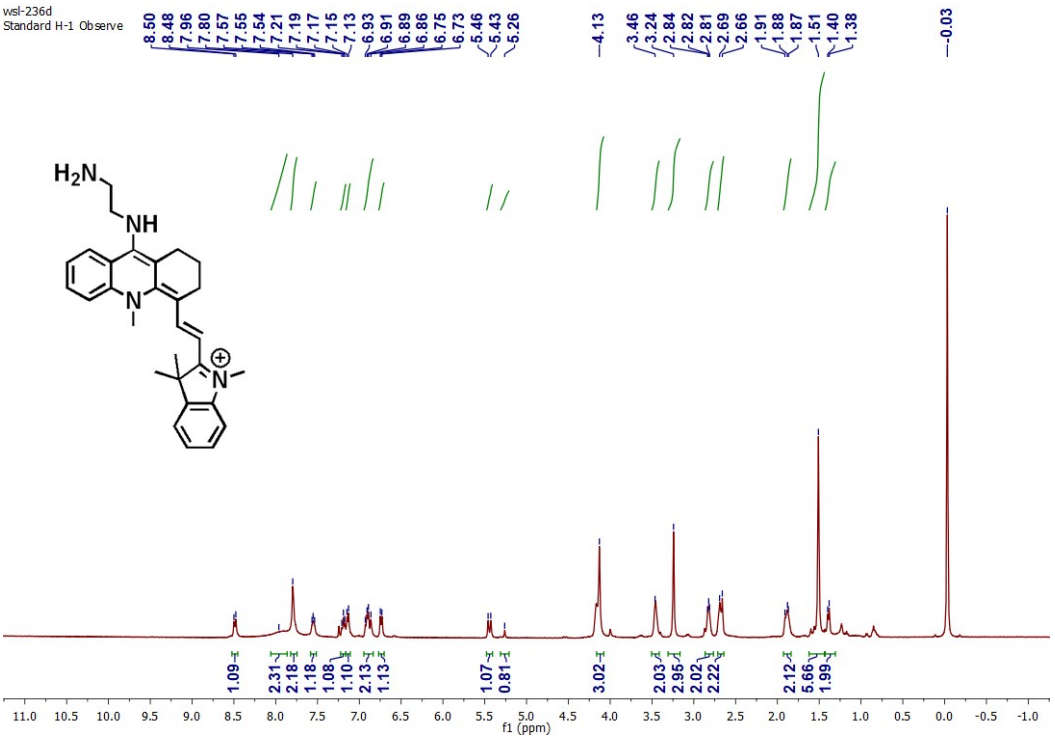


Figure S9. ^{1}H NMR spectrum of probe **C** in $CDCl_3$ solution.

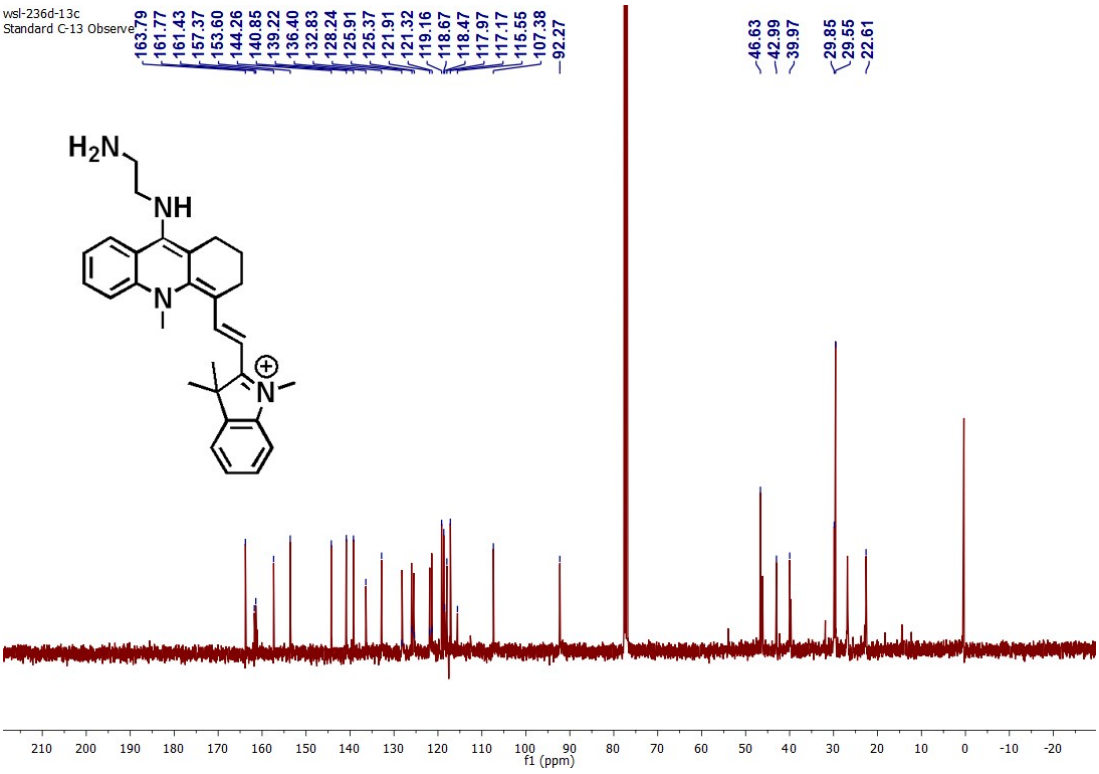


Figure S10. ^{13}C NMR spectrum of probe **C** in $CDCl_3$ solution.

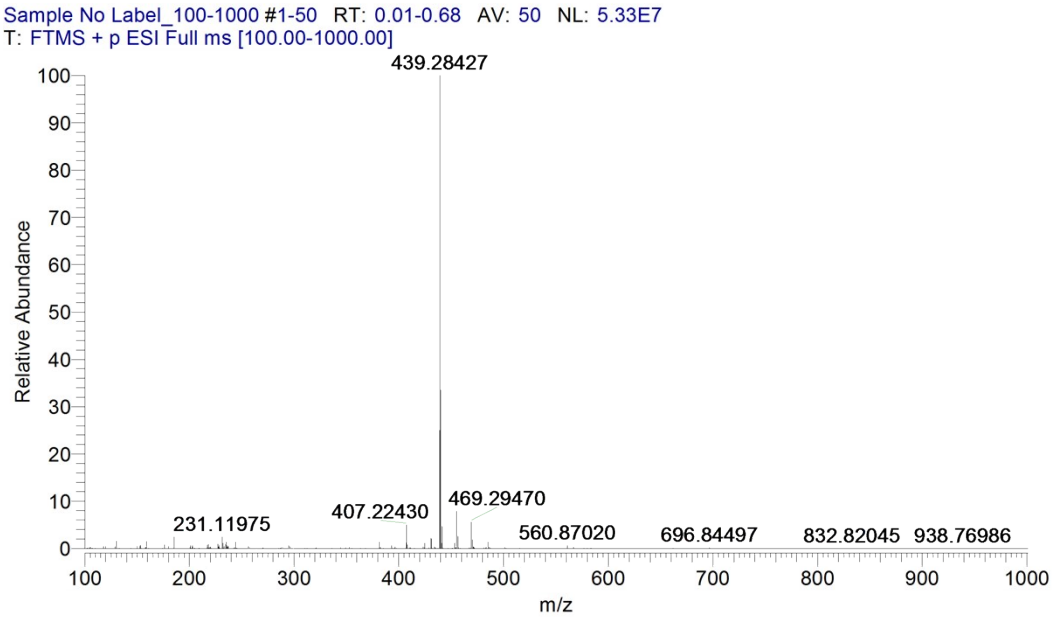


Figure S11. High-resolution ESI mass spectrum of probe **C**.

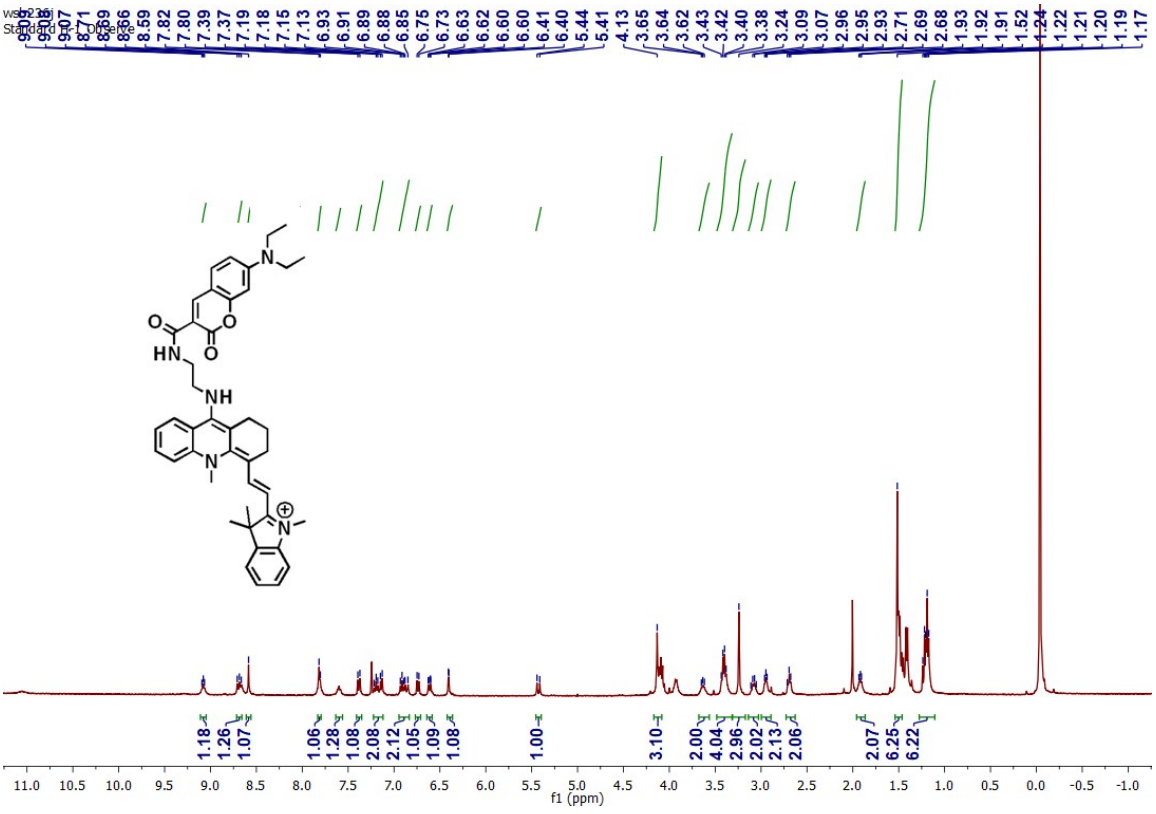


Figure S12. ^{1}H NMR spectrum of probe **D** in $CDCl_3$ solution.

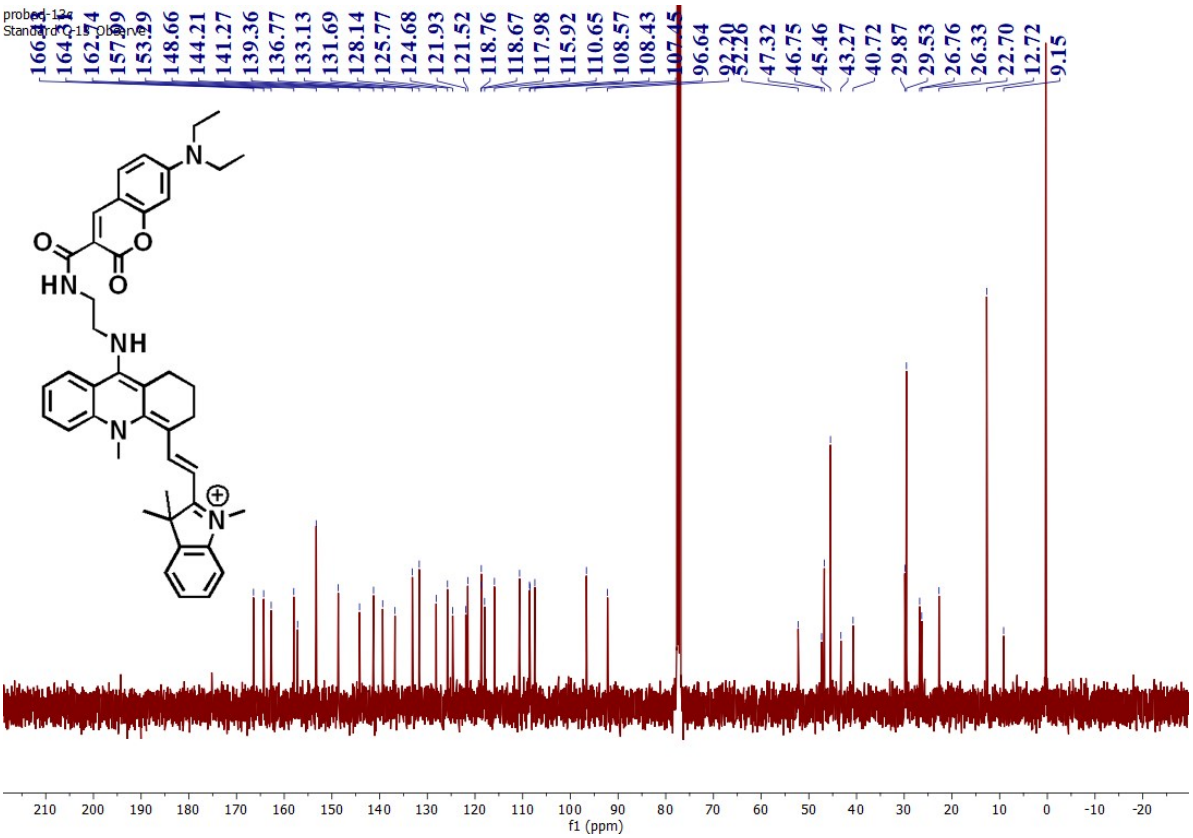


Figure S13. ^{13}C NMR spectrum of probe **D** in $CDCl_3$ solution**.**

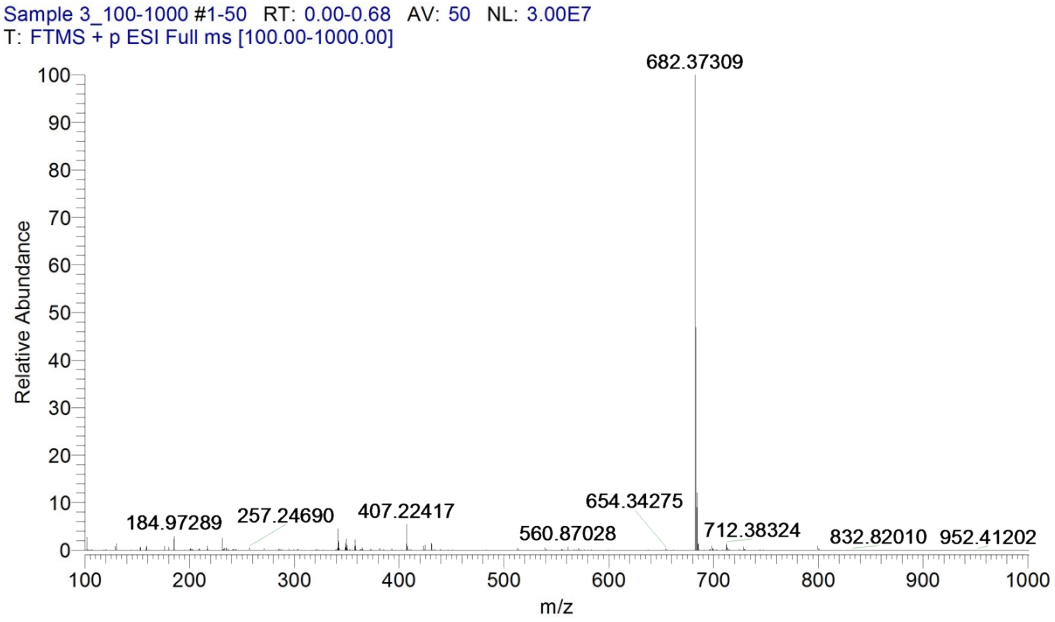


Figure S14. High-resolution ESI mass spectrum of probe **D**.

Calculation of fluorescence quantum yields of the probes.

The fluorescence quantum yields of the fluorescent probes were calculated by employing cresyl violet in methanol with a fluorescent quantum yield of 54% as a reference standard. The UV-Vis absorption spectra of the probes were collected in the range from 300 to 800 nm with increments of 1 nm. For pH detection, we used 0.1 M citrate-phosphate buffer for the pH range from 2.0 to 7.8 and 0.1 M carbonate-bicarbonate buffer for a pH range from 8.0 to 10.0. The pH-responsive fluorescent spectra of probe **A**, **B,** and **C** were collected under 520 nm excitation with increments of 1 nm. The fluorescent measurements for probe **D** were conducted under 400 nm and 520 nm excitation with increments of 1 nm, respectively. The excitation and emission slit widths were set to 5 nm. The fluorescence quantum yields were calculated according to the literature, using the equation below:

$$\Phi_s = \Phi_r(A_r F_s / A_s F_r)/(\eta_s^2/\eta_r^2)$$

Φ is the fluorescence quantum yield, the subscripts 'r' and 's' stand for standard and sample, respectively, 'A' stands for absorbance, and η is the refractive index.

Table S1. Absorption and emission peaks, Stokes shift, molar absorptivity and fluorescence quantum yields of the probes **A**, **B**, **C** and **D**.

Probe	***Solvent***	***λ_{abs} (nm)***	***λ_{em} (nm)***	***Stoke Shift (nm)***	***Φ_f (%)***	***ε ($10^5 M^{-1} cm^{-1}$)***
Probe **A**	Buffer 7.0 containing 0.1 % DMSO	592 nm	660 nm	68 nm	0.53	0.25
Probe **B**	Buffer 7.0 containing 0.1 % DMSO	509 nm	640 nm	131 nm	18.2	0.21
Probe **C**	Buffer 7.0 containing 0.1 % DMSO	512 nm	646 nm	134 nm	10	0.17
Probe **D**	Buffer 7.0 containing 0.1 % DMSO	434 and 535 nm	466 and 647 nm	32 and 112 nm	5.2	0.28/0.12

Fluorescence quantum yields of fluorescence probes are significantly dependents on organic solvent percent concentration in buffer solutions. Probe **A** shows low fluorescence of 2% due to an electron-withdrawing chlorine atom, while probes **B-D** display high fluorescence quantum yields of 60%, 32%, and 35% in aqueous solutions, respectively. However, Probe **A** shows low fluorescence quantum yield of 0.53% in pH 7.0 buffer solution containing 0.1% dimethyl

sulfoxide (DMSO). Probes **B**, **C** and **D** display reduced fluorescence quantum yields of 18.2%, 10% and 4.8% in pH 7.0 buffer solution containing 0.1% DMSO. Fluorescence quenching of the probes in almost pure buffer solvents are due to aggregation-induced quenching effect, especially for probe **D**, which is more hydrophobic.

Determination of the probe pK_a values by fluorometric titration

The pK_a values of fluorescent probes **A**, **B**, **C,** and **D** were calculated using the following equation by fluorometric titration as a function of the pH, which was obtained by using the fluorescence spectra. The expression of the steady-state fluorescence intensity F as a function of the proton concentration has been extended for the case of n: 1 complex between H^+ and a fluorescent dye.

$$F = \frac{F_{\min}[H^+]^n + F_{\max}K_a}{K_a + [H^+]^n}$$

$F_{\min}$ and $F_{\max}$ stand for the fluorescence intensities at maximal and minimal H^+ concentrations, respectively, while n is the apparent stoichiometry of H^+ binding to the probes **A, B,** and **C**. Fluorescence titration data were plotted as a function of the H^+ concentration and a nonlinear fitting is displayed in Figures S17, S19 and S20.

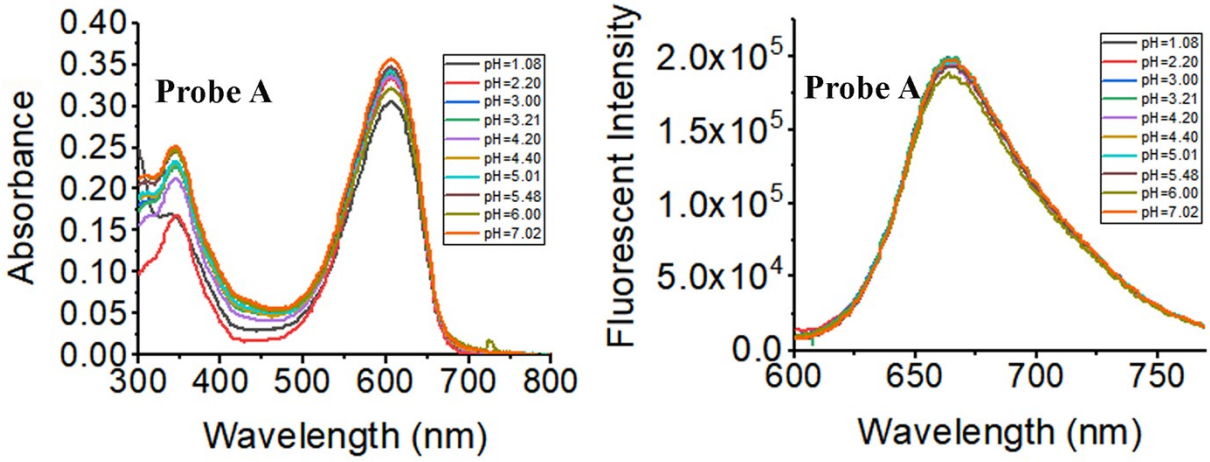


Figure S15. Absorption and fluorescence spectra of 5 μM probe **A** in different pH buffer solutions.

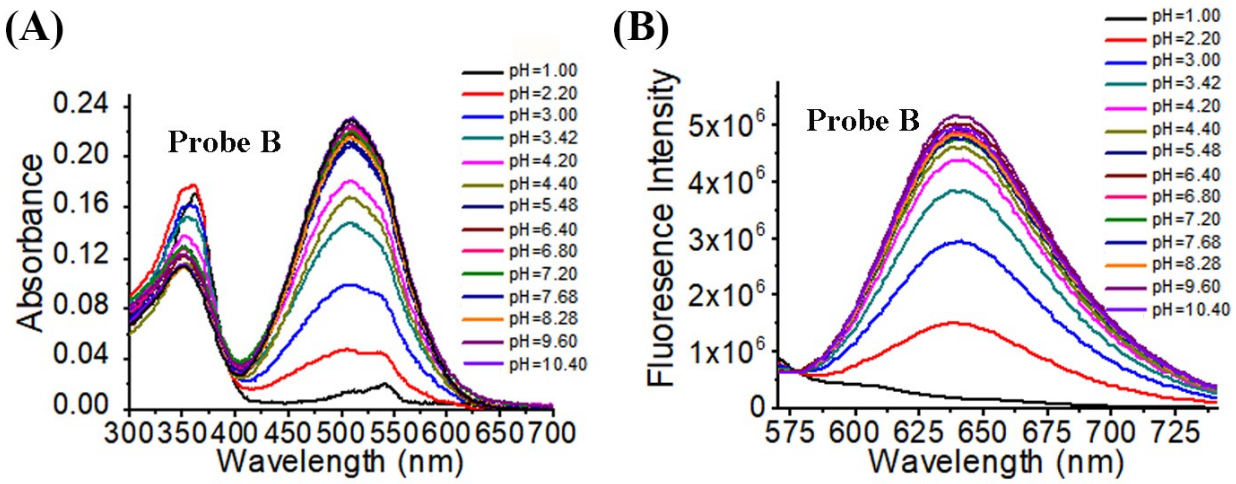


Figure S16. Absorbance (A) and fluorescence (B) responses of 5 μM probe **B** to pH changes in buffer solutions.

