Unexpected changes in the population of coordination isomers for the lanthanide ion complexes of DOTMA-tetraglycinate

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HPLC profiles

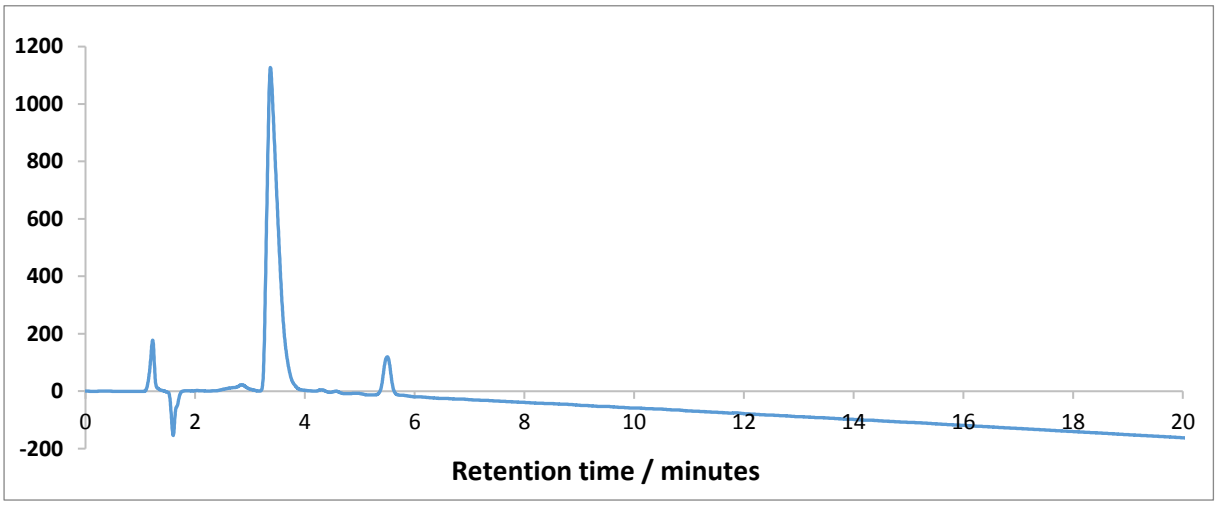

Figure S1: Analytical HPLC profile for DOTMA-Gly_4 (scheme 1); solvent gradient conditions - 98% Water (1% TFA): 2% ACN to 40% water (1% TFA): 60% ACN in 30 min, 215 nm.