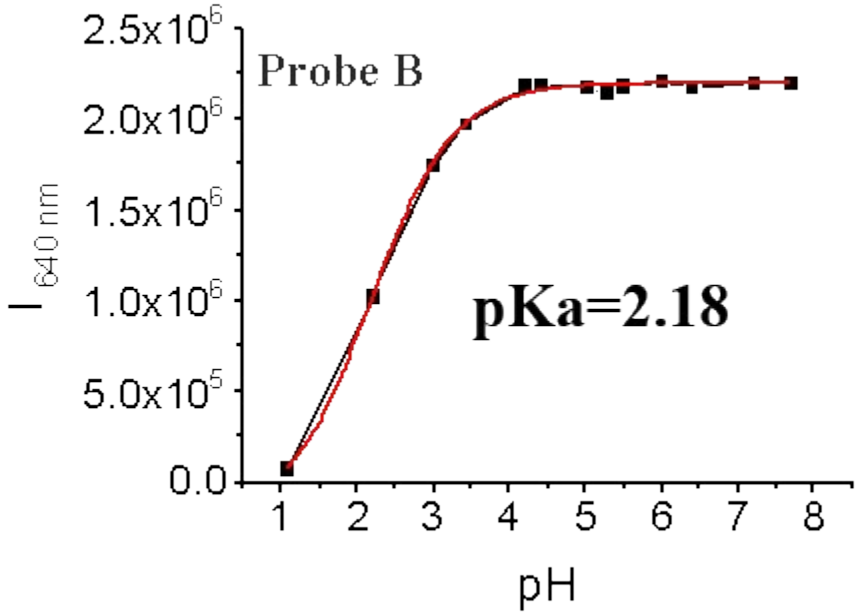


Figure S17. Fluorescence responses of 5 μM probe **B** to pH changes in buffer solutions.

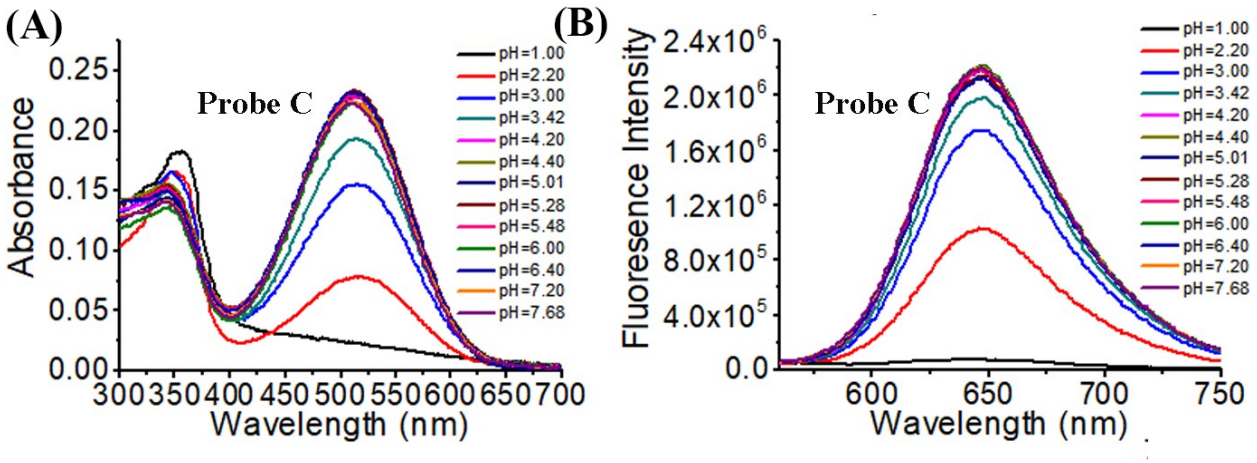


Figure S18. Absorbance (A) and fluorescence (B) responses of 5 μM probe **C** to pH changes in buffer solutions.

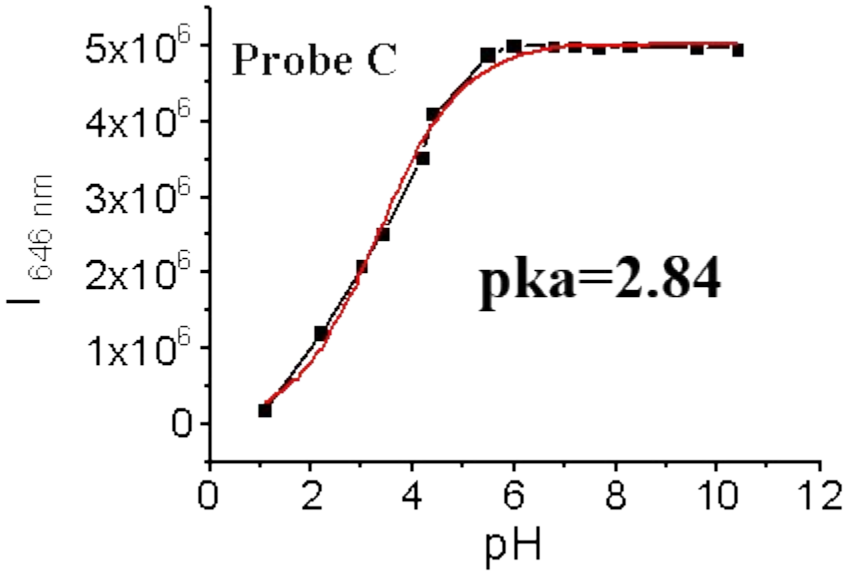


Figure S19. Fluorescence responses of 5 μM probe **C** to pH changes in buffer solutions.

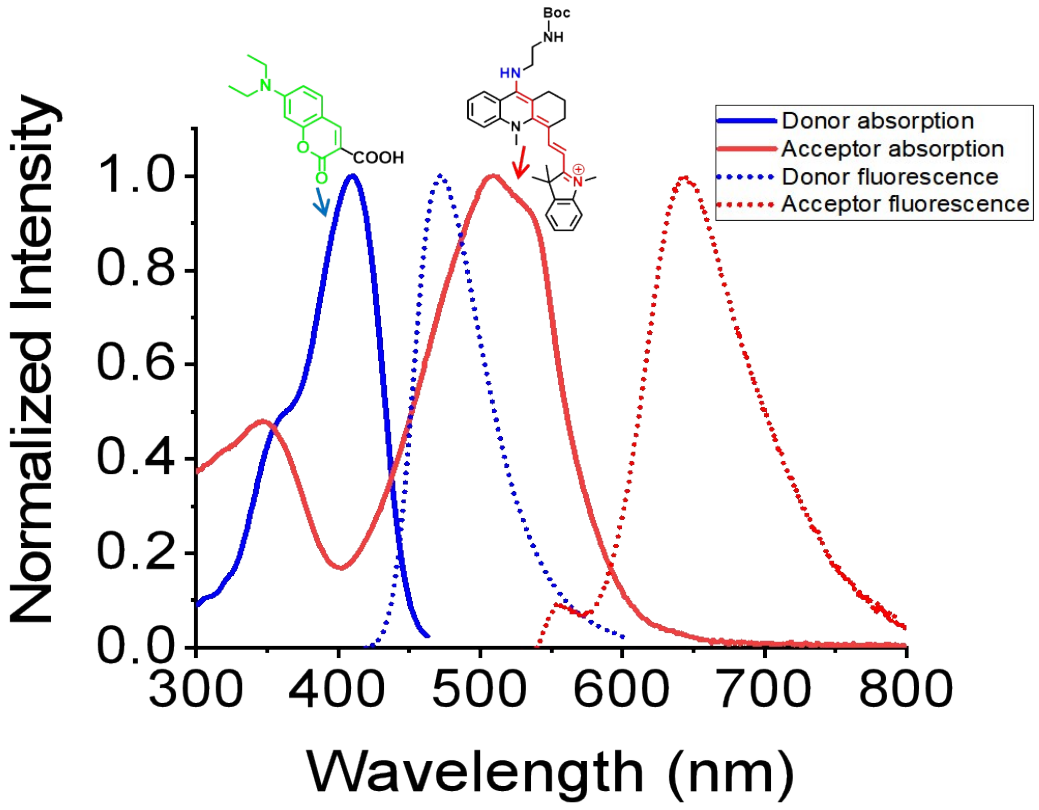


Figure S20. Absorption and emission spectra of 7-(diethylamino)coumarin-3-carboxylic acid and probe **B** in pH 7.0 buffer solution containing 1% dimethyl sulfoxide.

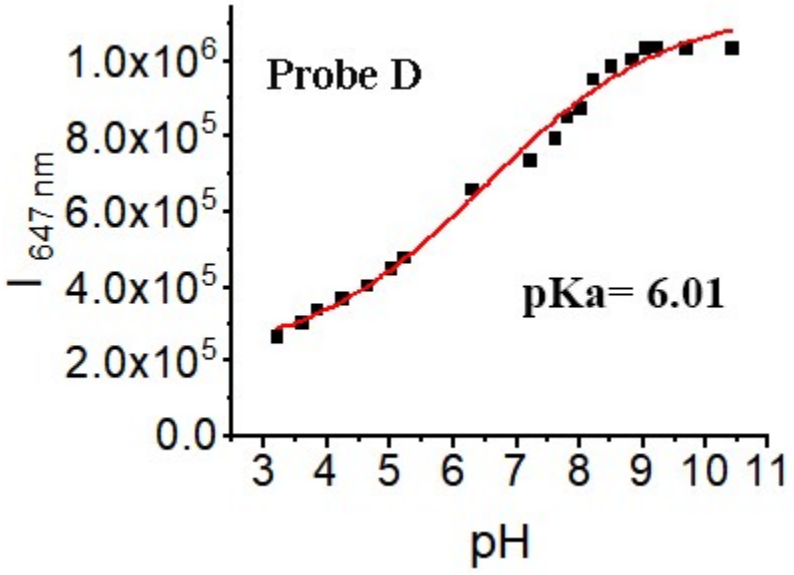


Figure S21. Fluorescence responses of 5 μM probe **D** to pH changes in buffer solutions.

The reversible fluorescence responses of the probes to pH changes

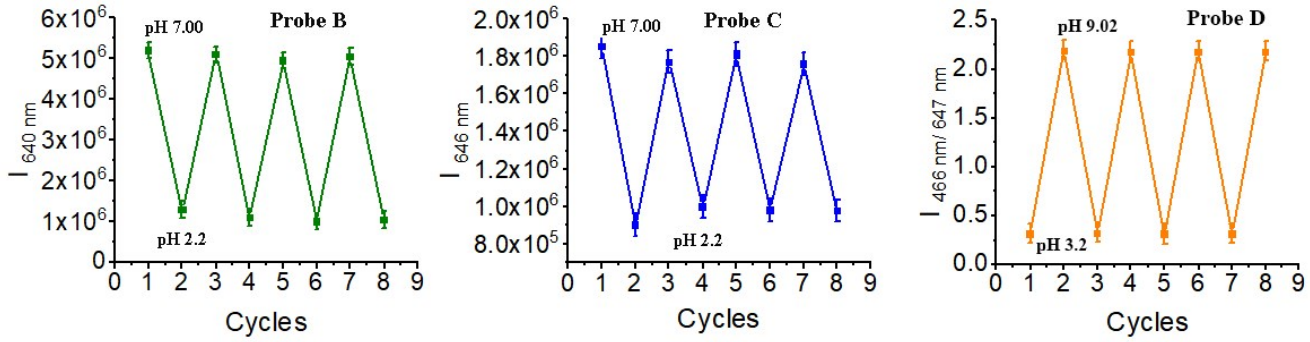


Figure S22. Fluorescence responses of 5 μM probes **B**, **C**, and **D** to pH change from 7.00 to 2.20 under 540 nm excitation.

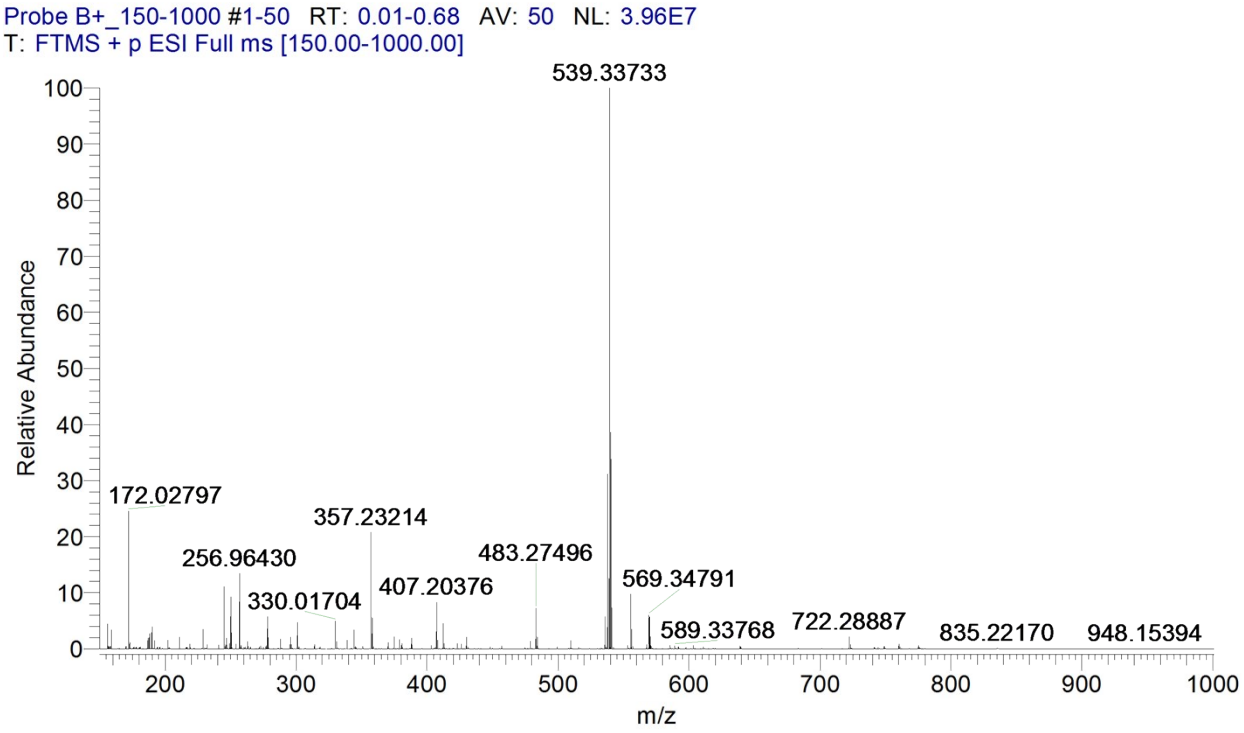


Figure S23. HR-ESI-MS spectrum of probe **B** after treatment with TFA.

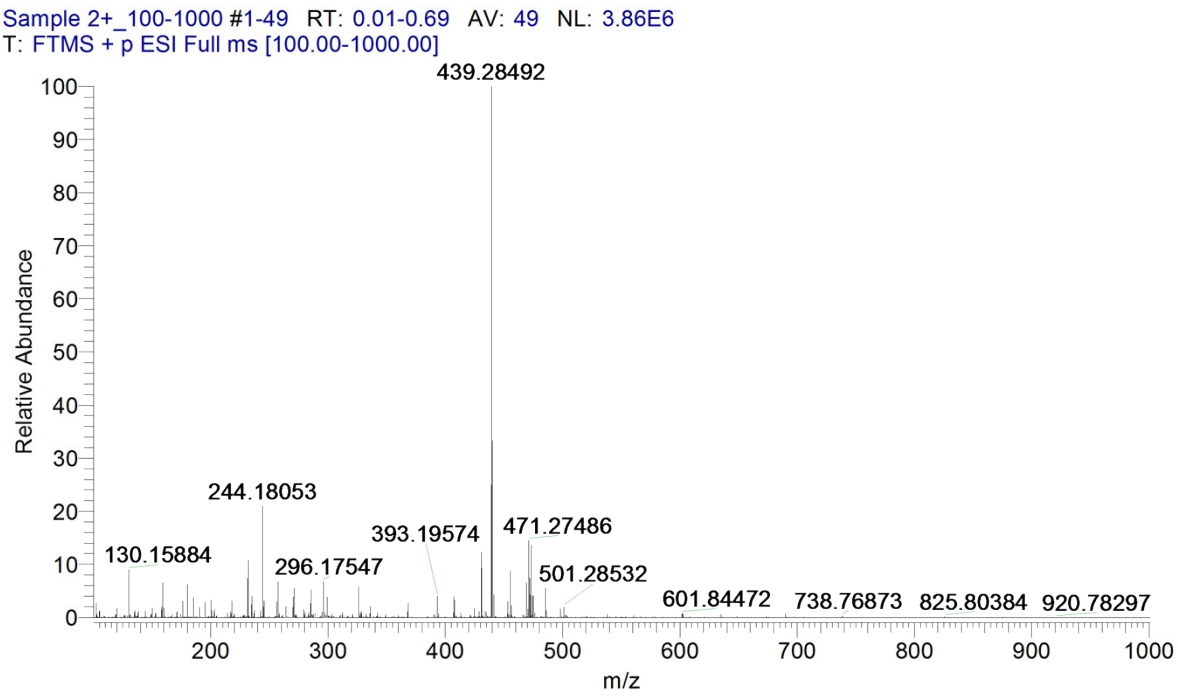


Figure S24. HR-ESI-MS spectrum of probe **C** after treatment with TFA.

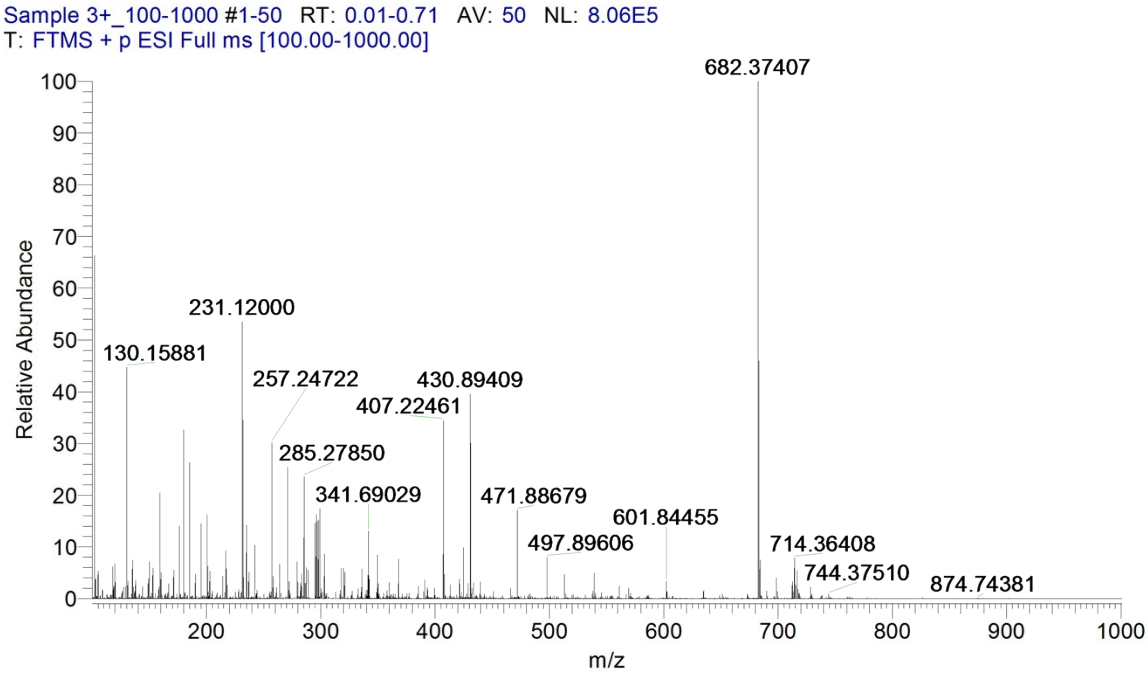


Figure S25. HR-ESI-MS spectrum of probe **D** after treatment with TFA.

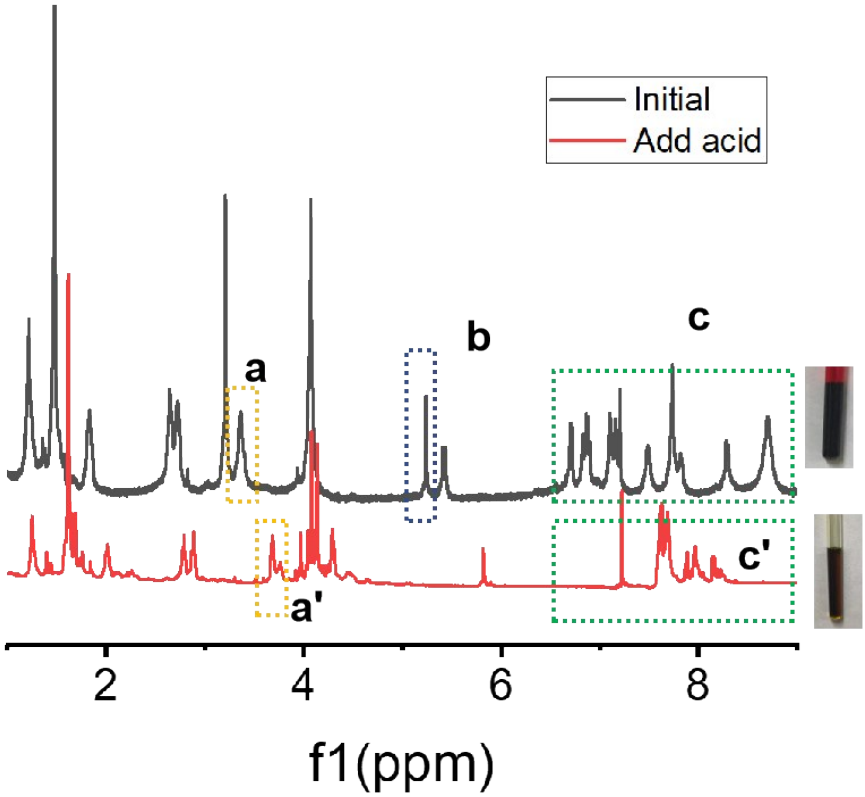


Figure S26. ^{1}H NMR spectrum of probe **C** before (gray line) and after (red line) treatment with TFA.

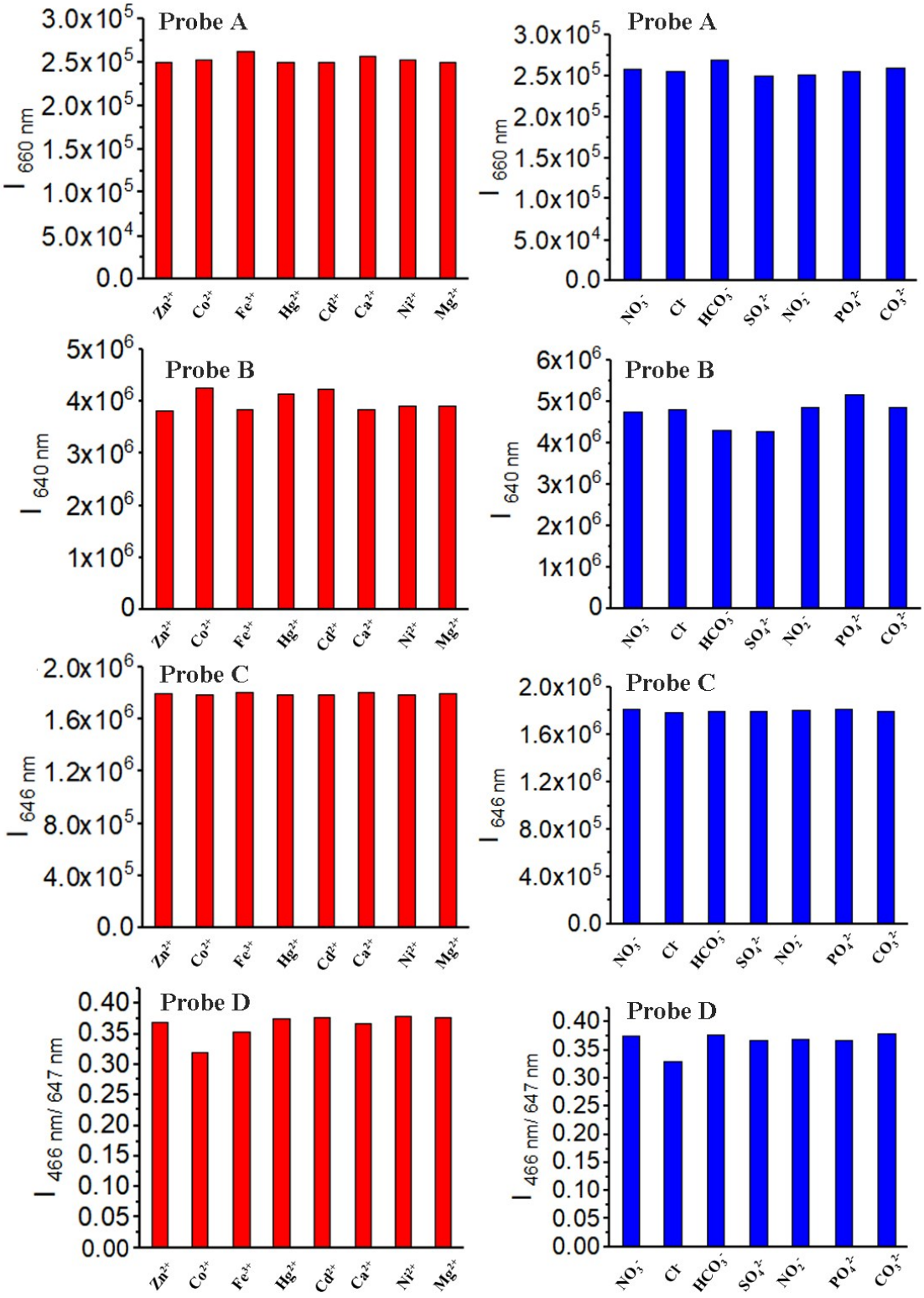


Figure S27. Fluorescent response of 5 μM probes **A**, **B**, **C**, and **D** to different anions and cations in PBS=7.0 buffer.

Photostability of the probes

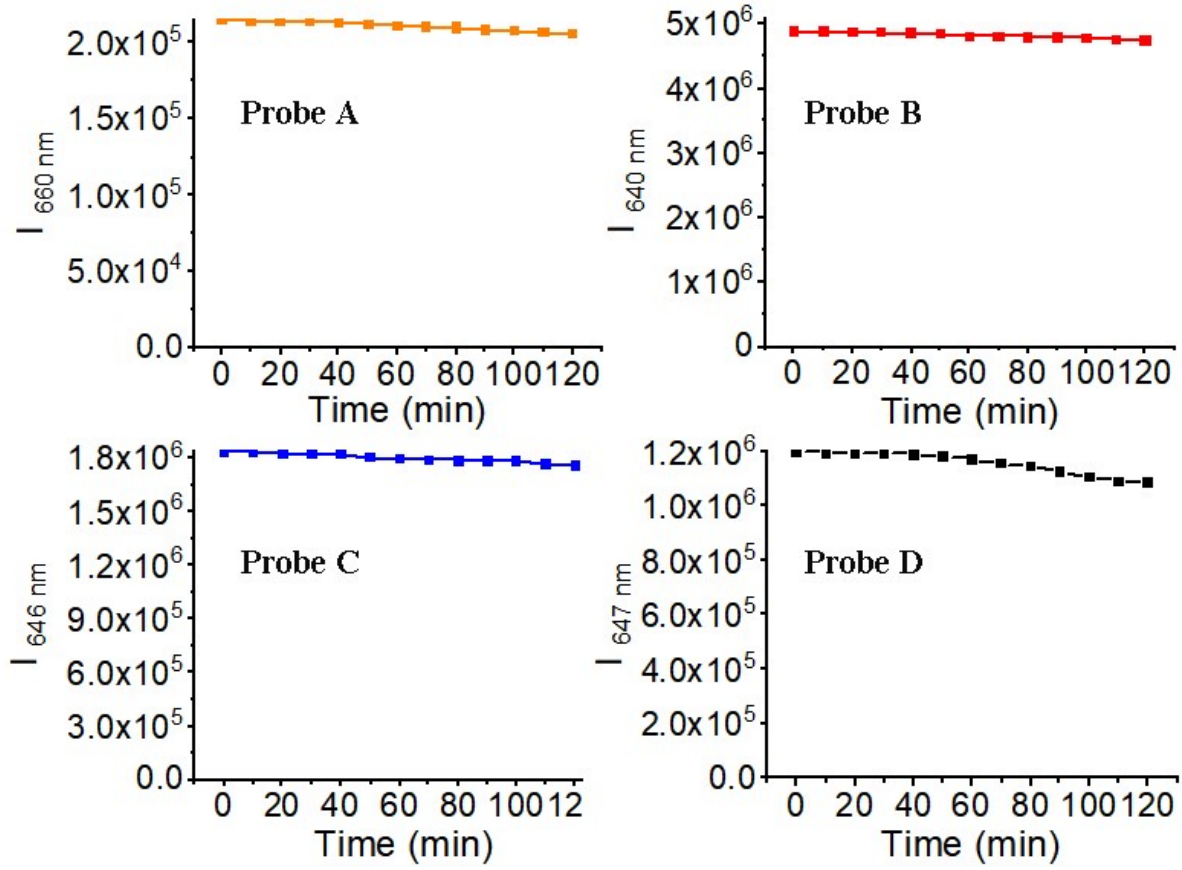


Figure S28. Fluorescence intensity of 5 μM probes **A**, **B**, **C**, and **D** under continuous 520 nm excitation.

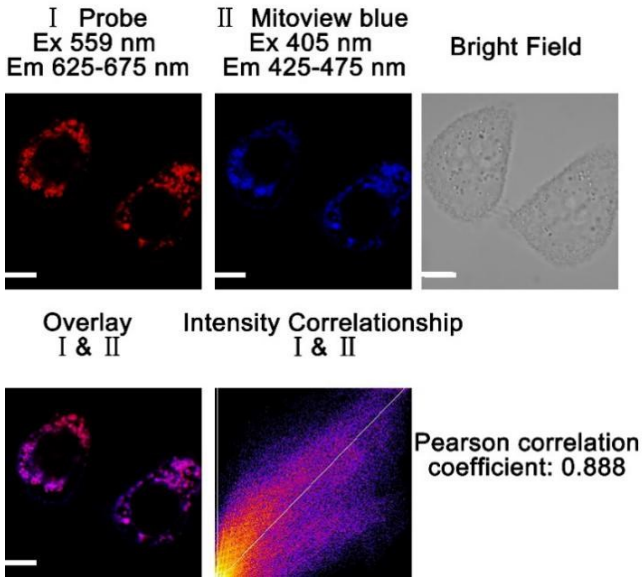


Figure S29. Fluorescence images of HeLa cells incubated with 5 μM probe **A** and 5 μM Mitoview blue for 15 minutes under excitation of 559 nm and 405 nm for probe **A** and Mitoview blue, respectively. Scale bar: 20 μm. The cells were washed with cell culture medium twice after 15-minute incubation of HeLa cells with probe A and Mitoview blue before cellular imaging was conducted, using confocal fluorescence microscope.

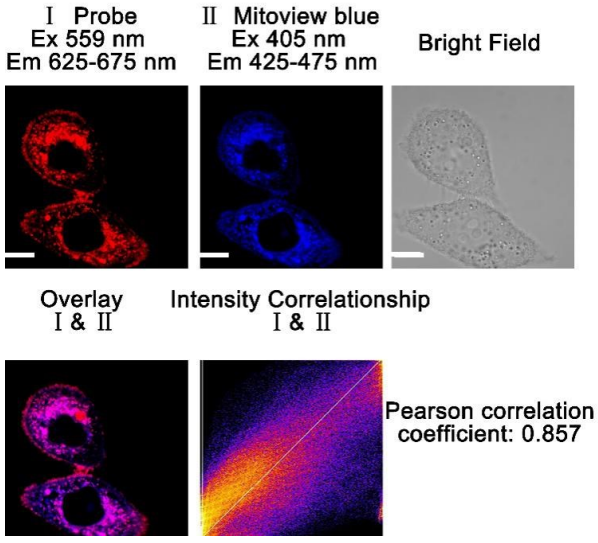


Figure S30. Fluorescence images of 5 μM probe **B** and Mitoview blue in HeLa cells under excitation of 559 nm and 405 nm for probe **B** and Mitoview blue, respectively. Scale bar: 20 μm. The cells were washed with cell culture medium twice after a 15-minute incubation of HeLa cells with probe **B** and Mitoview blue before cellular imaging was conducted, using confocal fluorescence microscope.

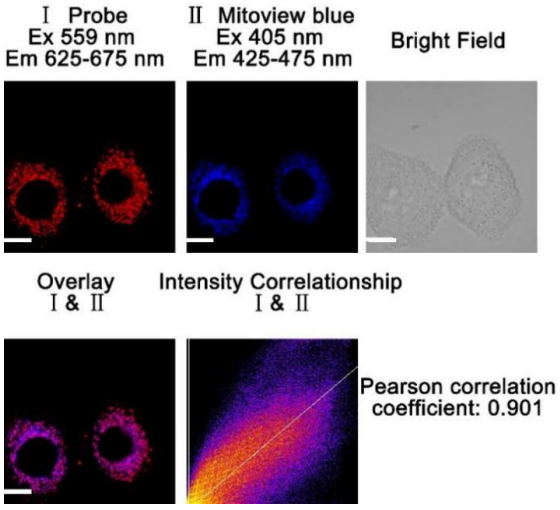


Figure S31. Fluorescence images of 5 μM probe **C** and Mitoview blue in HeLa cells under excitation of 559 nm and 405 nm for probe **C** and Mitoview blue, respectively. Scale bar: 20 μm. The cells were washed with cell culture medium twice after a 15-minute incubation of HeLa cells with probe **C** and Mitoview blue before cellular imaging was conducted, using confocal fluorescence microscope.

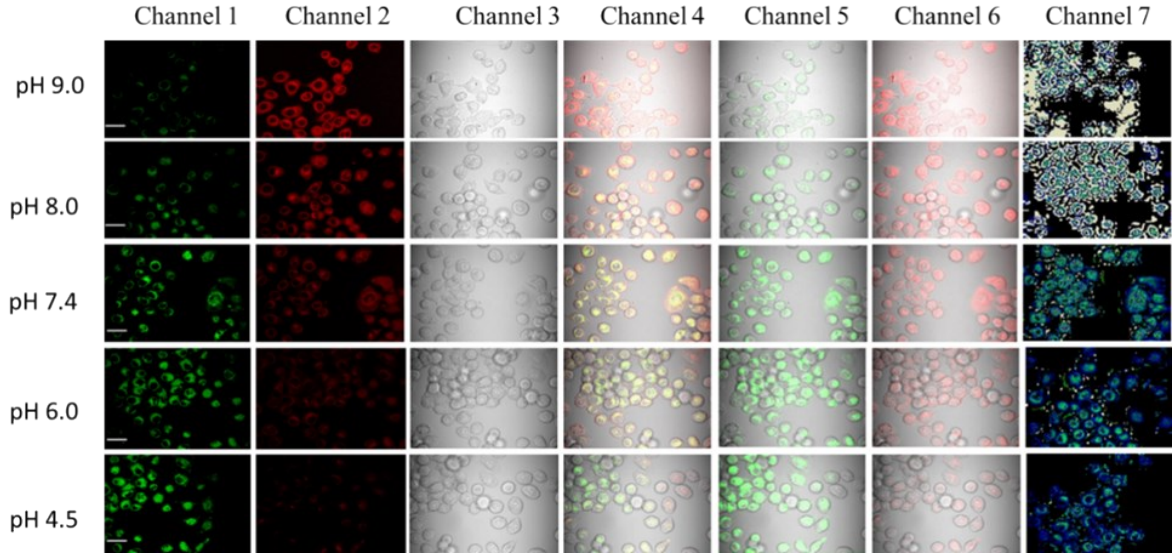


Figure S32. Fluorescence images of HeLa cells incubated with 5 μM probe **D** under different cellular pH conditions with scale bars of 50 μm. HeLa cells were incubated with 5 μM probe **D** for 15 minutes, washed with cell culture medium twice, and then incubated in different pH buffers in the presence of 5 μg/mL nigericin, a K+/H+ ionophore for 20 minutes. Channel 1: the coumarin donor fluorescence collected from 475 to 525 nm at 405 nm excitation. Channel 2: the cyanine acceptor fluorescence collected from 600 to700 nm at 405 nm excitation. Channel 3 bright-field. Channel 4: merged images of channels 1, 2, and 3. Channel 5: merged images of channels 1 and 3. Channel 6: merged images of channels 2 and 3. Channel 7: ratiometric images of channel 1 divided by channel 2, which were acquired with Image-Pro software.

Theoretical Calculations

Methods.

Models of all probes were generated using ChemDraw, with (molecular mechanics) MM2 minimizing the energies of a Chem3d model[1], followed by force field (UFF) calculations in Avogadro.[2] Convergence of the atomic positions in the models were obtained using Gaussian 16[3] and density functional theory (DFT), using the APFD functional[4] and electron basis sets at the 6-311g(d) level in a Polarizable Continuum Model (PCM) of water.[5] Imaginary frequencies were not obtained. Excited states were assessed, using TD-DFT optimizations in a Polarizable Continuum Model (PCM)[5]] in water with the 6-311+g(d) basis set. Results were interpreted using GaussView 6[6] for all data and figures. For the pKa calculations on probes **B-D**, the SMD[7] implicit solvation model in water was conducted. The effect of adding one water molecule directly hydrogen-bonded to the site being protonated/deprotonated was investigated.[8]

Results of the theoretical calculations.

Theoretical calculations for probe A.

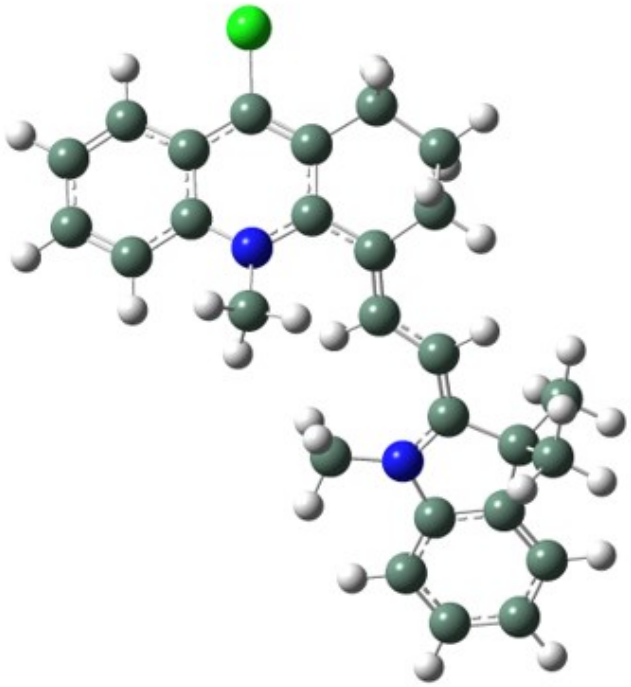


Figure S33**.** GaussView representation of probe **A**.

Table S2. Calculated atomic coordinates for probe **A** in water.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	5.631056	2.858637	-0.50616	30	Cl	5.639127	-1.53469	-1.05066
2	C	4.568939	3.53248	0.109956	31	H	6.501783	3.408708	-0.84679
3	C	3.456947	2.84357	0.547897	32	H	4.609996	4.609145	0.239006
4	C	3.375941	1.45187	0.37263	33	H	2.640448	3.392591	0.997807
5	C	4.439396	0.765731	-0.25811	34	H	6.381271	0.973887	-1.16956
6	C	5.563924	1.495642	-0.68706	35	H	1.337972	-2.93043	1.842599
7	N	2.274355	0.742702	0.823748	36	H	-0.17241	-3.09894	0.971995
8	C	2.067002	-0.56386	0.483712	37	H	1.601063	-4.40874	-0.18343
9	C	3.146856	-1.31109	-0.08911	38	H	1.058964	-3.07658	-1.1996
10	C	4.300866	-0.64598	-0.4067	39	H	3.617691	-3.29861	0.44504
11	C	0.80049	-1.19773	0.693564	40	H	3.471584	-3.05581	-1.27775
12	C	0.8341	-2.6874	0.898625	41	H	-0.23856	0.545754	0.172281
13	C	1.583489	-3.31808	-0.26852	42	H	-1.78124	-2.04294	0.793924
14	C	3.012132	-2.79555	-0.32095	43	H	-4.62221	3.241698	-0.77162
15	C	-0.36353	-0.4818	0.488795	44	H	-7.04557	3.107902	-1.22588
16	C	-1.66984	-0.99524	0.538206	45	H	-8.25382	0.958877	-1.05927
17	C	-2.86706	-0.37371	0.24073	46	H	-7.03557	-1.11822	-0.42588
18	N	-3.1302	0.930068	-0.00649	47	H	-3.79079	-2.5218	1.8199
19	C	-4.48536	1.12082	-0.32605	48	H	-4.5789	-1.00392	2.288786
20	C	-5.15164	-0.09806	-0.23932	49	H	-5.50121	-2.26482	1.45406
21	C	-4.16796	-1.16573	0.151932	50	H	-3.32935	-3.01022	-0.65258
22	C	-5.13853	2.292258	-0.68269	51	H	-5.03982	-2.75929	-1.02396
23	C	-6.5076	2.208211	-0.94273	52	H	-3.80288	-1.8342	-1.89096
24	C	-7.18922	0.996483	-0.85048	53	H	-1.53029	2.086713	-0.74232
25	C	-6.50761	-0.17134	-0.49586	54	H	-1.61915	1.90945	1.035214
26	C	-4.52675	-1.77353	1.515238	55	H	-2.75328	2.95946	0.192844
27	C	-4.07437	-2.25726	-0.92101	56	H	2.105317	1.885975	2.55693
28	C	-2.19955	2.027001	0.11908	57	H	0.777073	2.144648	1.408115
29	C	1.444629	1.399377	1.840502	58	H	0.857139	0.645674	2.354761

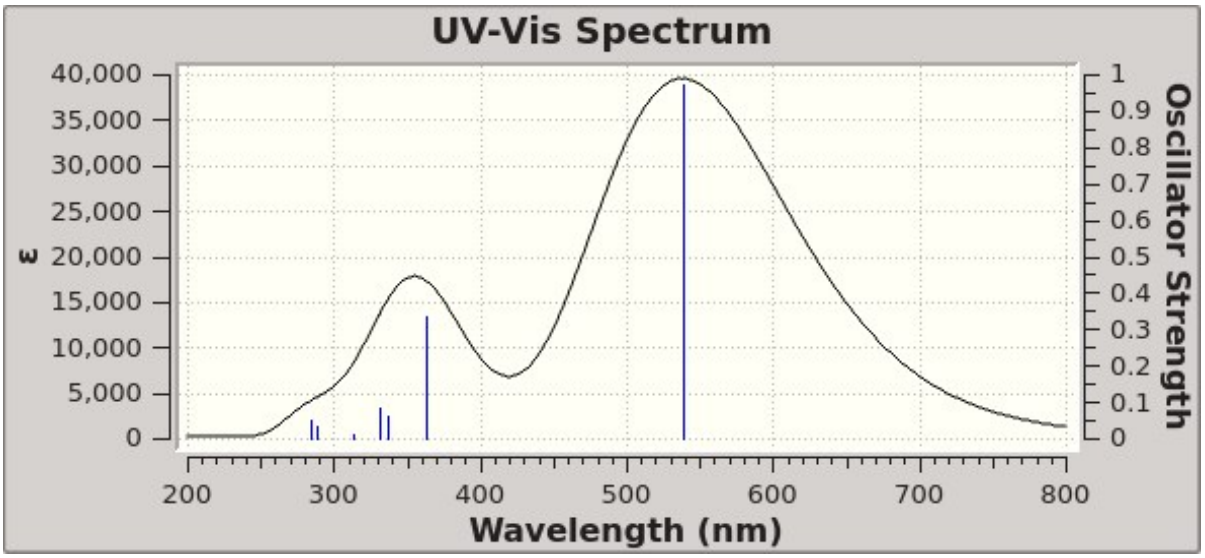


Figure S34. Calculated UV-Vis spectrum for probe **A** in water.

Table S3. Excitation energies and oscillator strengths listing for Probe **A** in water.

Excited State	Nature	E (eV)	λ (nm)	*f*	Orbital transitions	Normalized coefficient
1:	A	2.3030	538.37	0.9728	110 ->111	0.70704
2:	A	3.4109	363.50	0.3379	110 ->112	0.69163
3:	A	3.6758	337.30	0.0631	108 ->111	-0.21913
					109 ->111	0.65445
					110 ->113	-0.10503
4:	A	3.7412	331.40	0.0839	108 ->111	0.65776
					109 ->111	0.22307
5:	A	3.9635	312.82	0.0116	107 ->111	0.68956
					110 ->114	-0.11646
6:	A	4.2879	289.15	0.0351	106 ->111	-0.14388
					110 ->113	0.64771
					110 ->114	-0.18699
7:	A	4.3686	283.81	0.0487	106 ->111	0.62694
					108 ->112	-0.25433
					108 ->113	-0.10809

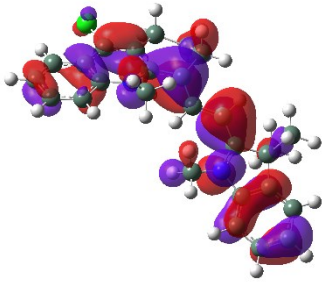
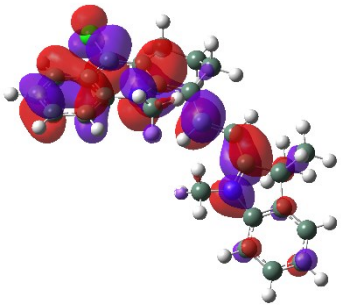
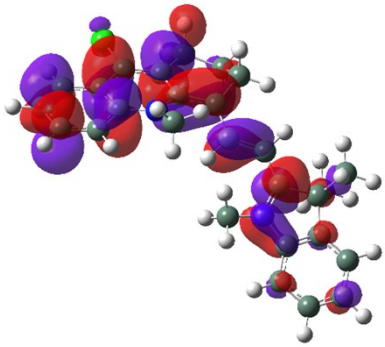
110-HOMO	
111-LUMO	
112-LUMO+1	

Figure S35. Drawings of selected molecular orbitals listed in Table S2.

Theoretical calculations for probe B.