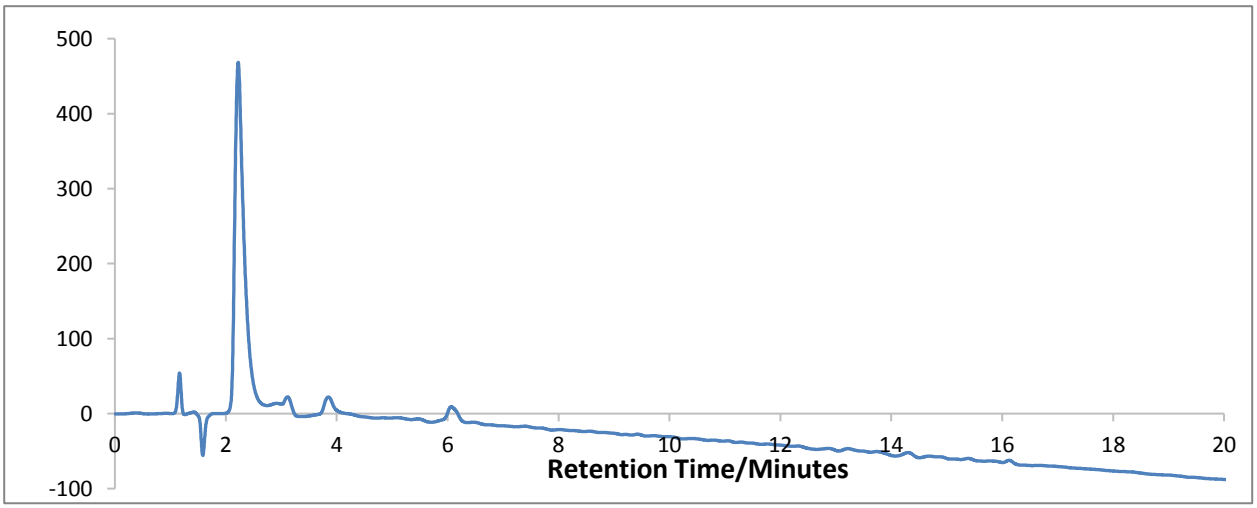

Figure S2: Analytical HPLC profile for ErDOTMA-Gly_4; solvent gradient conditions - 98% Water (1% TFA): 2% ACN to 40% water (1% TFA): 60% ACN in 30 min.

NMR Measurements ^{1}H NMR spectra and CEST spectra were recorded on a 400 MHz Bruker Avance III spectrometer. High resolution ^{1}H NMR samples were prepared either in D_2O or H_2O. Samples for studies were prepared by dissolving the appropriate amount of agent in water and the pH was adjusted to neutral pH.

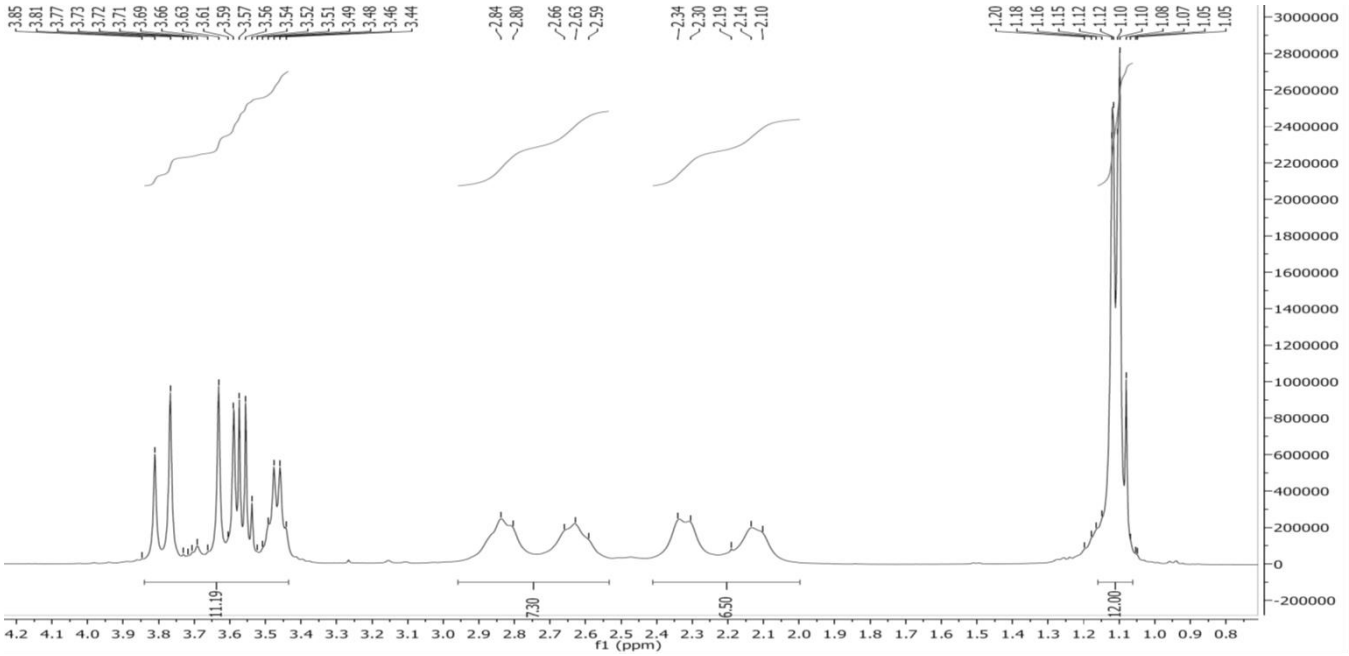

Figure S3. ^{1}H NMR spectra of the ethyl ester of DOTMA-(gly)$_4$.