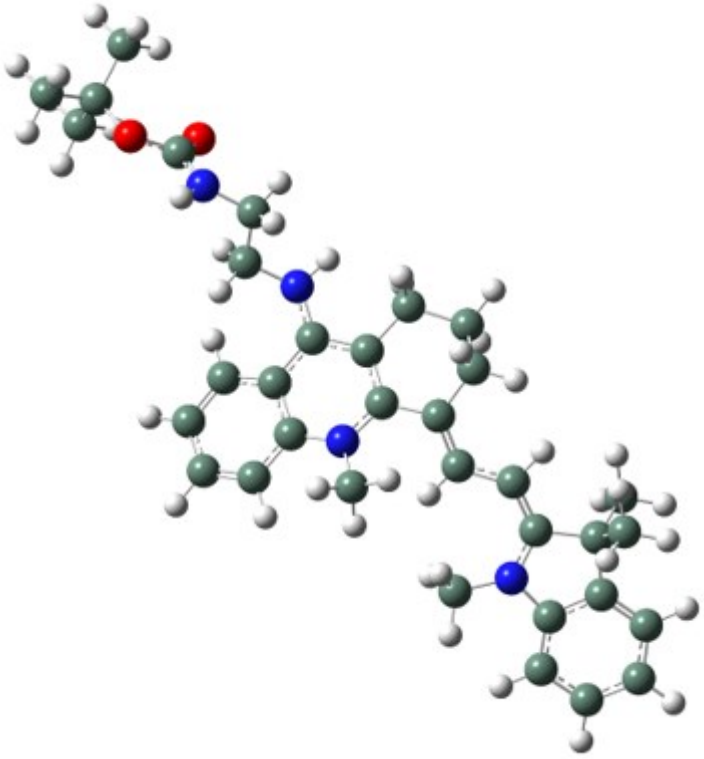


Figure S36. GaussView representation of probe **B.**

Table S4. Calculated atomic coordinates for probe **B** in water**.**

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	2.407113	3.717675	-0.61701	41	H	3.136747	4.372828	-1.08098
2	C	1.297163	4.253972	0.047815	42	H	1.172683	5.329958	0.117722
3	C	0.328953	3.427526	0.578087	43	H	-0.54938	3.867045	1.032919
4	C	0.456186	2.030633	0.473446	44	H	3.396357	1.942658	-1.24448
5	C	1.617205	1.479331	-0.11192	45	H	-0.99527	-2.54147	1.914253
6	C	2.559633	2.351575	-0.69322	46	H	-2.46972	-2.93737	1.05257
7	N	-0.55342	1.196218	0.913712	47	H	-0.53632	-3.99981	-0.10398
8	C	-0.56995	-0.12716	0.554508	48	H	-1.23903	-2.7494	-1.12642
9	C	0.586828	-0.72532	0.036682	49	H	1.315273	-2.66105	0.523794
10	C	1.744969	0.045892	-0.14927	50	H	1.12583	-2.39316	-1.20209
11	C	-1.77084	-0.92274	0.737687	51	H	-3.00554	0.664229	0.180097
12	C	-1.53194	-2.38779	0.968841	52	H	-4.2072	-2.13739	0.646094
13	C	-0.69573	-2.92065	-0.19065	53	H	-7.67008	2.812125	-0.74747
14	C	0.651849	-2.20986	-0.22914	54	H	-10.0395	2.393438	-1.30746
15	C	-2.99806	-0.37738	0.477364	55	H	-10.9627	0.10025	-1.29681
16	C	-4.23707	-1.06979	0.456326	56	H	-9.50383	-1.8324	-0.7138
17	C	-5.48931	-0.60052	0.158001	57	H	-6.18883	-2.91115	1.613121
18	N	-5.92079	0.680694	-0.02945	58	H	-7.1819	-1.52842	2.107528
19	C	-7.27122	0.707641	-0.39708	59	H	-7.90635	-2.86093	1.192482
20	C	-7.7775	-0.59056	-0.3994	60	H	-5.59182	-3.23477	-0.85851
21	C	-6.67887	-1.54502	-0.01475	61	H	-7.30761	-3.18432	-1.28324
22	C	-8.06225	1.801068	-0.72479	62	H	-6.17028	-2.07469	-2.06605
23	C	-9.39944	1.555956	-1.04568	63	H	-4.49705	2.131785	-0.6011
24	C	-9.92028	0.264276	-1.0417	64	H	-4.57892	1.752851	1.142493
25	C	-9.10214	-0.82268	-0.71575	65	H	-5.84814	2.710104	0.385708
26	C	-7.00563	-2.25185	1.307843	66	H	-0.90445	2.292976	2.650897
27	C	-6.41756	-2.56954	-1.12383	67	H	-2.22522	2.395086	1.470079
28	C	-5.16215	1.880073	0.229174	68	H	-1.98337	0.910588	2.408872
29	C	-1.47965	1.733689	1.913082	69	H	2.858645	-1.55254	-0.59691
30	N	2.914651	-0.55794	-0.43668	70	H	4.80496	0.169086	-1.03725
31	C	4.255809	-0.08777	-0.12598	71	H	4.192033	0.802982	0.499338
32	C	5.020464	-1.17891	0.627155	72	H	4.509012	-1.42127	1.561047
33	N	6.367687	-0.77048	0.927707	73	H	5.064965	-2.09067	0.024872
34	C	7.343881	-0.83236	-0.01537	74	H	6.560968	-0.27167	1.78067
35	O	8.476566	-0.30354	0.475094	75	H	11.143	-1.62266	-1.15749
36	O	7.191504	-1.31199	-1.12516	76	H	9.465429	-2.16171	-1.34953
37	C	9.699283	-0.25707	-0.33232	77	H	10.26072	-2.26784	0.234297
38	C	10.1633	-1.66755	-0.67461	78	H	11.6619	0.512499	0.120761
39	C	10.68812	0.415618	0.606477	79	H	10.34076	1.41426	0.883673
40	C	9.484431	0.59338	-1.57851	80	H	10.81576	-0.17169	1.519444

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
81	H	9.095956	1.578559	-1.3055	83	H	8.787283	0.118961	-2.26793
82	H	10.4392	0.736176	-2.09139					

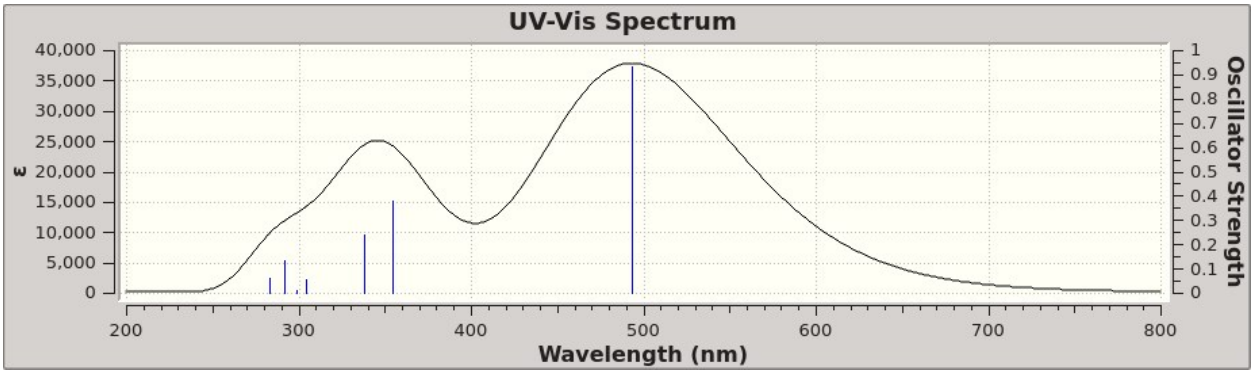


Figure S37. Calculated UV-Vis spectrum for probe **B** in water.

Table S5. Excitation energies and oscillator strengths listing for probe **B** in water.

Excited State	Nature	E (eV)	λ (nm)	*f*	Orbital transitions	Normalized coefficient
1:	A	2.5117	493.63	0.9308	145 ->146	0.70485
2:	A	3.4974	354.50	0.3804	145 ->147	0.69690
3:	A	3.6642	338.37	0.2396	144 ->146	0.69056
4:	A	4.0789	303.96	0.0578	143 ->146	0.68051
					144 ->147	-0.12591
5:	A	4.1533	298.52	0.0113	142 ->146	-0.11620
					145 ->148	0.52148
					145 ->149	-0.43846
6:	A	4.2457	292.02	0.1351	142 ->146	0.14668
					144 ->147	0.13023
					145 ->148	0.43757
					145 ->149	0.50100
7:	A	4.3797	283.09	0.0632	140 ->146	-0.22798
					141 ->146	0.53287
					143 ->146	-0.10832
					144 ->147	-0.34612

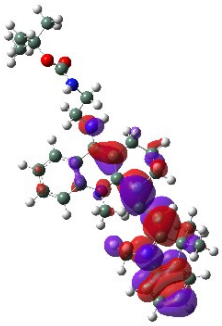
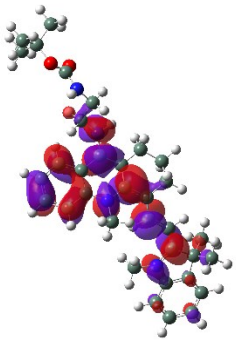
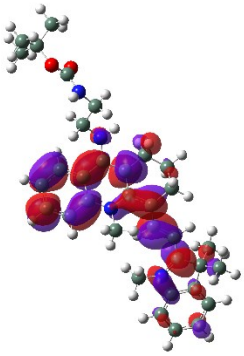
145-HOMO	
146-LUMO	
147-LUMO+1	

Figure S38. Drawings of selected molecular orbitals for probe **B** listed in Table S4.

Theoretical calculations for probe BH^+.

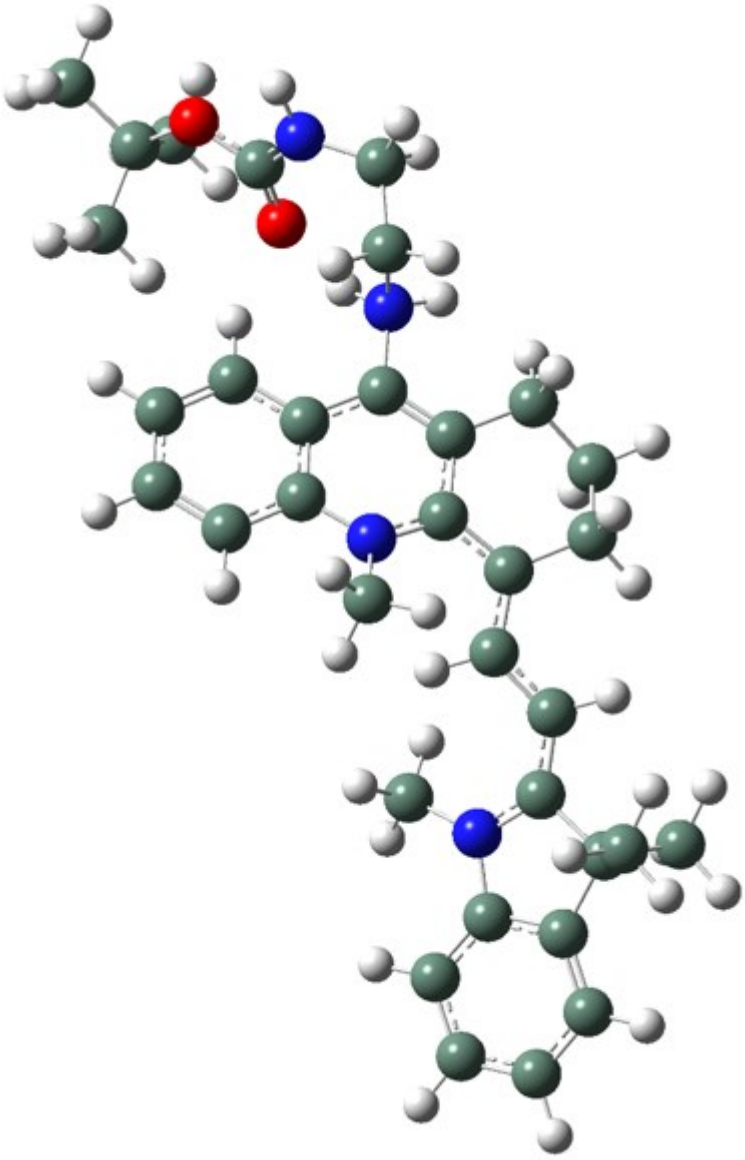


Figure S39. GaussView representation of probe **BH^+**.

Table S6. Calculated atomic coordinates for probe $\mathbf{BH^+}$ in water.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	3.250393	2.511813	1.356922	41	H	4.146021	3.12347	1.346938
2	C	2.073603	2.992191	1.939744	42	H	2.051234	3.982693	2.382115
3	C	0.924593	2.225053	1.945278	43	H	0.019576	2.631884	2.37698
4	C	0.927088	0.950137	1.360058	44	H	4.180959	0.910226	0.31826
5	C	2.114756	0.454623	0.777811	45	H	-1.55706	-3.62395	0.764434
6	C	3.26787	1.257696	0.784366	46	H	-2.56663	-3.21762	-0.61312
7	N	-0.2147	0.164013	1.358048	47	H	-0.48534	-4.37268	-1.3501
8	C	-0.31392	-0.96841	0.587329	48	H	-0.59012	-2.73854	-2.00156
9	C	0.911896	-1.58025	0.131463	49	H	1.050751	-3.67871	0.397395
10	C	2.066117	-0.86986	0.249288	50	H	1.654564	-3.15273	-1.16191
11	C	-1.56692	-1.5333	0.258897	51	H	-2.51091	0.354703	0.247863
12	C	-1.61701	-2.98777	-0.128	52	H	-4.20305	-2.19368	0.006443
13	C	-0.47324	-3.31401	-1.07688	53	H	-6.38186	3.508139	-1.06377
14	C	0.861826	-2.97931	-0.42751	54	H	-8.83815	3.806374	-1.03924
15	C	-2.68694	-0.70912	0.151668	55	H	-10.3421	1.901297	-0.5856
16	C	-3.99361	-1.13256	-0.07294	56	H	-9.40905	-0.36169	-0.14232
17	C	-5.12336	-0.34533	-0.27427	57	H	-7.82705	-2.31447	-1.14014
18	N	-5.20294	0.952651	-0.60781	58	H	-6.6142	-1.58616	-2.20552
19	C	-6.54222	1.387093	-0.63241	59	H	-6.14646	-2.85402	-1.05602
20	C	-7.37488	0.306352	-0.36222	60	H	-7.7764	-1.82307	1.383749
21	C	-6.5279	-0.9134	-0.12958	61	H	-6.08863	-2.34568	1.453845
22	C	-7.03251	2.661214	-0.87647	62	H	-6.5429	-0.73702	2.043878
23	C	-8.41865	2.82218	-0.85508	63	H	-3.69115	2.343128	-0.1402
24	C	-9.26771	1.747161	-0.59704	64	H	-3.33877	1.228207	-1.49047
25	C	-8.74685	0.474636	-0.34677	65	H	-4.48372	2.551482	-1.71355
26	C	-6.7881	-1.98187	-1.20242	66	H	-0.71909	0.687467	3.309912
27	C	-6.74193	-1.48817	1.276173	67	H	-1.8341	1.348819	2.096204
28	C	-4.11164	1.818222	-1.0005	68	H	-1.87988	-0.37725	2.496131
29	C	-1.23025	0.48344	2.368985	69	H	3.154295	-2.23157	-0.80796
30	N	3.329164	-1.48565	-0.13469	70	H	4.000253	-0.84414	-0.62859
31	C	4.081555	-2.06611	1.038861	71	H	4.120931	-1.29523	1.807468
32	C	5.478317	-2.51523	0.633424	72	H	3.490341	-2.90292	1.408554
33	N	6.465494	-1.45614	0.592915	73	H	5.817088	-3.23297	1.379799
34	C	6.457047	-0.4854	-0.34317	74	H	5.450847	-3.05192	-0.32134
35	O	7.580062	0.210978	-0.328	75	H	7.300279	-1.55356	1.148544
36	O	5.498933	-0.27764	-1.09709	76	H	8.009247	1.919926	-3.2309
37	C	7.756064	1.421209	-1.15974	77	H	6.775338	0.68098	-2.95014
38	C	7.753258	1.041564	-2.63302	78	H	8.499088	0.267356	-2.83107
39	C	9.128871	1.905841	-0.72625	79	H	9.402334	2.801133	-1.28866
40	C	6.689637	2.455778	-0.82592	80	H	9.135522	2.151913	0.338432

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
81	H	9.885489	1.139375	-0.91054	83	H	6.958355	3.4077	-1.29012
82	H	6.632658	2.613212	0.254655	84	H	5.708713	2.16253	-1.19776

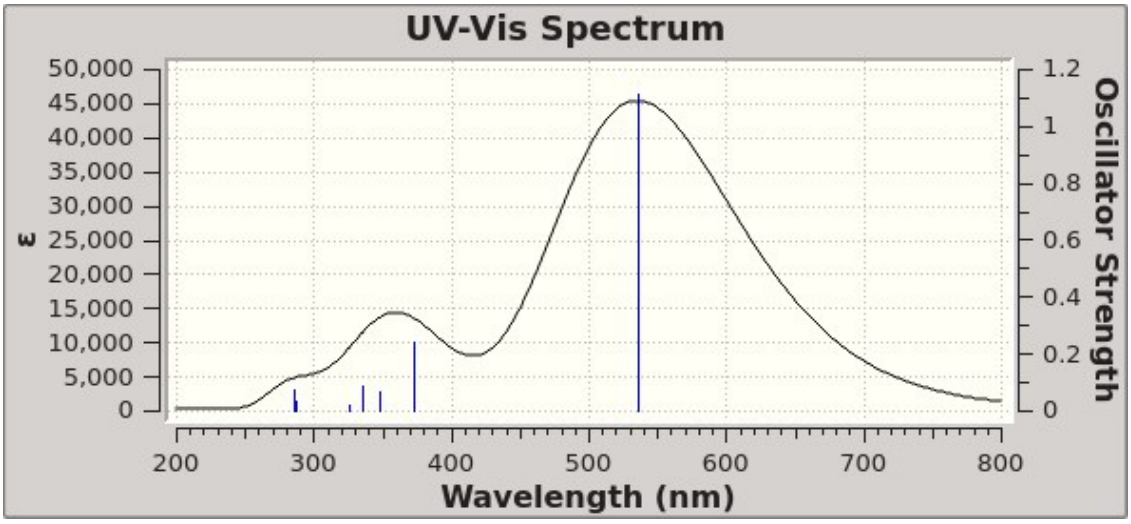


Figure S40. Calculated UV-Vis spectrum for probe **BH**$^+$ in water.

Table S7. Excitation energies and oscillator strengths listing for probe **BH**$^+$ in water.

Excited State	Nature	E (eV)	λ (nm)	*f*	Orbital transitions	Normalized coefficient
1:	A	2.3144	535.71	1.1162	145 ->146	0.70790
2:	A	3.3298	372.35	0.2386	144 ->146	0.12370
					145 ->147	0.68292
3:	A	3.5613	348.14	0.0701	144 ->146	0.68429
					145 ->147	-0.11099
4:	A	3.6954	335.51	0.0848	142 ->146	0.68871
5:	A	3.8006	326.22	0.0193	143 ->146	0.68892
6:	A	4.3217	286.88	0.0313	140 ->146	0.63454
					142 ->147	-0.15780
					145 ->148	-0.21125
7:	A	4.3435	285.45	0.0720	140 ->146	0.15066
					141 ->146	-0.10333
					142 ->147	-0.19059
					145 ->148	0.62862

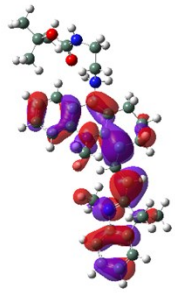
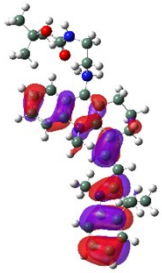
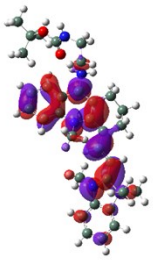
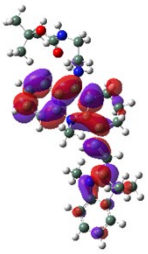
144-HOMO-1	
145-HOMO	
146-LUMO	
147-LUMO+1	

Figure S41. Drawings of selected molecular orbitals for probe **BH**$^{+}$ listed in Table S6.

Theoretical calculations for probe C.

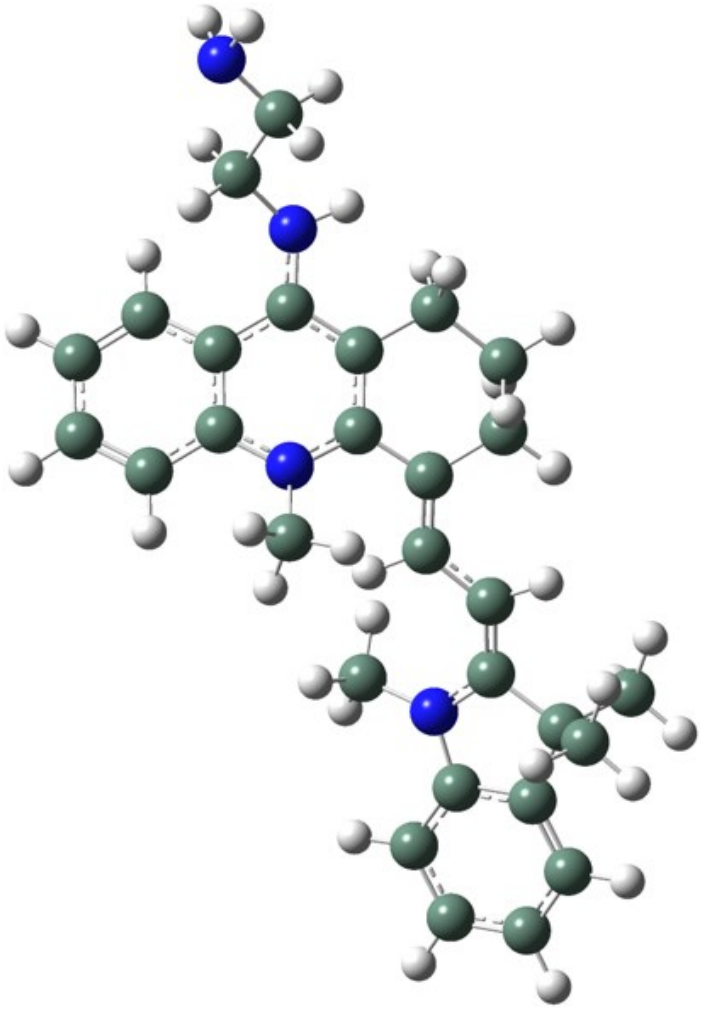


Figure S42. GaussView representation of probe **C.**

Table S8. Calculated atomic coordinates for probe **C** in water.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	4.589212	3.45938	-0.40435	35	H	3.407252	4.995769	0.547285
2	C	3.501299	3.934857	0.337957	36	H	1.653958	3.47104	1.289056
3	C	2.516302	3.072033	0.770495	37	H	5.525003	1.755667	-1.26442
4	C	2.603266	1.697563	0.485133	38	H	0.940847	-3.01396	1.275653
5	C	3.743111	1.194945	-0.1812	39	H	-0.52767	-3.17518	0.32343
6	C	4.703411	2.110456	-0.65665	40	H	1.386765	-4.14482	-0.93735
7	N	1.573814	0.842665	0.828186	41	H	0.76309	-2.71937	-1.76296
8	C	1.511362	-0.41918	0.290988	42	H	3.279317	-2.99625	-0.0732
9	C	2.646038	-0.97898	-0.30851	43	H	3.147342	-2.48801	-1.75007
10	C	3.830383	-0.22694	-0.39848	44	H	-0.88709	0.527589	0.0943
11	C	0.279509	-1.18168	0.365903	45	H	-2.25923	-2.19111	0.562291
12	C	0.440871	-2.67503	0.358741	46	H	-5.30231	2.943287	-1.12059
13	C	1.278542	-3.05878	-0.8577	47	H	-7.76587	2.900497	-0.93034
14	C	2.656988	-2.41562	-0.77074	48	H	-8.94387	0.882615	-0.1243
15	C	-0.92356	-0.54641	0.228095	49	H	-7.65263	-1.15129	0.507935
16	C	-2.20763	-1.15092	0.257824	50	H	-5.88052	-2.9826	-0.41037
17	C	-3.42957	-0.57336	0.034235	51	H	-4.85265	-2.21732	-1.63334
18	N	-3.72958	0.66106	-0.46572	52	H	-4.13324	-3.26401	-0.39659
19	C	-5.1088	0.894821	-0.42932	53	H	-5.77165	-2.18664	2.028411
20	C	-5.76105	-0.24429	0.03861	54	H	-4.02724	-2.47381	2.040125
21	C	-4.73591	-1.30372	0.34404	55	H	-4.66393	-0.87999	2.479403
22	C	-5.80486	2.044002	-0.78101	56	H	-2.35205	2.259428	-0.342
23	C	-7.19674	2.014514	-0.66428	57	H	-2.02602	1.036353	-1.59867
24	C	-7.86149	0.878827	-0.20841	58	H	-3.34001	2.189416	-1.81159
25	C	-7.13792	-0.2644	0.148283	59	H	1.291766	1.710485	2.702276
26	C	-4.90734	-2.51498	-0.5834	60	H	-0.06153	1.987096	1.585332
27	C	-4.79967	-1.73479	1.812555	61	H	0.176807	0.391379	2.314058
28	C	-2.80676	1.588518	-1.07611	62	H	4.894091	-1.80513	-1.00191
29	C	0.684914	1.263057	1.9151	63	H	6.840249	-0.03641	-1.34798
30	N	4.977827	-0.83035	-0.75487	64	H	6.328507	0.375407	0.29372
31	C	6.341626	-0.42115	-0.45021	65	H	6.654072	-1.92833	1.040415
32	C	7.124817	-1.60401	0.107192	66	H	7.062954	-2.45192	-0.59498
33	N	8.487884	-1.17609	0.392795	67	H	8.96314	-1.89272	0.929288
34	H	5.333182	4.148853	-0.78915	68	H	9.007367	-1.08948	-0.47462

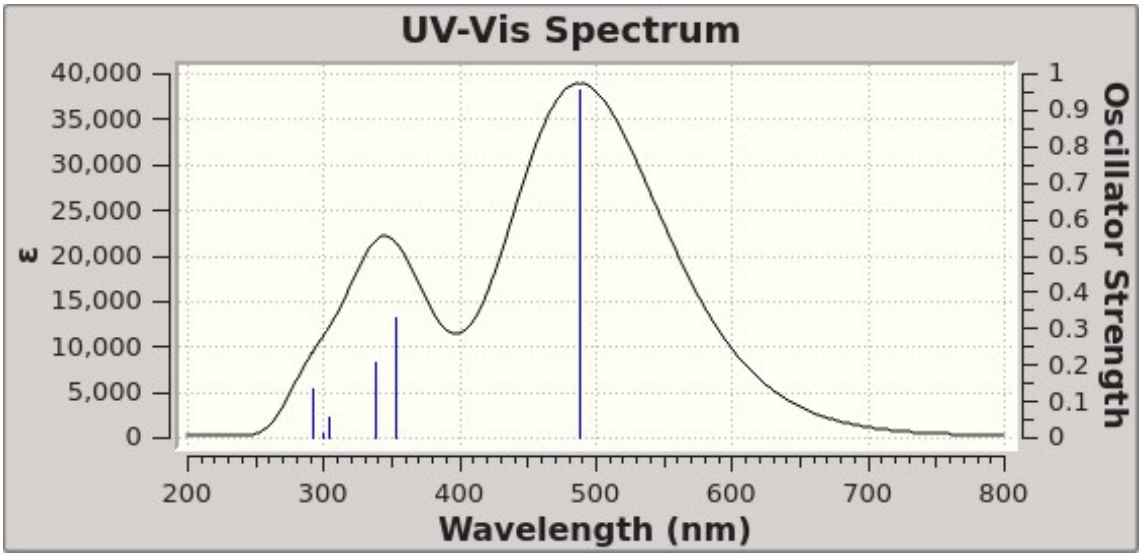


Figure S43. Calculated UV-Vis spectrum for probe **C** in water.

Table S9. Excitation energies and oscillator strengths listing for probe **C** in water.

Excited State	Nature	E (eV)	λ (nm)	*f*	Orbital transitions	Normalized coefficient
1:	A	2.5374	488.62	0.9565	118 ->119	0.70487
2:	A	3.5057	353.66	0.3320	118 ->120	0.69500
3:	A	3.6673	338.08	0.2092	117 ->119	0.68850
4:	A	4.0878	303.30	0.0551	116 ->119	0.67201
					117 ->120	-0.12887
5:	A	4.1365	299.73	0.0017	115 ->119	0.65240
					116 ->119	-0.10254
					118 ->121	0.18654
					118 ->122	-0.13071
6:	A	4.1454	299.09	0.0102	114 ->119	0.10201
					115 ->119	-0.23942
					118 ->121	0.50229
					118 ->122	-0.40135
7:	A	4.2430	292.21	0.1321	114 ->119	-0.13275
					117 ->120	0.14244
					118 ->121	0.41532
					118 ->122	0.51972

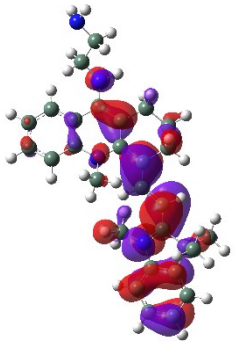
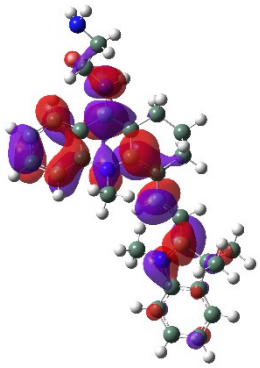
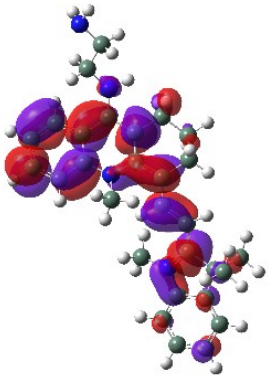
118-HOMO	
119-LUMO	
120-LUMO+1	

Figure S44. Drawings of selected molecular orbitals for probe **C** listed in Table S8.

Theoretical calculations for probe CH^+.

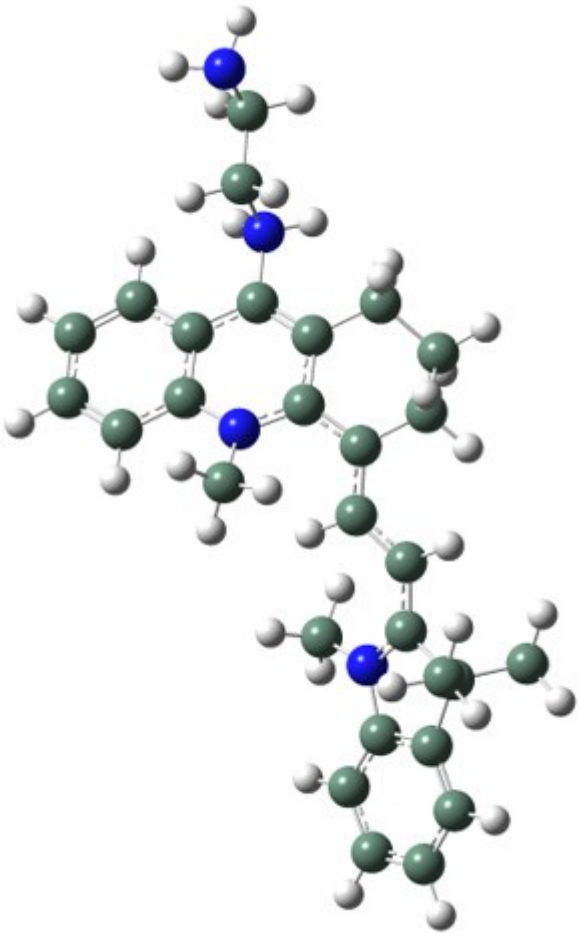


Figure S45. GaussView representation of probe **CH^+**.

Table S10. Calculated atomic coordinates for probe **CH**$^{+}$ in water.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	4.5303	3.583013	-0.1971	36	H	1.456662	3.503215	1.230604
2	C	3.365981	4.021666	0.440296	37	H	5.589668	1.940957	-1.01028
3	C	2.356987	3.13316	0.758031	38	H	0.705356	-3.10284	1.120618
4	C	2.487956	1.774631	0.435129	39	H	-0.5246	-3.13461	-0.13133
5	C	3.659572	1.323988	-0.21309	40	H	1.601665	-4.14136	-0.9533
6	C	4.672987	2.249407	-0.51777	41	H	1.154714	-2.71181	-1.88234
7	N	1.491652	0.865736	0.757919	42	H	3.219377	-2.89771	0.361355
8	C	1.476428	-0.41014	0.2483	43	H	3.52803	-2.64337	-1.34401
9	C	2.714143	-0.94185	-0.27259	44	H	-0.92222	0.517439	-0.01138
10	C	3.739869	-0.07534	-0.47952	45	H	-2.2173	-2.21517	0.507245
11	C	0.298836	-1.18806	0.229722	46	H	-5.34708	2.788473	-1.38257
12	C	0.430786	-2.68568	0.143262	47	H	-7.79978	2.782405	-1.05634
13	C	1.479745	-3.05643	-0.89485	48	H	-8.93312	0.861824	0.003798
14	C	2.81692	-2.42085	-0.54198	49	H	-7.62169	-1.11055	0.765534
15	C	-0.94163	-0.55551	0.1308	50	H	-5.89142	-3.03023	-0.03881
16	C	-2.18276	-1.1794	0.187945	51	H	-4.93455	-2.41605	-1.3966
17	C	-3.4357	-0.60193	-0.00237	52	H	-4.15031	-3.32742	-0.09169
18	N	-3.7485	0.566234	-0.583	53	H	-5.62114	-1.98238	2.296976
19	C	-5.13215	0.817364	-0.49945	54	H	-3.87802	-2.27124	2.221304
20	C	-5.75343	-0.26712	0.110455	55	H	-4.49293	-0.63753	2.53127
21	C	-4.71043	-1.29034	0.462043	56	H	-2.42117	2.201675	-0.6508
22	C	-5.83499	1.933206	-0.92851	57	H	-2.06315	0.855589	-1.7686
23	C	-7.2171	1.924542	-0.73529	58	H	-3.41432	1.941502	-2.09759
24	C	-7.85678	0.841026	-0.1351	59	H	1.151882	1.727205	2.624187
25	C	-7.12336	-0.26818	0.294679	60	H	-0.19466	1.959484	1.489205
26	C	-4.92901	-2.5955	-0.31918	61	H	0.086243	0.369564	2.219987
27	C	-4.66793	-1.55893	1.971387	62	H	5.412837	0.064312	-1.66416
28	C	-2.85444	1.444526	-1.30761	63	H	4.86483	-1.44835	-1.49909
29	C	0.569164	1.26055	1.829998	64	H	6.123154	0.098505	0.670673
30	N	5.017643	-0.58314	-0.97824	65	H	5.566243	-1.57845	0.790964
31	C	6.015444	-0.84094	0.12772	66	H	7.188359	-2.29158	-0.93728
32	C	7.340056	-1.33346	-0.43146	67	H	7.719036	-0.62602	-1.18456
33	N	8.233177	-1.53975	0.697547	68	H	9.004916	-2.13402	0.420338
34	H	5.318708	4.285603	-0.44315	69	H	8.64209	-0.65715	0.983626
35	H	3.243091	5.071944	0.682991					

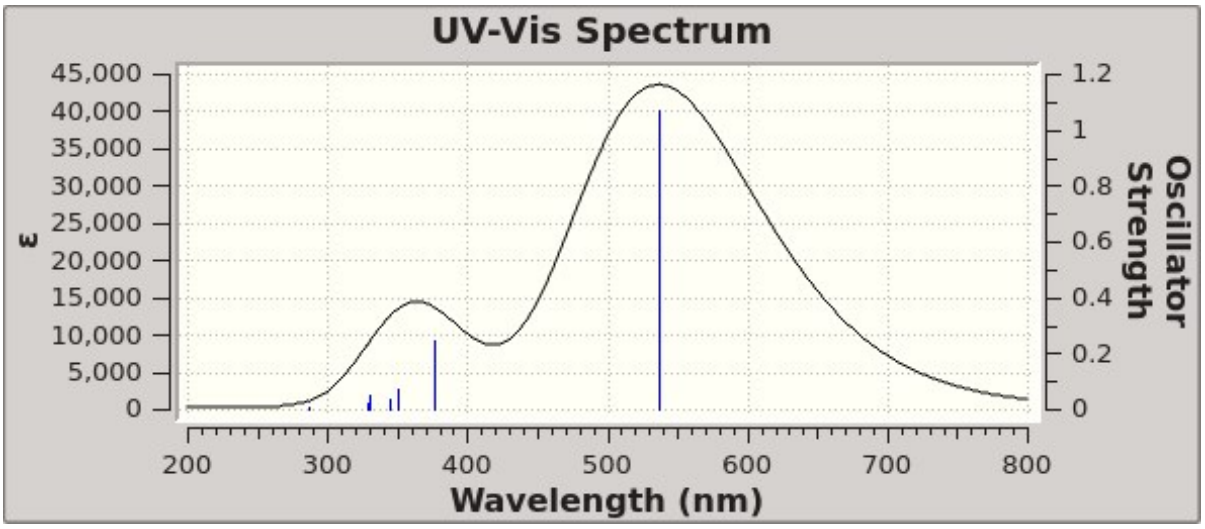


Figure S46. Calculated UV-Vis spectrum for probe **CH**$^{+}$ in water.

Table S11. Excitation energies and oscillator strengths listing for probe **CH**$^{+}$ in water.

Excited State	Nature	E (eV)	λ (nm)	*f*	Orbital transitions	Normalized coefficient
1:	A	2.3097	536.79	1.0710	118 ->119	0.70787
2:	A	3.2980	375.94	0.2460	117 ->119	0.11251
					118 ->120	0.68550
3:	A	3.5397	350.27	0.0703	117 ->119	0.68503
					118 ->120	-0.10110
4:	A	3.6023	344.18	0.0362	114 ->119	-0.14456
					116 ->119	0.68809
5:	A	3.7529	330.37	0.0482	114 ->119	0.67494
					116 ->119	0.14909
6:	A	3.7690	328.96	0.0194	115 ->119	0.69028
7:	A	4.3204	286.97	0.0108	113 ->119	0.52985
					118 ->121	0.43450

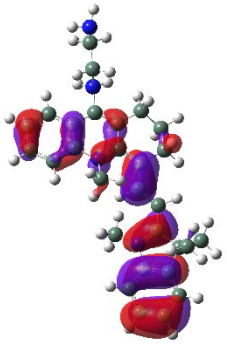
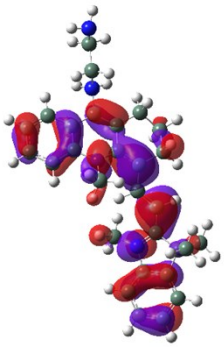
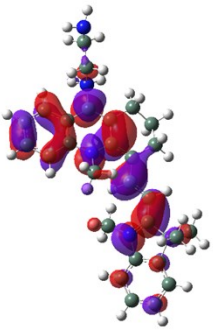
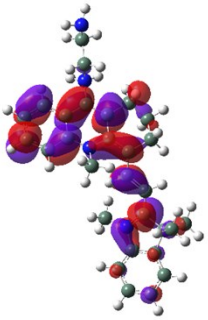
117-HOMO-1	
118-HOMO	
119-LUMO	
120-LUMO+1	

Figure S47. Drawings of selected molecular orbitals for probe **CH**$^{+}$ listed in Table S10.

Theoretical calculations for probe D.

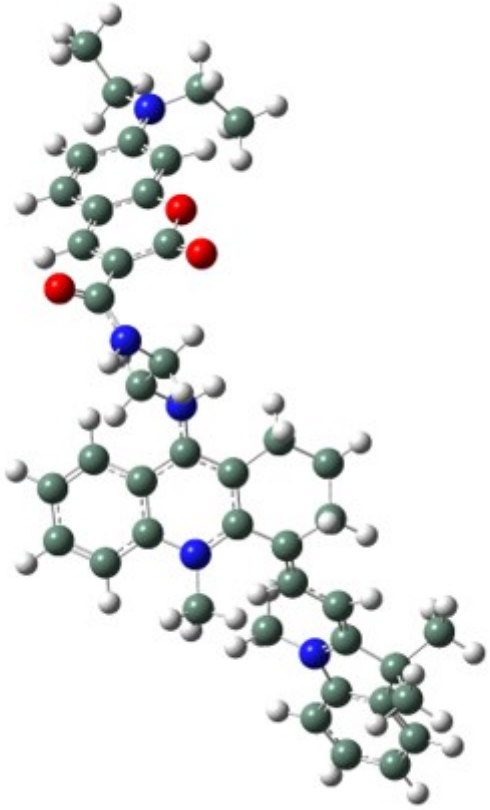


Figure S48. GaussView representation of probe **D**.

Table S12. Calculated atomic coordinates for probe **D** in water.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	0.274072	2.939838	-3.51328	40	O	6.037623	-0.06932	1.81036
2	C	-1.05703	3.306912	-3.74744	41	C	5.590529	1.234206	1.960866
3	C	-2.02959	3.065498	-2.79995	42	O	5.201791	1.545784	3.064799
4	C	-1.69043	2.463026	-1.57503	43	C	7.211756	-0.29111	-1.64673
5	C	-0.33708	2.173905	-1.29318	44	C	7.601639	-1.60411	-1.68539
6	C	0.622941	2.381901	-2.30395	45	C	7.467831	-2.44879	-0.54077
7	N	-2.66951	2.130041	-0.65849	46	C	6.925781	-1.87931	0.633895
8	C	-2.38772	1.287167	0.386849	47	C	9.815663	-4.21173	-2.02814
9	C	-1.05392	1.03405	0.738654	48	C	8.318027	-4.38327	-1.80556
10	C	-0.01622	1.61098	-0.00804	49	N	7.850863	-3.74821	-0.58046
11	C	-3.46227	0.682503	1.149962	50	C	7.812248	-4.59653	0.602315
12	C	-3.12971	0.34197	2.574987	51	C	6.456646	-5.26136	0.810644
13	C	-1.87693	-0.52889	2.577484	52	H	1.024875	3.081357	-4.28345
14	C	-0.70659	0.234792	1.970949	53	H	-1.33942	3.752658	-4.69598
15	C	-4.60218	0.267715	0.51737	54	H	-3.06066	3.301725	-3.02944
16	C	-5.72203	-0.35938	1.122825	55	H	1.644167	2.06124	-2.14454
17	C	-6.83642	-0.88767	0.52522	56	H	-2.94892	1.254554	3.158061
18	N	-7.08293	-1.13183	-0.79438	57	H	-3.95524	-0.18261	3.057127
19	C	-8.39146	-1.59133	-0.98372	58	H	-1.61816	-0.84845	3.591571
20	C	-9.01931	-1.72724	0.252831	59	H	-2.07584	-1.43533	1.995171
21	C	-8.05936	-1.31444	1.336458	60	H	-0.29944	0.926485	2.723591
22	C	-9.04437	-1.89001	-2.17254	61	H	0.099498	-0.47373	1.740446
23	C	-10.36	-2.35182	-2.08576	62	H	-4.65333	0.446187	-0.54943
24	C	-10.995	-2.50286	-0.85556	63	H	-5.75199	-0.37901	2.20723
25	C	-10.3198	-2.18692	0.328405	64	H	-8.57162	-1.76198	-3.14032
26	C	-7.71593	-2.50211	2.246169	65	H	-10.8949	-2.59108	-3.00022
27	C	-8.61556	-0.14774	2.158929	66	H	-12.0189	-2.86159	-0.81564
28	C	-6.12714	-1.05947	-1.87402	67	H	-10.8131	-2.30003	1.290029
29	C	-3.94137	2.853309	-0.74551	68	H	-8.61667	-2.85207	2.757763
30	N	1.251582	1.550408	0.447255	69	H	-7.30504	-3.33404	1.669083
31	C	2.331381	2.489954	0.193526	70	H	-6.98483	-2.21527	3.006664
32	C	2.920414	2.955644	1.527885	71	H	-9.53099	-0.4543	2.67224
33	N	4.044486	3.846147	1.338062	72	H	-7.89748	0.177213	2.916119
34	C	5.266676	3.523566	0.843733	73	H	-8.85105	0.706789	1.520063
35	O	6.029675	4.38276	0.415423	74	H	-6.07109	-0.05625	-2.30528
36	C	5.658052	2.083067	0.791512	75	H	-5.14331	-1.35676	-1.51162
37	C	6.230283	1.614608	-0.35881	76	H	-6.41965	-1.75799	-2.65663
38	C	6.668193	0.280706	-0.47978	77	H	-3.72681	3.90409	-0.93977
39	C	6.545843	-0.55512	0.646382	78	H	-4.58301	2.467091	-1.53869

Row	Symbol	X	Y	Z
79	H	-4.46242	2.769534	0.202357
80	H	1.384487	0.992173	1.277048
81	H	3.118653	2.028064	-0.41164
82	H	1.944943	3.351376	-0.35164
83	H	2.160831	3.494073	2.098038
84	H	3.22588	2.106283	2.140042
85	H	3.842969	4.834612	1.284835
86	H	6.335169	2.288533	-1.2052
87	H	7.32696	0.330423	-2.53014
88	H	8.030265	-1.9911	-2.60048
89	H	6.779333	-2.455	1.538042
90	H	10.12855	-4.71857	-2.94525
91	H	10.08801	-3.1566	-2.11372
92	H	10.38479	-4.63761	-1.19703
93	H	8.065305	-5.44422	-1.73329
94	H	7.745008	-4.00405	-2.65469
95	H	8.590694	-5.35425	0.482013
96	H	8.105031	-4.01147	1.477519
97	H	6.473722	-5.89801	1.699552
98	H	5.663488	-4.52044	0.939574
99	H	6.193818	-5.88702	-0.04696

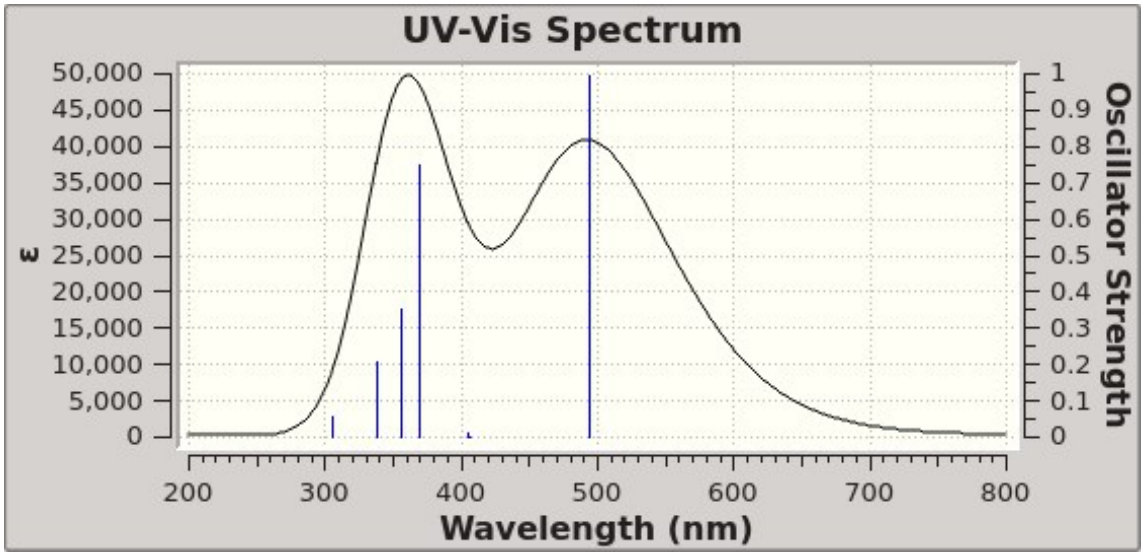


Figure S49. Calculated UV-Vis spectrum for probe **D** in water.

Table S13. Excitation energies and oscillator strengths listing for probe **D** in water.

Excited State	**Nature**	**E (eV)**	**λ (nm)**	***f***	**Orbital transitions**	**Normalized coefficient**
1:	A	2.5073	494.49	0.9919	182 -> 183	0.70438
2:	A	3.0463	407.00	0.0027	182 -> 184	0.70519
3:	A	3.0627	404.82	0.0107	181 -> 183	0.70523
4:	A	3.3554	369.51	0.7471	181 -> 184	0.69301
5:	A	3.4903	355.22	0.3499	182 -> 185	0.69436
6:	A	3.6657	338.22	0.2063	180 -> 183	0.68423
7:	A	4.0632	305.14	0.0562	179 -> 183	0.67700

180 -> 185	0.12036

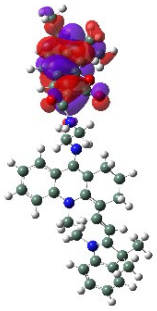
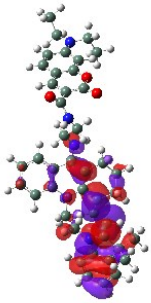
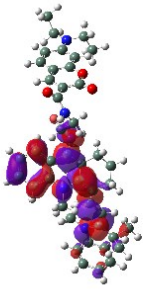
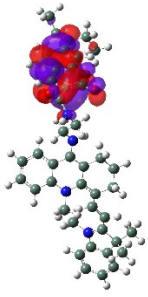
181-HOMO-1	
182-HOMO	
183-LUMO	
184-LUMO+1	

Figure S50. Drawings of selected molecular orbitals for probe **D** listed in Table S12.

Theoretical calculations for probe DH^+.