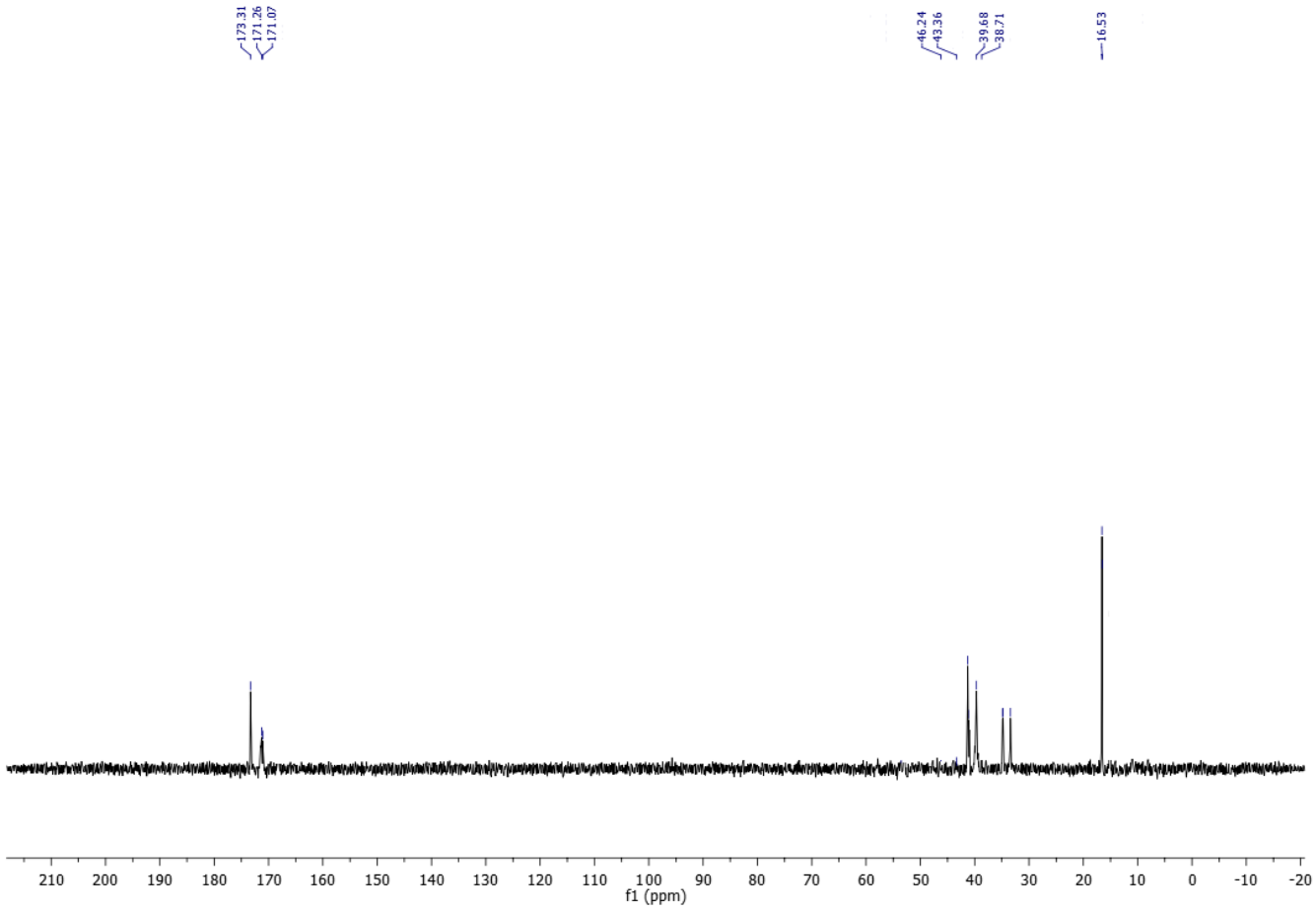

Figure S4. ^{13}C NMR spectra of