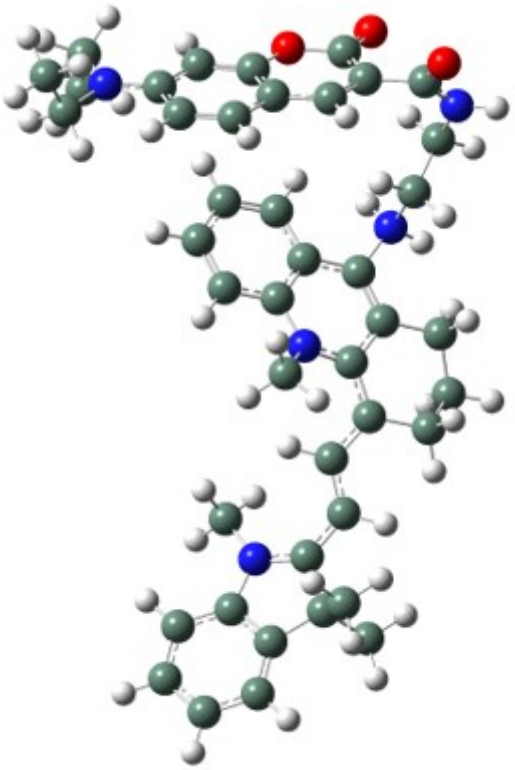


Figure S51. GaussView representation of probe **DH^+.**

Table S14. Calculated atomic coordinates for probe **DH**$^+$ in water.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	-2.45883	-1.22716	-0.97208	41	C	-5.94112	1.89521	-0.36888
2	C	-1.54908	-2.0384	-0.28984	42	O	-6.40099	2.70943	-1.13837
3	C	-0.33353	-1.53562	0.131991	43	C	-4.48021	-1.30326	1.995603
4	C	0.002836	-0.19909	-0.12826	44	C	-4.66451	-2.58429	1.547295
5	C	-0.91821	0.630826	-0.80509	45	C	-5.3603	-2.84687	0.327883
6	C	-2.14909	0.09307	-1.2179	46	C	-5.86116	-1.73848	-0.39306
7	N	1.216152	0.325912	0.290361	47	C	-5.79675	-5.83682	1.636015
8	C	1.68356	1.534644	-0.17164	48	C	-4.91684	-5.25947	0.534408
9	C	0.715595	2.461934	-0.71221	49	N	-5.52105	-4.11161	-0.12892
10	C	-0.52805	1.992169	-0.9852	50	C	-6.28956	-4.40085	-1.33145
11	C	3.046876	1.881812	-0.09485	51	C	-5.45517	-4.30072	-2.60306
12	C	3.426072	3.338803	-0.1467	52	H	-3.40797	-1.62958	-1.3037
13	C	2.601596	4.062077	-1.20103	53	H	-1.79108	-3.07801	-0.09628
14	C	1.115256	3.899823	-0.91567	54	H	0.365975	-2.19122	0.634026
15	C	4.01821	0.87693	-0.10387	55	H	-2.87779	0.696068	-1.74958
16	C	5.38021	1.060343	0.095773	56	H	3.276608	3.807591	0.834552
17	C	6.384561	0.095724	0.029622	57	H	4.485847	3.442531	-0.38488
18	N	6.356719	-1.11055	-0.55381	58	H	2.849834	5.126568	-1.22386
19	C	7.552767	-1.81574	-0.31405	59	H	2.833682	3.653276	-2.19028
20	C	8.420552	-1.00263	0.406749	60	H	0.85412	4.457341	-0.00662
21	C	7.746756	0.314682	0.66972	61	H	0.555302	4.363898	-1.73399
22	C	7.885698	-3.10634	-0.6968	62	H	3.664346	-0.13004	-0.28449
23	C	9.153254	-3.56609	-0.33708	63	H	5.716923	2.028501	0.450028
24	C	10.0395	-2.76024	0.376213	64	H	7.199195	-3.74734	-1.23868
25	C	9.675605	-1.4653	0.755164	65	H	9.448012	-4.57283	-0.61651
26	C	8.487629	1.466504	-0.02723	66	H	11.01883	-3.14443	0.643763
27	C	7.606084	0.586637	2.172693	67	H	10.36509	-0.83944	1.31413
28	C	5.318641	-1.63839	-1.41377	68	H	9.495723	1.551529	0.385473
29	C	1.873036	-0.36258	1.408522	69	H	8.570521	1.29144	-1.10217
30	N	-1.57371	2.926505	-1.40759	70	H	7.977438	2.419358	0.132572
31	C	-2.39563	3.376181	-0.22425	71	H	8.59706	0.666663	2.625908
32	C	-3.65178	4.131119	-0.64723	72	H	7.074174	1.523561	2.354078
33	N	-4.43231	4.461565	0.52271	73	H	7.064897	-0.22092	2.670951
34	C	-5.07197	3.581639	1.340029	74	H	4.590175	-2.22081	-0.84589
35	O	-5.47178	3.910309	2.447584	75	H	4.82035	-0.81705	-1.92588
36	C	-5.23047	2.175761	0.858402	76	H	5.777394	-2.2807	-2.16324
37	C	-4.80385	1.152027	1.657083	77	H	1.109005	-0.62515	2.140462
38	C	-4.96433	-0.19504	1.274731	78	H	2.394358	-1.26781	1.097212
39	C	-5.65202	-0.46035	0.075806	79	H	2.585414	0.317436	1.866434
40	O	-6.1256	0.560976	-0.68875	80	H	-1.16333	3.739945	-1.86821

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
81	H	-2.17473	2.498564	-2.11511	91	H	-5.98781	-5.10074	2.421181
82	H	-2.6528	2.471546	0.324409	92	H	-6.76278	-6.15686	1.235628
83	H	-1.74042	3.987429	0.395185	93	H	-5.31738	-6.70607	2.094696
84	H	-3.39562	5.064734	-1.15041	94	H	-3.93212	-4.98264	0.917355
85	H	-4.25405	3.54824	-1.34584	95	H	-4.7299	-6.01275	-0.23509
86	H	-4.31027	5.382146	0.919964	96	H	-6.69304	-5.41051	-1.22175
87	H	-4.3106	1.382871	2.598011	97	H	-7.15888	-3.74029	-1.37372
88	H	-3.94823	-1.13081	2.926763	98	H	-5.05181	-3.29408	-2.74032
89	H	-4.28028	-3.40202	2.142568	99	H	-4.61263	-4.99711	-2.57131
90	H	-6.3805	-1.85179	-1.3353	100	H	-6.06093	-4.54387	-3.48024

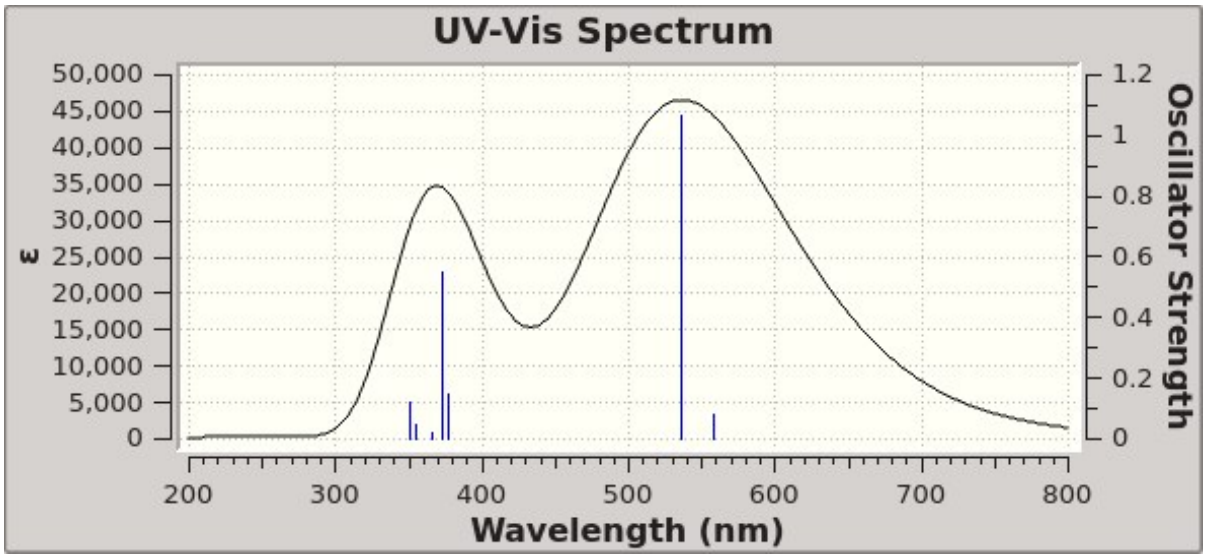
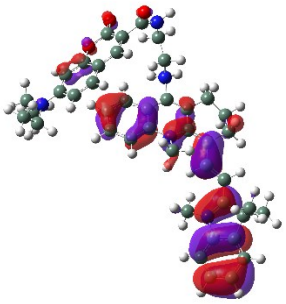
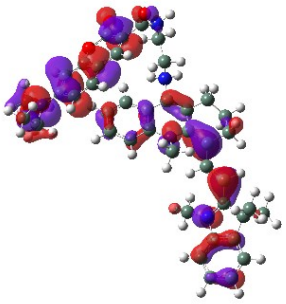
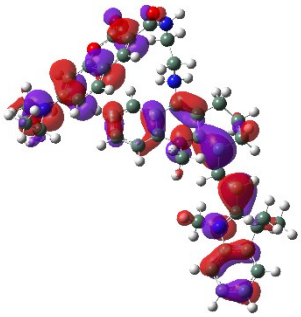
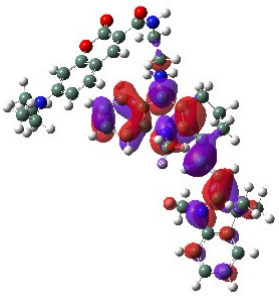


Figure S52. Calculated UV-Vis spectrum for probe **DH**$^{+}$ in water**.**

Table S15. Excitation energies and oscillator strengths listing for probe **DH**$^{+}$ in water.

Excited State	Nature	E (eV)	λ (nm)	*f*	Orbital transitions	Normalized coefficient
1:	A	2.2191	558.70	0.0814	181 -> 183	-0.43006
					182 -> 183	0.55906
2:	A	2.3141	535.78	1.0657	181 -> 183	0.56043
					182 -> 183	0.43212
3:	A	3.2912	376.72	0.1479	181 -> 185	0.29854
					182 -> 184	-0.44124
					182 -> 185	0.44096
4:	A	3.3287	372.47	0.5530	180 -> 183	-0.10017
					181 -> 184	0.50423
					181 -> 185	-0.22410
					182 -> 184	-0.37584
					182 -> 185	-0.17046
5:	A	3.3894	365.80	0.0197	181 -> 184	0.47805
					181 -> 185	0.16026
					182 -> 184	0.39491
					182 -> 185	0.28350
6:	A	3.4982	354.42	0.0460	181 -> 185	0.56598
					182 -> 185	-0.40084
7:	A	3.5321	351.02	0.1184	179 -> 183	-0.10698
					180 -> 183	0.66897
					182 -> 185	-0.12681

180-HOMO-2	
181-HOMO-1	
182-HOMO	
183-LUMO	

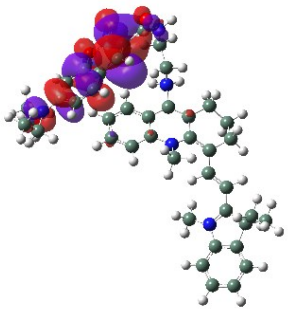
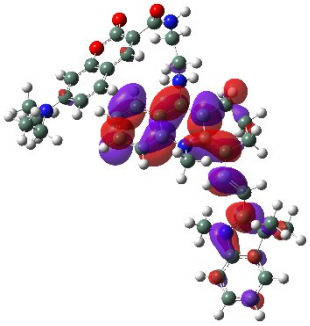
184-LUMO+1	
185-LUMO+1	

Figure S53. Drawings of selected molecular orbitals for probe **DH**$^{+}$ listed in Table S14.

Results for the pKa Calculations.

Atomic coordinates for the models refined with the SMD[7] implicit solvation model in water are listed below together with those involving the added water molecule including pictures of the water adduct.

Table S16. Calculated atomic coordinates for probe **B.**

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	-1.26568	4.545837	-0.14846	8	C	-2.20331	-0.37087	0.185614
2	C	-2.6451	4.379004	-0.35092	9	C	-0.84578	-0.24733	0.531745
3	C	-3.22839	3.131227	-0.23331	10	C	-0.3043	1.024814	0.787883
4	C	-2.44321	2.010746	0.108679	11	C	-2.75752	-1.68822	-0.0883
5	C	-1.0673	2.182546	0.394355	12	C	-2.05734	-2.8301	0.595932
6	C	-0.49358	3.4615	0.217358	13	C	-0.57585	-2.77048	0.232189
7	N	-3.00251	0.745774	0.143775	14	C	0.030456	-1.46265	0.730188

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
15	C	-3.70514	-1.84883	-1.06654	50	H	1.000153	-1.30366	0.239171
16	C	-4.22065	-3.08645	-1.54505	51	H	-4.03417	-0.9509	-1.576
17	C	-5.15724	-3.29642	-2.5277	52	H	-3.79656	-4.00033	-1.13943
18	N	-5.96861	-2.39339	-3.1614	53	H	-7.82118	-1.37939	-5.07601
19	C	-6.72447	-3.01794	-4.16114	54	H	-8.96695	-2.8845	-6.6772
20	C	-6.45332	-4.38845	-4.16321	55	H	-8.51472	-5.31964	-6.6842
21	C	-5.44008	-4.68946	-3.09046	56	H	-6.88543	-6.29631	-5.05833
22	C	-7.62255	-2.44663	-5.05892	57	H	-5.30964	-5.78563	-1.21257
23	C	-8.26199	-3.30384	-5.96355	58	H	-6.93491	-5.15907	-1.56979
24	C	-8.00687	-4.67744	-5.96954	59	H	-6.30957	-6.56773	-2.45378
25	C	-7.09187	-5.22847	-5.05968	60	H	-3.43006	-5.50077	-2.89642
26	C	-6.03729	-5.60428	-2.01015	61	H	-4.41902	-6.27825	-4.14845
27	C	-4.17229	-5.31961	-3.68029	62	H	-3.72604	-4.66836	-4.43832
28	C	-6.17747	-1.01522	-2.77368	63	H	-5.41951	-0.34876	-3.19689
29	C	-4.45993	0.661803	0.297472	64	H	-6.16317	-0.93823	-1.68414
30	N	0.938693	1.132912	1.309785	65	H	-7.15925	-0.69233	-3.12139
31	C	1.428216	2.195662	2.179772	66	H	-4.76964	1.403971	1.035773
32	C	2.056559	1.572278	3.426838	67	H	-4.98447	0.848156	-0.64221
33	N	2.624078	2.574251	4.300017	68	H	-4.72079	-0.32723	0.664784
34	C	3.826747	3.137441	4.029668	69	H	1.415389	0.251207	1.450557
35	O	4.047991	4.17881	4.851089	70	H	2.174905	2.814383	1.66946
36	O	4.60466	2.724893	3.16691	71	H	0.596377	2.837643	2.477321
37	C	5.31457	4.933615	4.823688	72	H	1.30301	1.014906	3.989023
38	C	6.481112	4.024476	5.191643	73	H	2.847227	0.871621	3.14164
39	C	5.092286	5.976952	5.909081	74	H	2.008756	3.07546	4.927045
40	C	5.503322	5.607795	3.469802	75	H	7.379467	4.636761	5.318241
41	H	-0.80403	5.518568	-0.29079	76	H	6.679306	3.279912	4.418491
42	H	-3.26037	5.228164	-0.63579	77	H	6.282102	3.510983	6.137919
43	H	-4.28384	3.017095	-0.44982	78	H	5.977114	6.614683	5.991331
44	H	0.575611	3.590908	0.334826	79	H	4.231611	6.609685	5.669655
45	H	-2.17067	-2.74559	1.686298	80	H	4.919456	5.498896	6.87859
46	H	-2.4896	-3.7894	0.305055	81	H	4.615509	6.192006	3.206622
47	H	-0.02664	-3.61369	0.665765	82	H	6.355365	6.291984	3.532208
48	H	-0.47576	-2.84234	-0.858	83	H	5.70014	4.884249	2.676824
49	H	0.243293	-1.55312	1.807161					

Table S17. Calculated atomic coordinates for probe **BH$^+$.**

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	-3.83785	2.283156	2.025899	42	H	-4.73512	1.281678	3.718792
2	C	-3.83599	1.40868	3.122472	43	H	-2.73207	0.004857	4.290995
3	C	-2.69969	0.68997	3.452605	44	H	-2.72017	3.097124	0.411185
4	C	-1.53136	0.825456	2.679975	45	H	3.589165	0.114806	2.336707
5	C	-1.53023	1.693606	1.56182	46	H	3.689807	-1.53603	1.732914
6	C	-2.69833	2.421512	1.2574	47	H	3.886505	0.097396	-0.14317
7	N	-0.37735	0.126125	3.006972	48	H	2.435371	-0.89722	-0.31188
8	C	0.69612	0.051929	2.154823	49	H	2.633624	1.969226	0.733575
9	C	0.779585	1.013197	1.080437	50	H	1.862313	1.42112	-0.74601
10	C	-0.31199	1.793337	0.824099	51	H	0.341338	-2.34388	3.072346
11	C	1.711772	-0.93392	2.311316	52	H	3.332413	-3.05657	2.936901
12	C	3.069043	-0.63793	1.727887	53	H	-1.13335	-7.09109	4.466132
13	C	2.911057	-0.12317	0.302924	54	H	-0.40708	-9.26567	5.412743
14	C	2.061417	1.140373	0.293676	55	H	1.994468	-9.78221	5.716695
15	C	1.38369	-2.18147	2.835154	56	H	3.733145	-8.10633	5.074332
16	C	2.270017	-3.2423	3.059856	57	H	4.691999	-6.61172	3.067238
17	C	1.97197	-4.5212	3.516696	58	H	3.381352	-6.24973	1.923354
18	N	0.770136	-5.12103	3.666144	59	H	4.524215	-4.97383	2.40506
19	C	0.914576	-6.4029	4.229545	60	H	4.629271	-5.64682	5.433947
20	C	2.270723	-6.67432	4.40728	61	H	4.458678	-4.01605	4.754728
21	C	3.070105	-5.48709	3.946416	62	H	3.272948	-4.6155	5.936789
22	C	-0.07698	-7.31078	4.585311	63	H	-0.9229	-3.90437	3.993184
23	C	0.341926	-8.53346	5.122596	64	H	-0.41647	-4.09826	2.289539
24	C	1.698115	-8.82493	5.296327	65	H	-1.21156	-5.43177	3.133065
25	C	2.676879	-7.88789	4.937507	66	H	-0.69238	0.371009	5.052442
26	C	3.970301	-5.85048	2.754602	67	H	-0.85828	-1.31775	4.509602
27	C	3.907072	-4.89914	5.09132	68	H	0.750349	-0.56733	4.624687
28	C	-0.51702	-4.60124	3.253038	69	H	0.434521	2.480385	-0.94672
29	C	-0.29486	-0.39108	4.380096	70	H	-1.13746	2.891476	-0.78457
30	N	-0.24471	2.7803	-0.24016	71	H	-0.45096	4.404297	1.093931
31	C	0.162517	4.155608	0.226849	72	H	1.208333	4.084907	0.531273
32	C	-0.00862	5.167476	-0.89446	73	H	0.596869	6.040146	-0.64576
33	N	-1.36794	5.642268	-1.08432	74	H	0.384244	4.760203	-1.83398
34	C	-2.34103	4.881095	-1.61896	75	H	-1.56799	6.622144	-0.93499
35	O	-3.43983	5.591831	-1.86679	76	H	-5.58323	3.973399	-3.94818
36	O	-2.21495	3.662649	-1.83951	77	H	-3.92519	3.44304	-3.6123
37	C	-4.71847	4.946814	-2.24824	78	H	-4.22162	5.034585	-4.35283
38	C	-4.5923	4.306597	-3.62414	79	H	-6.6663	5.788977	-2.58728
39	C	-5.67026	6.13228	-2.29373	80	H	-5.74442	6.610257	-1.31183
40	C	-5.1492	3.955	-1.17453	81	H	-5.33184	6.874488	-3.02358
41	H	-4.73193	2.844853	1.772142	82	H	-5.1227	4.425572	-0.18654

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
83	H	-6.17948	3.646993	-1.37775	84	H	-4.52169	3.062515	-1.16247

Table S18. Calculated atomic coordinates for probe **C.**

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	-4.76584	2.652567	0.511325	35	H	-5.35235	1.146817	1.950398
2	C	-4.53095	1.580554	1.386542	36	H	-3.11901	0.175594	2.149308
3	C	-3.26562	1.038069	1.510174	37	H	-3.91294	4.001426	-0.89562
4	C	-2.19007	1.56659	0.76646	38	H	2.752285	1.931147	-0.00483
5	C	-2.39943	2.699534	-0.05787	39	H	3.272516	0.48888	-0.87024
6	C	-3.71393	3.198975	-0.19659	40	H	2.961172	2.362765	-2.48872
7	N	-0.9426	0.971035	0.821473	41	H	1.75583	1.10445	-2.77571
8	C	0.022317	1.286575	-0.10481	42	H	1.423954	3.807368	-1.41278
9	C	-0.11816	2.437521	-0.89698	43	H	0.546252	3.1741	-2.7991
10	C	-1.26589	3.244544	-0.76474	44	H	0.14329	-1.31176	0.102685
11	C	1.209932	0.45614	-0.2473	45	H	3.1115	-1.4484	-0.74637
12	C	2.435052	1.176811	-0.73914	46	H	-0.20744	-6.44052	0.863631
13	C	2.086169	1.864391	-2.05664	47	H	0.814655	-8.65585	0.421786
14	C	0.980152	2.889945	-1.83086	48	H	3.05595	-8.84027	-0.61644
15	C	1.120082	-0.90708	-0.13358	49	H	4.324643	-6.77602	-1.23373
16	C	2.165645	-1.84073	-0.38517	50	H	4.321561	-4.54398	-2.63686
17	C	2.135383	-3.2102	-0.29081	51	H	2.638013	-4.02747	-2.87103
18	N	1.163384	-4.02707	0.223102	52	H	3.889502	-2.83054	-2.47129
19	C	1.510279	-5.37441	0.067691	53	H	5.358491	-4.49655	-0.30208
20	C	2.77087	-5.46635	-0.52753	54	H	4.922005	-2.78282	-0.1514
21	C	3.291056	-4.07783	-0.79084	55	H	4.396672	-3.94605	1.086682
22	C	0.779782	-6.50719	0.41682	56	H	0.277044	-2.77875	1.637658
23	C	1.363466	-7.75439	0.160433	57	H	-0.81809	-3.29127	0.323196
24	C	2.626957	-7.85997	-0.42658	58	H	-0.34144	-4.43297	1.594177
25	C	3.340974	-6.7035	-0.7754	59	H	-1.05034	0.618932	2.875307
26	C	3.546986	-3.8537	-2.2867	60	H	-1.09639	-0.86699	1.895814
27	C	4.570072	-3.80598	0.015162	61	H	0.425197	0.030108	2.094178
28	C	0.003095	-3.60383	0.975772	62	H	-0.46734	4.691494	-1.8879
29	C	-0.652	0.126623	1.98615	63	H	-2.95232	5.741019	-1.64148
30	N	-1.29808	4.454022	-1.36071	64	H	-2.46265	5.48586	0.042824
31	C	-2.08893	5.617743	-0.97565	65	H	-0.41382	6.784971	-0.30721
32	C	-1.22013	6.866053	-1.04349	66	H	-0.74732	6.921343	-2.0374
33	N	-2.02875	8.039223	-0.72484	67	H	-1.42863	8.858041	-0.68361
34	H	-5.77024	3.044666	0.380976	68	H	-2.68496	8.209782	-1.48344

Table S19. Calculated atomic coordinates for probe **CH^+.**

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	-2.87096	2.242988	3.724713	36	H	-1.64849	-0.8876	3.218763
2	C	-2.63843	0.872745	3.912503	37	H	-2.4545	3.96518	2.54659
3	C	-1.79504	0.17466	3.064978	38	H	2.562074	0.745856	-1.65955
4	C	-1.16614	0.837201	1.995066	39	H	1.715531	-0.09288	-2.95511
5	C	-1.40501	2.217486	1.791371	40	H	1.602922	2.37982	-3.274
6	C	-2.26091	2.904499	2.675688	41	H	-0.00524	1.685691	-3.03776
7	N	-0.30242	0.157807	1.146372	42	H	1.723514	3.129976	-0.96776
8	C	0.128837	0.69996	-0.04073	43	H	0.224393	3.701199	-1.68091
9	C	0.022342	2.131919	-0.20755	44	H	-0.51531	-1.7548	-0.53068
10	C	-0.71921	2.830506	0.69914	45	H	1.545507	-2.04666	-2.79724
11	C	0.67407	-0.103	-1.07855	46	H	-2.44224	-6.53611	-1.19559
12	C	1.569166	0.561449	-2.0933	47	H	-2.10224	-8.75356	-2.24609
13	C	0.95584	1.879965	-2.54576	48	H	-0.26952	-9.12788	-3.86703
14	C	0.747556	2.797948	-1.34941	49	H	1.278052	-7.2599	-4.46918
15	C	0.254327	-1.42632	-1.2141	50	H	1.40162	-4.76137	-5.4546
16	C	0.714717	-2.34272	-2.16421	51	H	-0.10968	-3.91064	-5.0676
17	C	0.295087	-3.654	-2.37235	52	H	1.45405	-3.08607	-4.86883
18	N	-0.74309	-4.32433	-1.82876	53	H	2.988238	-5.47017	-3.5797
19	C	-0.76392	-5.66302	-2.26874	54	H	3.046015	-3.78886	-3.01683
20	C	0.2786	-5.86195	-3.17298	55	H	2.593429	-5.08848	-1.88999
21	C	1.038438	-4.57501	-3.33221	56	H	-1.3651	-3.68321	0.080126
22	C	-1.63832	-6.6835	-1.90989	57	H	-2.09	-2.81621	-1.30745
23	C	-1.43776	-7.93369	-2.50604	58	H	-2.60639	-4.46008	-0.92413
24	C	-0.40189	-8.14605	-3.42053	59	H	0.525149	-0.94287	2.710663
25	C	0.46858	-7.10192	-3.76081	60	H	-0.43066	-1.93385	1.579728
26	C	0.93518	-4.04458	-4.77154	61	H	1.170718	-1.32388	1.102691
27	C	2.508946	-4.73753	-2.92291	62	H	-1.73974	4.612032	0.864628
28	C	-1.75141	-3.78654	-0.93905	63	H	-0.72448	4.557324	-0.40614
29	C	0.265516	-1.09594	1.661575	64	H	0.128825	4.689675	2.412734
30	N	-0.81085	4.281567	0.57889	65	H	1.20944	4.669407	0.99609
31	C	0.243109	5.007539	1.373932	66	H	0.202426	6.777335	0.163945
32	C	0.063986	6.50566	1.215462	67	H	-0.96622	6.777959	1.492009
33	N	1.081926	7.181391	2.011515	68	H	1.070162	8.173448	1.793355
34	H	-3.53153	2.78394	4.395506	69	H	0.844517	7.104662	2.997565
35	H	-3.12876	0.344835	4.725444					

Table S20. Calculated atomic coordinates for probe **D.**

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	-4.0018	0.17682	3.33858	42	O	3.546255	3.803449	4.957131
2	C	-4.56368	-1.09588	3.151736	43	C	-0.17118	6.454962	1.961875
3	C	-3.77084	-2.16785	2.786785	44	C	0.256578	6.99039	0.772189
4	C	-2.38184	-1.99747	2.612819	45	C	1.617946	6.85326	0.345354
5	C	-1.79298	-0.73666	2.878177	46	C	2.510777	6.151086	1.194739
6	C	-2.63861	0.348048	3.202196	47	C	0.91021	9.482035	-1.54125
7	N	-1.5993	-3.04657	2.164052	48	C	1.12197	7.99295	-1.78603
8	C	-0.32044	-2.82049	1.713945	49	N	2.039298	7.380431	-0.82948
9	C	0.321084	-1.60679	2.013628	50	C	3.4425	7.355983	-1.22936
10	C	-0.3632	-0.61277	2.737199	51	C	3.825873	6.089034	-1.98523
11	C	0.388875	-3.84186	0.959846	52	H	-4.63445	1.026586	3.577714
12	C	1.887488	-3.80393	1.083014	53	H	-5.63509	-1.24051	3.261063
13	C	2.362192	-2.40384	0.700725	54	H	-4.23411	-3.12778	2.5926
14	C	1.769866	-1.37271	1.65557	55	H	-2.2216	1.341167	3.311915
15	C	-0.2856	-4.62296	0.056553	56	H	2.191263	-4.02461	2.116335
16	C	0.287792	-5.61503	-0.78691	57	H	2.354628	-4.556	0.444059
17	C	-0.31375	-6.40444	-1.73722	58	H	3.455509	-2.33573	0.725041
18	N	-1.60254	-6.39871	-2.19851	59	H	2.045396	-2.19148	-0.32796
19	C	-1.80377	-7.4207	-3.13528	60	H	2.353858	-1.37201	2.589345
20	C	-0.60431	-8.1065	-3.34094	61	H	1.885745	-0.36945	1.222904
21	C	0.462468	-7.50105	-2.46815	62	H	-1.35118	-4.45363	-0.02547
22	C	-2.97248	-7.77409	-3.80491	63	H	1.345551	-5.82406	-0.65721
23	C	-2.90056	-8.84155	-4.70861	64	H	-3.91223	-7.2569	-3.63855
24	C	-1.7048	-9.52945	-4.92972	65	H	-3.79893	-9.13792	-5.2445
25	C	-0.54206	-9.16019	-4.23703	66	H	-1.6769	-10.3551	-5.63582
26	C	1.5935	-6.89914	-3.31518	67	H	0.392955	-9.69155	-4.39932
27	C	1.026405	-8.53554	-1.48506	68	H	2.080357	-7.69053	-3.89467
28	C	-2.63954	-5.46225	-1.82766	69	H	1.206609	-6.1479	-4.01075
29	C	-2.13029	-4.40315	2.342575	70	H	2.346787	-6.42809	-2.67559
30	N	0.320531	0.455538	3.200908	71	H	1.502069	-9.35064	-2.04043
31	C	-0.01614	1.304509	4.333135	72	H	1.778532	-8.0813	-0.83222
32	C	1.243197	1.547852	5.165067	73	H	0.232694	-8.95819	-0.86121
33	N	0.995125	2.500931	6.231035	74	H	-3.01711	-5.65982	-0.8187
34	C	0.826633	3.828499	6.068985	75	H	-2.2584	-4.43974	-1.88046
35	O	0.313865	4.538537	6.954798	76	H	-3.46764	-5.54776	-2.53003
36	C	1.255396	4.440719	4.779573	77	H	-2.60175	-4.46035	3.325268
37	C	0.346874	5.152904	4.035643	78	H	-2.8624	-4.66567	1.575021
38	C	0.71047	5.751217	2.813004	79	H	-1.30713	-5.11219	2.308873
39	C	2.049266	5.628003	2.382751	80	H	1.302619	0.47118	2.955888
40	O	2.961745	4.956556	3.158514	81	H	-0.41696	2.268781	3.999352
41	C	2.626592	4.351579	4.353361	82	H	-0.77258	0.8146	4.950805

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
83	H	1.581541	0.615605	5.623242	92	H	1.860978	10.02357	-1.59224
84	H	2.05645	1.906289	4.529827	93	H	1.548603	7.83171	-2.78099
85	H	0.631027	2.132368	7.103159	94	H	0.171066	7.453046	-1.77595
86	H	-0.68089	5.238193	4.382723	95	H	3.609074	8.236085	-1.85826
87	H	-1.2072	6.573921	2.269298	96	H	4.073329	7.492077	-0.34615
88	H	-0.45161	7.539373	0.163335	97	H	4.879505	6.131011	-2.28137
89	H	3.548446	5.990625	0.929434	98	H	3.679828	5.196846	-1.36778
90	H	0.240989	9.89806	-2.30205	99	H	3.222138	5.979664	-2.89259
91	H	0.465398	9.66456	-0.55763					

Table S21. Calculated atomic coordinates for probe **DH$^+$.**

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	1.846766	1.712032	1.179588	30	N	4.094316	-1.36408	3.116828
2	C	0.652257	1.37367	0.527556	31	C	3.799646	-1.15312	4.585785
3	C	0.186604	0.071755	0.536326	32	C	4.639047	-0.01563	5.152271
4	C	0.929603	-0.93638	1.175424	33	N	4.258667	0.238325	6.526928
5	C	2.161557	-0.61253	1.791741	34	C	3.118129	0.845308	6.912972
6	C	2.587517	0.7321	1.810581	35	O	2.732413	0.836114	8.093688
7	N	0.458446	-2.24216	1.224594	36	C	2.310583	1.534131	5.860844
8	C	1.257643	-3.29472	1.595904	37	C	1.081821	1.049398	5.492584
9	C	2.495134	-3.00002	2.279115	38	C	0.295971	1.716318	4.530451
10	C	2.887188	-1.69695	2.366449	39	C	0.773777	2.937771	4.010952
11	C	0.891589	-4.64711	1.342649	40	O	1.999563	3.419752	4.399089
12	C	1.507388	-5.70787	2.21881	41	C	2.815677	2.749943	5.284575
13	C	3.011885	-5.48402	2.305364	42	O	3.907132	3.264845	5.520221
14	C	3.299872	-4.11383	2.902233	43	C	-0.94273	1.252319	4.035308
15	C	0.123763	-4.96173	0.224276	44	C	-1.64006	1.955746	3.085211
16	C	-0.33212	-6.2386	-0.12698	45	C	-1.13639	3.189485	2.561084
17	C	-1.04738	-6.61684	-1.25759	46	C	0.082672	3.6847	3.083298
18	N	-1.34541	-5.9042	-2.36644	47	C	-4.26595	3.5296	1.4147
19	C	-2.14148	-6.65852	-3.25039	48	C	-2.88936	3.231941	0.83381
20	C	-2.33963	-7.93038	-2.71384	49	N	-1.80203	3.85366	1.584236
21	C	-1.63875	-8.01544	-1.38669	50	C	-1.34053	5.140954	1.077026
22	C	-2.68398	-6.27847	-4.47349	51	C	-0.28088	5.017182	-0.0134
23	C	-3.43822	-7.23528	-5.16217	52	H	2.187847	2.742112	1.194874
24	C	-3.63882	-8.51663	-4.64049	53	H	0.075175	2.139515	0.018632
25	C	-3.08614	-8.87229	-3.40269	54	H	-0.73978	-0.16547	0.027457
26	C	-0.52636	-9.07581	-1.4122	55	H	3.499599	1.023841	2.323084
27	C	-2.62934	-8.30228	-0.24915	56	H	1.070085	-5.6737	3.226391
28	C	-0.88949	-4.56529	-2.67723	57	H	1.30195	-6.70107	1.815487
29	C	-0.98938	-2.4109	1.033946	58	H	3.486917	-6.25206	2.924621

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
59	H	3.447161	-5.55502	1.300998	80	H	4.802011	-2.09899	3.013805
60	H	3.063747	-4.1269	3.975409	81	H	4.530633	-0.524	2.718991
61	H	4.373523	-3.90889	2.823204	82	H	2.737478	-0.91837	4.656746
62	H	-0.11833	-4.14364	-0.43957	83	H	3.991036	-2.09995	5.092015
63	H	-0.17726	-7.04734	0.58043	84	H	5.701796	-0.26598	5.133708
64	H	-2.54349	-5.28432	-4.88597	85	H	4.502585	0.889631	4.554395
65	H	-3.87718	-6.96978	-6.12041	86	H	4.758468	-0.2605	7.255236
66	H	-4.22967	-9.23935	-5.1968	87	H	0.71119	0.125498	5.932711
67	H	-3.23999	-9.86602	-2.98922	88	H	-1.34217	0.318123	4.422391
68	H	-0.9708	-10.0603	-1.58957	89	H	-2.5882	1.564264	2.736484
69	H	0.194798	-8.87069	-2.20914	90	H	0.538828	4.594646	2.714147
70	H	0.00473	-9.10479	-0.45585	91	H	-4.36129	3.141922	2.434187
71	H	-3.10215	-9.27513	-0.41654	92	H	-4.45114	4.608898	1.441254
72	H	-2.11422	-8.33123	0.71575	93	H	-5.04426	3.066768	0.798285
73	H	-3.41138	-7.5381	-0.20698	94	H	-2.7143	2.153929	0.767658
74	H	-1.44328	-3.81502	-2.10383	95	H	-2.82673	3.610607	-0.19146
75	H	0.177553	-4.48024	-2.46017	96	H	-2.22166	5.661611	0.688859
76	H	-1.03746	-4.37468	-3.73872	97	H	-0.97485	5.745311	1.91236
77	H	-1.4949	-1.59328	1.550256	98	H	0.622197	4.521063	0.355684
78	H	-1.27488	-2.40527	-0.02025	99	H	-0.66198	4.44401	-0.8653
79	H	-1.2922	-3.35412	1.484307	100	H	0.001144	6.011994	-0.37488

Table S22. Calculated atomic coordinates for probe **B** with one added water molecule.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	2.434986	3.583459	-1.03909	17	C	-5.55165	-0.56687	0.230879
2	C	1.300491	4.200552	-0.48792	18	N	-5.96534	0.713555	-0.02393
3	C	0.307731	3.445429	0.108082	19	C	-7.32409	0.740283	-0.36214
4	C	0.432204	2.042674	0.182317	20	C	-7.85131	-0.55077	-0.27608
5	C	1.609214	1.418538	-0.29704	21	C	-6.75895	-1.50011	0.140046
6	C	2.581945	2.214158	-0.94295	22	C	-8.10681	1.827563	-0.74218
7	N	-0.59561	1.274048	0.698791	23	C	-9.45744	1.582785	-1.02152
8	C	-0.61465	-0.08591	0.506746	24	C	-9.99922	0.298464	-0.92811
9	C	0.552768	-0.74963	0.090221	25	C	-9.18807	-0.78276	-0.55181
10	C	1.723011	-0.01211	-0.16257	26	C	-7.06672	-2.12934	1.507558
11	C	-1.82112	-0.85516	0.771363	27	C	-6.53904	-2.59418	-0.91217
12	C	-1.58894	-2.28445	1.179396	28	C	-5.18892	1.917048	0.181532
13	C	-0.73667	-2.95651	0.105776	29	C	-1.55378	1.946117	1.584644
14	C	0.613694	-2.25611	0.000809	30	N	2.895265	-0.65802	-0.35614
15	C	-3.05348	-0.34063	0.459792	31	C	4.226083	-0.15708	-0.03296
16	C	-4.29496	-1.03383	0.528997	32	C	4.970894	-1.19979	0.80198

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
33	N	6.316101	-0.77167	1.113257	60	H	-5.7168	-3.25402	-0.61764
34	C	7.305909	-0.86246	0.190743	61	H	-7.44637	-3.19898	-1.012
35	O	8.398318	-0.20304	0.61412	62	H	-6.30338	-2.15868	-1.88821
36	O	7.208648	-1.48636	-0.86791	63	H	-4.56559	2.158344	-0.68509
37	C	9.649631	-0.1918	-0.16657	64	H	-4.55522	1.794551	1.062493
38	C	10.20473	-1.60509	-0.29642	65	H	-5.86533	2.751734	0.368492
39	C	10.5644	0.660229	0.7014	66	H	-0.99821	2.617771	2.241811
40	C	9.428897	0.476168	-1.51885	67	H	-2.29653	2.521305	1.027714
41	H	3.188054	4.176219	-1.55004	68	H	-2.05828	1.199388	2.1923
42	H	1.178851	5.278144	-0.55691	69	H	2.829901	-1.6679	-0.33369
43	H	-0.5846	3.937598	0.476232	70	H	4.801643	0.055017	-0.94099
44	H	3.441382	1.742883	-1.40427	71	H	4.138212	0.768064	0.541105
45	H	-1.06063	-2.32226	2.142916	72	H	4.44109	-1.38091	1.740776
46	H	-2.53366	-2.81473	1.314217	73	H	5.028608	-2.14801	0.26031
47	H	-0.57823	-4.01742	0.329754	74	H	6.450001	-0.1109	1.867462
48	H	-1.26676	-2.90148	-0.85315	75	H	11.20331	-1.55255	-0.74143
49	H	1.267224	-2.61474	0.81174	76	H	9.577968	-2.23358	-0.93169
50	H	1.102298	-2.54939	-0.93876	77	H	10.29493	-2.07268	0.68943
51	H	-3.0729	0.662518	0.050248	78	H	11.54562	0.745942	0.225804
52	H	-4.26958	-2.08569	0.798022	79	H	10.15325	1.666811	0.828628
53	H	-7.69869	2.829336	-0.83408	80	H	10.69748	0.20651	1.688711
54	H	-10.0912	2.413589	-1.32194	81	H	8.955268	1.454719	-1.38904
55	H	-11.0501	0.136035	-1.15265	82	H	10.40064	0.628869	-1.99886
56	H	-9.59991	-1.78677	-0.47916	83	H	8.810635	-0.13276	-2.18061
57	H	-6.25043	-2.78766	1.821959	84	O	3.323572	-4.55697	-0.01907
58	H	-7.20835	-1.35875	2.27175	85	H	3.367017	-3.78751	0.562476
59	H	-7.98338	-2.72469	1.440846	86	H	2.477917	-4.45999	-0.47425

Table S23. Calculated atomic coordinates for probe **BH**$^+$ with one added water molecule.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	3.301864	2.54198	1.17759	43	H	0.071965	2.755654	2.208429
2	C	2.129788	3.072343	1.734983	44	H	4.215575	0.868034	0.243681
3	C	0.971598	2.315857	1.795685	45	H	-1.5958	-3.5848	0.992738
4	C	0.95896	1.003665	1.288118	46	H	-2.59219	-3.24488	-0.41767
5	C	2.135212	0.462866	0.715001	47	H	-0.51466	-4.4845	-1.04674
6	C	3.300729	1.254431	0.6753	48	H	-0.58137	-2.8927	-1.81242
7	N	-0.19343	0.231811	1.349079	49	H	0.971981	-3.65602	0.709927
8	C	-0.31003	-0.95139	0.660338	50	H	1.662303	-3.30502	-0.86344
9	C	0.905622	-1.60332	0.228437	51	H	-2.50599	0.366057	0.299549
10	C	2.067903	-0.88828	0.249389	52	H	-4.21552	-2.17646	0.013794
11	C	-1.57658	-1.52715	0.36879	53	H	-6.42708	3.579206	-0.72697
12	C	-1.6413	-3.00224	0.06156	54	H	-8.89119	3.825978	-0.85527
13	C	-0.4883	-3.40788	-0.84814	55	H	-10.3768	1.852038	-0.70562
14	C	0.838476	-3.047	-0.19588	56	H	-9.40786	-0.43343	-0.41814
15	C	-2.68837	-0.69688	0.225709	57	H	-7.7193	-2.23779	-1.4759
16	C	-4.00054	-1.11297	-0.02259	58	H	-6.4472	-1.38595	-2.3779
17	C	-5.1263	-0.32024	-0.22737	59	H	-6.02811	-2.75318	-1.31942
18	N	-5.21408	1.01198	-0.4295	60	H	-7.87742	-1.98454	1.06384
19	C	-6.55928	1.420044	-0.51689	61	H	-6.18967	-2.51296	1.205843
20	C	-7.38373	0.300015	-0.41929	62	H	-6.7024	-0.97127	1.929318
21	C	-6.52619	-0.92252	-0.24476	63	H	-3.6888	2.234539	0.363423
22	C	-7.06802	2.705204	-0.66984	64	H	-3.34264	1.483609	-1.22264
23	C	-8.45987	2.835309	-0.73738	65	H	-4.48035	2.830399	-1.11081
24	C	-9.29944	1.720773	-0.65132	66	H	-0.7019	0.94715	3.239674
25	C	-8.76049	0.437132	-0.48941	67	H	-1.81676	1.48591	1.958705
26	C	-6.68411	-1.88486	-1.43332	68	H	-1.86022	-0.19626	2.535572
27	C	-6.83817	-1.64144	1.075077	69	H	3.150544	-2.18555	-0.97528
28	C	-4.11513	1.939893	-0.6009	70	H	3.966877	-0.83795	-0.62705
29	C	-1.21257	0.651226	2.321286	71	H	4.083865	-1.53803	1.769218
30	N	3.303321	-1.51117	-0.18877	72	H	3.456257	-3.09595	1.179753
31	C	4.044944	-2.21578	0.914325	73	H	5.798188	-3.38581	1.153746
32	C	5.437351	-2.62473	0.460069	74	H	5.391431	-3.08728	-0.53222
33	N	6.418692	-1.55429	0.463149	75	H	7.186449	-1.59336	1.119655
34	C	6.442082	-0.58945	-0.47646	76	H	8.097605	1.988277	-3.18507
35	O	7.532079	0.171763	-0.37018	77	H	6.888056	0.698545	-3.05625
36	O	5.54442	-0.44096	-1.32311	78	H	8.612163	0.335811	-2.80016
37	C	7.695799	1.412765	-1.16165	79	H	9.254809	2.891894	-1.14463
38	C	7.823487	1.078392	-2.64199	80	H	8.92819	2.178747	0.447909
39	C	9.007032	1.963437	-0.62241	81	H	9.822436	1.250413	-0.78008
40	C	6.551901	2.377322	-0.87305	82	H	6.429309	2.513086	0.20619
41	H	4.208143	3.138521	1.131249	83	H	6.795464	3.349136	-1.31348
42	H	2.119456	4.089675	2.115872	84	H	5.607644	2.035266	-1.29955

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
85	O	3.426607	-3.0187	-2.51174	87	H	3.354667	-3.97902	-2.43101
86	H	4.36423	-2.8545	-2.68339					

Table S24. Calculated atomic coordinates for probe **C** with one added water molecule near center N atom.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	-4.79902	2.668164	0.524227	37	H	-3.93866	4.023021	-0.87326
2	C	-4.56964	1.586797	1.389516	38	H	2.711859	1.948741	0.042934
3	C	-3.30733	1.036221	1.508172	39	H	3.252593	0.516697	-0.82642
4	C	-2.22886	1.565378	0.769048	40	H	2.949981	2.405798	-2.43257
5	C	-2.43365	2.703698	-0.04891	41	H	1.760401	1.139013	-2.7489
6	C	-3.74488	3.213143	-0.18122	42	H	1.388273	3.821887	-1.35861
7	N	-0.98295	0.965492	0.822855	43	H	0.537233	3.20208	-2.76793
8	C	-0.01208	1.288178	-0.09397	44	H	0.124047	-1.31179	0.096496
9	C	-0.1509	2.442504	-0.88242	45	H	3.102529	-1.42079	-0.7198
10	C	-1.30099	3.24507	-0.7585	46	H	-0.19019	-6.44787	0.833811
11	C	1.18212	0.466117	-0.23	47	H	0.855586	-8.65232	0.393286
12	C	2.409052	1.198238	-0.70135	48	H	3.108899	-8.81299	-0.6227
13	C	2.073734	1.894507	-2.01771	49	H	4.366059	-6.7352	-1.21792
14	C	0.957032	2.908258	-1.79657	50	H	4.360433	-4.49679	-2.61028
15	C	1.100632	-0.89853	-0.12577	51	H	2.675044	-3.99446	-2.86155
16	C	2.155987	-1.82254	-0.3711	52	H	3.911057	-2.78808	-2.44232
17	C	2.136324	-3.1927	-0.28325	53	H	5.369818	-4.45212	-0.26328
18	N	1.166185	-4.0198	0.217138	54	H	4.917872	-2.74252	-0.11119
19	C	1.526254	-5.36356	0.059885	55	H	4.387732	-3.91472	1.116365
20	C	2.79368	-5.44213	-0.52258	56	H	0.256295	-2.7846	1.628178
21	C	3.304787	-4.04815	-0.77427	57	H	-0.82214	-3.30062	0.301271
22	C	0.802107	-6.50404	0.396905	58	H	-0.3483	-4.44377	1.572013
23	C	1.399184	-7.74505	0.141302	59	H	-1.10536	0.591325	2.871932
24	C	2.669479	-7.83725	-0.43317	60	H	-1.14398	-0.88392	1.875892
25	C	3.377013	-6.67322	-0.76974	61	H	0.3759	0.011574	2.095594
26	C	3.575662	-3.81508	-2.2661	62	H	-0.51437	4.678361	-1.90665
27	C	4.57228	-3.7691	0.047446	63	H	-2.97824	5.768871	-1.5959
28	C	-0.00474	-3.60918	0.960419	64	H	-2.44784	5.494226	0.072595
29	C	-0.70048	0.108701	1.980571	65	H	-0.37193	6.748128	-0.34375
30	N	-1.33901	4.450201	-1.36649	66	H	-0.78454	6.924474	-2.05435
31	C	-2.09937	5.625891	-0.95493	67	H	-1.36076	8.84841	-0.64153
32	C	-1.20989	6.858826	-1.03996	68	H	-2.66804	8.2365	-1.38578
33	N	-1.97835	8.041901	-0.66344	69	O	1.981204	6.023879	-3.12993
34	H	-5.80114	3.067731	0.399078	70	H	2.256085	5.102229	-3.21343
35	H	-5.39318	1.152804	1.95003	71	H	1.384483	6.026	-2.37058
36	H	-3.16568	0.167889	2.140427					

Table S25. Calculated atomic coordinates for probe **CH**$^{+}$ with one added water molecule near center N atom.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	-2.76737	2.319962	3.801978	37	H	-2.32518	4.029546	2.614578
2	C	-2.56866	0.943398	3.985979	38	H	2.62091	0.785269	-1.50035
3	C	-1.75411	0.225672	3.127729	39	H	1.884912	-0.11682	-2.82078
4	C	-1.12233	0.873907	2.050024	40	H	1.685543	2.336353	-3.22592
5	C	-1.33675	2.257909	1.842493	41	H	0.095051	1.591968	-3.029
6	C	-2.16064	2.965692	2.742199	42	H	1.684909	3.21084	-0.98067
7	N	-0.27823	0.175055	1.197765	43	H	0.151633	3.626463	-1.7347
8	C	0.16245	0.706338	0.012113	44	H	-0.50807	-1.73675	-0.50595
9	C	0.050457	2.132163	-0.17895	45	H	1.578196	-2.04502	-2.74964
10	C	-0.66914	2.854493	0.730948	46	H	-2.48845	-6.49225	-1.23337
11	C	0.728344	-0.10938	-1.01073	47	H	-2.19986	-8.68721	-2.34308
12	C	1.663781	0.553326	-1.98794	48	H	-0.38958	-9.05348	-3.99162
13	C	1.028254	1.83647	-2.50675	49	H	1.186817	-7.19877	-4.55959
14	C	0.741282	2.786171	-1.35139	50	H	1.353342	-4.68457	-5.48148
15	C	0.282201	-1.41745	-1.17029	51	H	-0.13528	-3.80985	-5.06173
16	C	0.736432	-2.33404	-2.1281	52	H	1.448176	-3.02579	-4.8553
17	C	0.292212	-3.62968	-2.36304	53	H	2.938838	-5.47142	-3.63601
18	N	-0.7542	-4.29632	-1.82456	54	H	3.036169	-3.80588	-3.03355
19	C	-0.80478	-5.62129	-2.29928	55	H	2.564154	-5.12234	-1.93473
20	C	0.225265	-5.81675	-3.2189	56	H	-1.31789	-3.63552	0.09456
21	C	1.009857	-4.54149	-3.35194	57	H	-2.10324	-2.79824	-1.27681
22	C	-1.69394	-6.63544	-1.95876	58	H	-2.58391	-4.44292	-0.85143
23	C	-1.52244	-7.87347	-2.58865	59	H	0.517762	-0.95005	2.761227
24	C	-0.49971	-8.08138	-3.51857	60	H	-0.45188	-1.91495	1.618755
25	C	0.386661	-7.04444	-3.83984	61	H	1.165088	-1.3361	1.155905
26	C	0.908429	-3.97457	-4.77718	62	H	-1.7532	4.645252	0.727555
27	C	2.479534	-4.74495	-2.9583	63	H	-0.56052	4.550764	-0.39147
28	C	-1.7386	-3.75965	-0.90848	64	H	-0.0319	4.796631	2.489871
29	C	0.261291	-1.09253	1.710237	65	H	1.20164	4.675672	1.213209
30	N	-0.76457	4.298446	0.580735	66	H	0.332991	6.743555	0.148667
31	C	0.203119	5.047001	1.453268	67	H	-0.95903	6.851485	1.350892
32	C	0.08325	6.537535	1.19485	68	H	1.058626	8.218435	1.788861
33	N	1.038356	7.236453	2.048598	69	H	0.711798	7.206345	3.011567
34	H	-3.40107	2.877956	4.484759	70	O	-3.31267	5.311974	0.747761
35	H	-3.06064	0.427489	4.805618	71	H	-3.50512	5.709386	-0.11235
36	H	-1.63187	-0.83988	3.279969	72	H	-3.96815	4.608703	0.852231

Table S26. Calculated atomic coordinates for probe **C** with one added water molecule near outer N atom.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	4.675669	3.44776	-0.60515	37	H	5.6108	1.681913	-1.3382
2	C	3.573419	3.983022	0.07995	38	H	0.948589	-2.866	1.422678
3	C	2.563606	3.158787	0.540318	39	H	-0.52885	-3.09596	0.489657
4	C	2.638357	1.764958	0.339541	40	H	1.387535	-4.16013	-0.70061
5	C	3.78611	1.206745	-0.2738	41	H	0.771784	-2.79066	-1.63035
6	C	4.775236	2.081372	-0.77671	42	H	3.278715	-2.96275	0.094045
7	N	1.589248	0.946212	0.717368	43	H	3.16478	-2.56549	-1.61626
8	C	1.519157	-0.35086	0.266826	44	H	-0.88913	0.586606	0.015783
9	C	2.660401	-0.9624	-0.28048	45	H	-2.24262	-2.15099	0.484404
10	C	3.853835	-0.22779	-0.4095	46	H	-5.4667	2.997687	-0.82903
11	C	0.281705	-1.10525	0.385582	47	H	-7.93333	2.797013	-0.79891
12	C	0.442341	-2.59725	0.484372	48	H	-9.03195	0.636461	-0.28563
13	C	1.282301	-3.06947	-0.70004	49	H	-7.64631	-1.38448	0.213378
14	C	2.664543	-2.42806	-0.64801	50	H	-5.75229	-3.0119	-0.71067
15	C	-0.92486	-0.47676	0.210462	51	H	-4.70156	-2.10669	-1.82125
16	C	-2.20692	-1.09277	0.242022	52	H	-3.99089	-3.21546	-0.62713
17	C	-3.44694	-0.53176	0.049578	53	H	-5.79937	-2.41789	1.773994
18	N	-3.79095	0.74685	-0.2985	54	H	-4.03989	-2.63428	1.847689
19	C	-5.18339	0.899271	-0.33392	55	H	-4.77065	-1.10871	2.393477
20	C	-5.79274	-0.32181	-0.03589	56	H	-2.36477	2.192248	0.271192
21	C	-4.72622	-1.35126	0.226105	57	H	-2.13949	1.479547	-1.35208
22	C	-5.9314	2.041409	-0.60929	58	H	-3.4347	2.642632	-1.06816
23	C	-7.32638	1.920162	-0.58722	59	H	1.259322	1.979306	2.498511
24	C	-7.94729	0.702706	-0.29713	60	H	-0.0642	2.158053	1.320675
25	C	-7.17275	-0.43276	-0.01663	61	H	0.150836	0.63053	2.202742
26	C	-4.79319	-2.49119	-0.80067	62	H	4.902835	-1.86203	-0.88422
27	C	-4.83696	-1.91023	1.651198	63	H	6.900555	-0.10945	-1.26828
28	C	-2.87785	1.820105	-0.62212	64	H	6.320051	0.350443	0.341442
29	C	0.670099	1.46442	1.737496	65	H	6.609822	-1.94533	1.157699
30	N	4.999266	-0.8659	-0.73409	66	H	7.038629	-2.50993	-0.46485
31	C	6.356949	-0.46494	-0.38354	67	H	8.957672	-2.03701	0.968926
32	C	7.096118	-1.65169	0.221653	68	H	8.974856	-1.08549	-0.35209
33	N	8.476751	-1.26294	0.517845	69	O	8.334299	0.935251	2.149901
34	H	5.441608	4.103204	-1.00923	70	H	8.435509	0.130318	1.562546
35	H	3.489953	5.056819	0.224066	71	H	9.088356	0.905859	2.750032
36	H	1.694736	3.598555	1.015398					

Table S27. Calculated atomic coordinates for probe **CH**$^{+}$ with one added water molecule near outer N atom.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	4.400427	3.978598	-0.53978	37	H	5.591976	2.306328	-1.0631
2	C	3.189076	4.428833	0.006903	38	H	1.151758	-2.6834	1.593914
3	C	2.241415	3.525234	0.450059	39	H	-0.24111	-2.97744	0.555678
4	C	2.475242	2.13927	0.340124	40	H	1.812814	-3.88876	-0.52796
5	C	3.691018	1.671802	-0.21367	41	H	1.191901	-2.5232	-1.46294
6	C	4.643919	2.62245	-0.63967	42	H	3.499978	-2.59264	0.517487
7	N	1.527124	1.236346	0.787519	43	H	3.610452	-2.21254	-1.19273
8	C	1.611385	-0.09398	0.455629	44	H	-0.84513	0.652829	0.092738
9	C	2.84708	-0.6182	-0.00443	45	H	-2.02002	-2.14502	0.638948
10	C	3.914972	0.245506	-0.2641	46	H	-5.42414	2.736359	-1.14198
11	C	0.445111	-0.94026	0.541199	47	H	-7.88635	2.462161	-1.08549
12	C	0.695375	-2.42102	0.628929	48	H	-8.91421	0.326329	-0.36739
13	C	1.643603	-2.80633	-0.50367	49	H	-7.46744	-1.59394	0.317516
14	C	2.975656	-2.0938	-0.30772	50	H	-5.50626	-3.23741	-0.46452
15	C	-0.80491	-0.40508	0.312315	51	H	-4.4825	-2.40066	-1.65144
16	C	-2.03438	-1.10494	0.327672	52	H	-3.73914	-3.37005	-0.35893
17	C	-3.2991	-0.61821	0.069398	53	H	-5.58178	-2.42484	1.958173
18	N	-3.68157	0.599677	-0.40844	54	H	-3.81551	-2.56721	2.049417
19	C	-5.08028	0.703735	-0.4532	55	H	-4.60499	-1.02723	2.456181
20	C	-5.6477	-0.50174	-0.03642	56	H	-2.34131	2.195367	-0.06493
21	C	-4.54636	-1.46379	0.31966	57	H	-2.03166	1.218725	-1.52803
22	C	-5.86089	1.791619	-0.83324	58	H	-3.38903	2.347544	-1.48944
23	C	-7.25114	1.629027	-0.79524	59	H	1.079273	2.358014	2.487824
24	C	-7.83218	0.424902	-0.38883	60	H	-0.25427	2.301838	1.309238
25	C	-7.02371	-0.65443	-0.0035	61	H	0.126496	0.875	2.29689
26	C	-4.56548	-2.69403	-0.60041	62	H	5.760686	0.500961	-1.01005
27	C	-4.63888	-1.89456	1.789664	63	H	6.68232	-0.71538	0.654076
28	C	-2.80806	1.64766	-0.89022	64	H	5.236243	-1.69269	0.827456
29	C	0.551762	1.726164	1.770483	65	H	5.738533	-2.83663	-1.35674
30	N	5.141662	-0.22015	-0.66694	66	H	7.156705	-1.79059	-1.56931
31	C	5.88638	-1.22035	0.091256	67	H	7.756657	-3.92844	-0.68841
32	C	6.500564	-2.25841	-0.83214	68	H	8.077526	-2.73439	0.448396
33	N	7.325049	-3.21549	-0.05058	69	H	6.759056	-3.70529	0.646593
34	H	5.145625	4.690183	-0.88296	70	O	8.382654	-5.04648	-1.82473
35	H	2.984942	5.493539	0.079392	71	H	9.319628	-4.88074	-1.99391
36	H	1.303955	3.892485	0.850505	72	H	7.950644	-4.91792	-2.67977

Table S28. Calculated atomic coordinates for probe **D** with one added water molecule.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	-4.10989	0.166512	3.304272	43	C	-0.05148	6.579578	2.130322
2	C	-4.64733	-1.11648	3.11375	44	C	0.333798	7.070993	0.908806
3	C	-3.83324	-2.17514	2.756394	45	C	1.619472	6.758485	0.355282
4	C	-2.44589	-1.98036	2.595214	46	C	2.483384	5.926223	1.112611
5	C	-1.88291	-0.70948	2.865042	47	C	1.12904	9.504916	-1.42773
6	C	-2.74885	0.361708	3.179644	48	C	1.091756	8.007161	-1.70549
7	N	-1.63859	-3.01422	2.154051	49	N	1.997378	7.245836	-0.84955
8	C	-0.35748	-2.76607	1.722418	50	C	3.34115	7.038956	-1.38057
9	C	0.259901	-1.5412	2.033724	51	C	3.473929	5.746581	-2.1774
10	C	-0.45542	-0.558	2.738835	52	H	-4.76021	1.004895	3.536058
11	C	0.379492	-3.77422	0.977895	53	H	-5.71699	-1.27961	3.213676
12	C	1.875662	-3.71426	1.12184	54	H	-4.27849	-3.14295	2.559428
13	C	2.336136	-2.30482	0.757282	55	H	-2.34809	1.361998	3.290769
14	C	1.714992	-1.29074	1.711277	56	H	2.168734	-3.93884	2.157426
15	C	-0.2701	-4.56514	0.064407	57	H	2.362453	-4.45438	0.483613
16	C	0.32908	-5.54858	-0.77055	58	H	3.427813	-2.22128	0.799449
17	C	-0.24944	-6.34633	-1.72859	59	H	2.03217	-2.08803	-0.27436
18	N	-1.5303	-6.3535	-2.21046	60	H	2.276207	-1.30267	2.659331
19	C	-1.70795	-7.38146	-3.1456	61	H	1.836205	-0.28169	1.294622
20	C	-0.49931	-8.05708	-3.32981	62	H	-1.33684	-4.41194	-0.03267
21	C	0.548119	-7.43861	-2.44272	63	H	1.386998	-5.74537	-0.62449
22	C	-2.86329	-7.74828	-3.83094	64	H	-3.81006	-7.23906	-3.68039
23	C	-2.768	-8.81875	-4.72888	65	H	-3.65546	-9.12577	-5.27679
24	C	-1.56272	-9.49641	-4.92903	66	H	-1.51661	-10.3246	-5.63118
25	C	-0.41397	-9.11378	-4.22046	67	H	0.528118	-9.63732	-4.36637
26	C	1.687535	-6.83	-3.27363	68	H	2.190301	-7.61929	-3.8423
27	C	1.105132	-8.46406	-1.44627	69	H	1.305625	-6.08446	-3.97801
28	C	-2.58014	-5.42205	-1.86381	70	H	2.426422	-6.35054	-2.62357
29	C	-2.14854	-4.38016	2.323947	71	H	1.596875	-9.27713	-1.99046
30	N	0.192164	0.53625	3.198545	72	H	1.842574	-8.00056	-0.78329
31	C	-0.13761	1.332849	4.371778	73	H	0.305169	-8.89124	-0.8336
32	C	1.12078	1.461083	5.233087	74	H	-2.98123	-5.62186	-0.8644
33	N	0.975702	2.448991	6.284997	75	H	-2.20182	-4.39813	-1.90679
34	C	0.947065	3.785488	6.118181	76	H	-3.39056	-5.51042	-2.58622
35	O	0.563868	4.549545	7.025147	77	H	-2.63079	-4.44685	3.30078
36	C	1.350243	4.349708	4.798491	78	H	-2.86682	-4.65334	1.547133
37	C	0.479246	5.188952	4.14121	79	H	-1.31321	-5.07515	2.299097
38	C	0.802535	5.749112	2.892651	80	H	1.171126	0.585402	2.946416
39	C	2.06644	5.452883	2.336499	81	H	-0.48677	2.330902	4.082655
40	O	2.945007	4.653647	3.023437	82	H	-0.93007	0.844835	4.94246
41	C	2.645212	4.08799	4.240729	83	H	1.352307	0.50439	5.706509
42	O	3.541592	3.40625	4.747921	84	H	1.975811	1.719926	4.607528

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
85	H	0.63873	2.127869	7.185806	94	H	0.076943	7.61352	-1.60294
86	H	-0.49055	5.405803	4.584375	95	H	3.567683	7.900267	-2.0165
87	H	-1.02849	6.832396	2.534476	96	H	4.063077	7.070112	-0.55944
88	H	-0.34563	7.719804	0.369386	97	H	4.490648	5.65077	-2.57314
89	H	3.459938	5.630317	0.750074	98	H	3.266019	4.870893	-1.55415
90	H	0.45955	10.03271	-2.11539	99	H	2.777747	5.738003	-3.02298
91	H	0.812324	9.727037	-0.40347	100	O	4.195736	1.298685	2.940797
92	H	2.140482	9.901683	-1.56658	101	H	3.539989	0.633764	3.184936
93	H	1.390582	7.806608	-2.73913	102	H	4.045566	2.031456	3.565146

Table S29. Calculated atomic coordinates for probe **DH**$^{+}$ with one added water molecule.

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
1	C	5.853032	0.972263	0.365785	31	C	4.761133	-0.66016	5.015921
2	C	4.823409	1.173409	-0.56219	32	C	4.767737	0.83146	4.700814
3	C	3.606998	0.526949	-0.41948	33	N	4.253665	1.570989	5.834983
4	C	3.392507	-0.34946	0.660954	34	C	2.945004	1.726094	6.114126
5	C	4.426691	-0.555	1.605244	35	O	2.547489	2.132168	7.219559
6	C	5.65247	0.117385	1.434174	36	C	1.958996	1.39865	5.039394
7	N	2.177151	-1.00357	0.808409	37	C	1.101766	0.338036	5.172432
8	C	2.004811	-2.03153	1.714662	38	C	0.108613	0.07995	4.203228
9	C	2.94273	-2.08456	2.822407	39	C	-0.03061	0.988429	3.132684
10	C	4.110293	-1.39103	2.724192	40	O	0.843587	2.040346	3.003283
11	C	0.975433	-2.99575	1.60393	41	C	1.864247	2.271917	3.900384
12	C	0.680678	-3.92005	2.765779	42	O	2.604117	3.221914	3.653091
13	C	1.912962	-4.20871	3.608314	43	C	-0.77891	-1.01673	4.228085
14	C	2.560299	-2.89936	4.025134	44	C	-1.72714	-1.19294	3.25039
15	C	0.279497	-3.18326	0.396233	45	C	-1.87617	-0.24652	2.188113
16	C	-0.82106	-4.02289	0.244219	46	C	-1.00985	0.874187	2.171021
17	C	-1.55356	-4.32288	-0.90738	47	C	-4.84159	-1.75301	1.815501
18	N	-1.50072	-3.76076	-2.12744	48	C	-3.52249	-1.68712	1.056142
19	C	-2.37609	-4.41037	-3.02128	49	N	-2.81171	-0.42386	1.222077
20	C	-3.05761	-5.41643	-2.33862	50	C	-3.15408	0.62643	0.269976
21	C	-2.59816	-5.43153	-0.90804	51	C	-2.36004	0.564718	-1.02927
22	C	-2.58164	-4.1559	-4.37241	52	H	6.805407	1.480727	0.250279
23	C	-3.52343	-4.95282	-5.0324	53	H	4.975875	1.834098	-1.41097
24	C	-4.22238	-5.96007	-4.35986	54	H	2.839211	0.682251	-1.16722
25	C	-3.99124	-6.19879	-2.99863	55	H	6.466469	-0.0111	2.140974
26	C	-1.96048	-6.77728	-0.53324	56	H	-0.10468	-3.48953	3.400168
27	C	-3.75996	-5.10162	0.04382	57	H	0.28572	-4.86376	2.380416
28	C	-0.71482	-2.60641	-2.51079	58	H	1.636916	-4.78712	4.496125
29	C	1.049609	-0.4242	0.072984	59	H	2.632332	-4.80865	3.035688
30	N	5.086204	-1.48509	3.806459	60	H	1.839233	-2.31107	4.60849

Row	Symbol	X	Y	Z	Row	Symbol	X	Y	Z
61	H	3.409533	-3.08805	4.681831	83	H	5.504893	-0.91314	5.774664
62	H	0.675588	-2.69544	-0.48312	84	H	5.780183	1.181196	4.486737
63	H	-1.18162	-4.54811	1.122824	85	H	4.154888	1.048208	3.824311
64	H	-2.03912	-3.38173	-4.90562	86	H	4.889673	1.756921	6.603435
65	H	-3.7091	-4.78325	-6.08973	87	H	1.185698	-0.32149	6.033878
66	H	-4.94732	-6.56424	-4.89868	88	H	-0.70037	-1.73128	5.043532
67	H	-4.52933	-6.98246	-2.47124	89	H	-2.38549	-2.05167	3.306475
68	H	-2.70769	-7.57092	-0.63081	90	H	-1.03688	1.604261	1.371188
69	H	-1.11712	-7.00983	-1.19041	91	H	-4.68599	-1.65429	2.894684
70	H	-1.60536	-6.76216	0.501585	92	H	-5.5201	-0.9554	1.494423
71	H	-4.50768	-5.89821	-0.02105	93	H	-5.33487	-2.71289	1.627803
72	H	-3.41435	-5.03751	1.079757	94	H	-2.86126	-2.50695	1.345796
73	H	-4.23609	-4.15673	-0.22986	95	H	-3.70065	-1.80985	-0.01725
74	H	-0.76797	-1.85936	-1.71953	96	H	-4.22351	0.523729	0.056352
75	H	0.327036	-2.88301	-2.69567	97	H	-3.03351	1.598724	0.756217
76	H	-1.1352	-2.17458	-3.41746	98	H	-1.28856	0.697789	-0.85326
77	H	1.094488	0.660068	0.185492	99	H	-2.51208	-0.39363	-1.53557
78	H	1.077119	-0.67714	-0.98891	100	H	-2.69366	1.3587	-1.70634
79	H	0.124652	-0.78186	0.512334	101	O	5.686482	-4.10174	4.299784
80	H	5.223861	-2.50036	4.079495	102	H	4.986505	-4.69154	3.986118
81	H	6.012013	-1.20874	3.466643	103	H	5.766132	-4.29225	5.244209
82	H	3.776384	-0.971	5.36649					

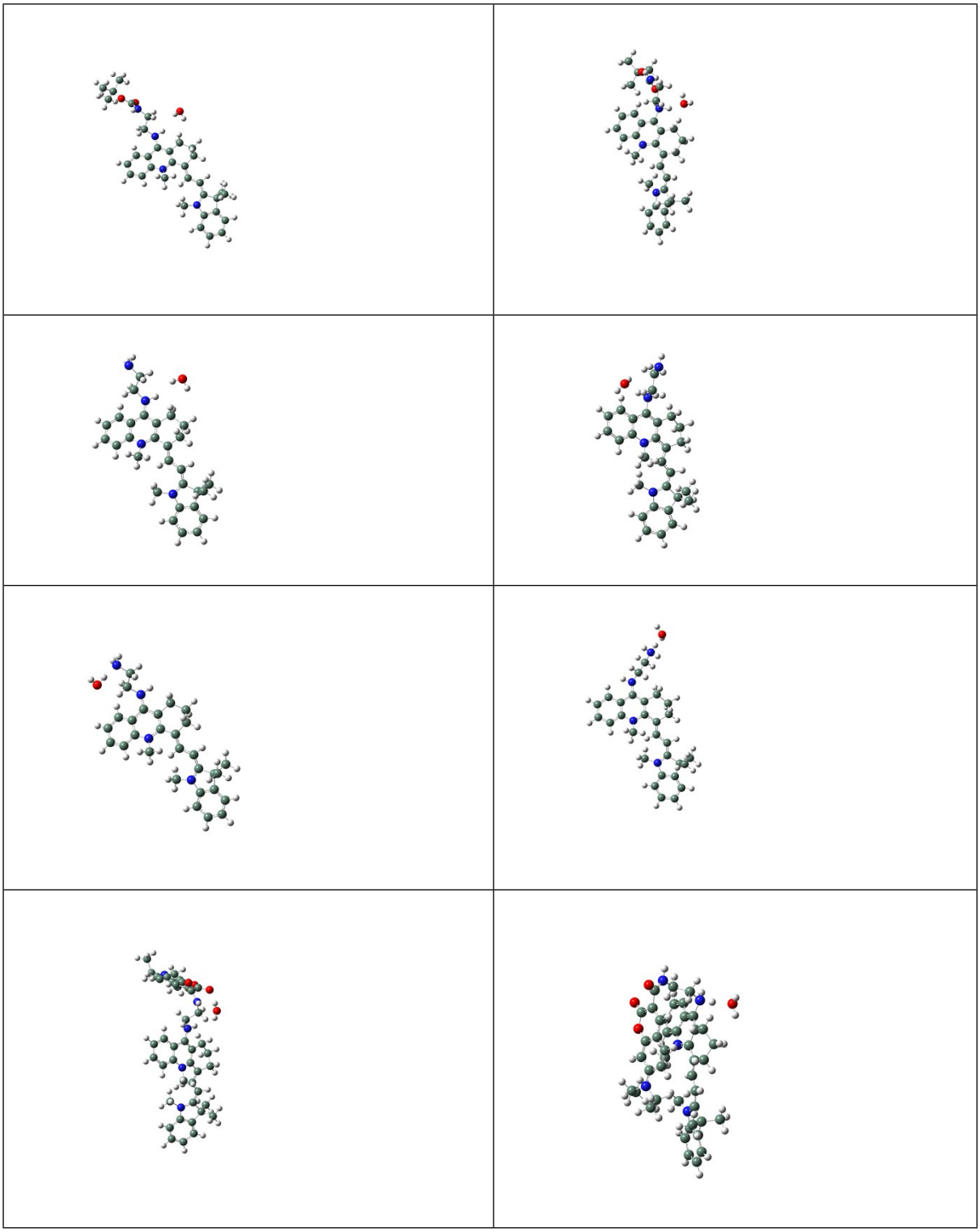


Figure S54. Drawings of the probes showing the orientation of the added water molecule. **B**, **C** (water in middle), **C** (water at the end) and **D** from top to bottom on the left side and on the right are the corresponding protonated probes.

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