the ethyl ester of DOTMA-(gly)$_4$.

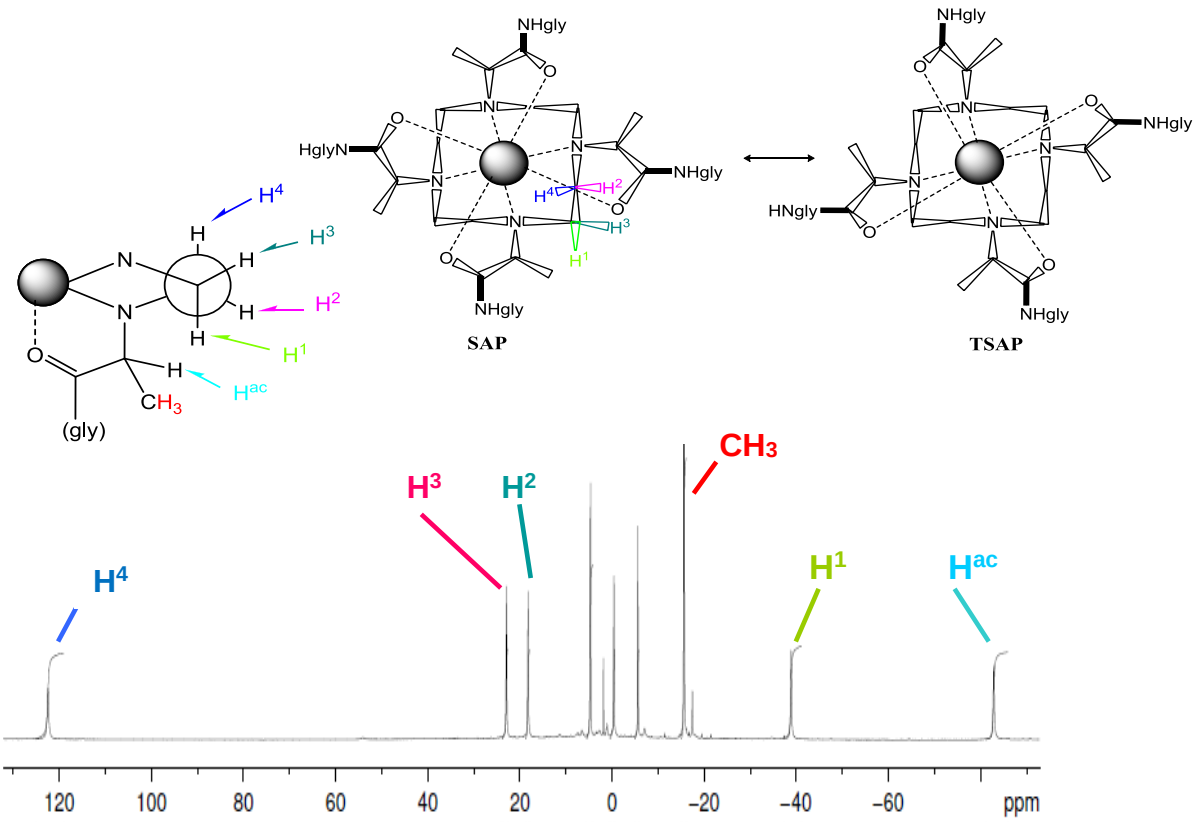

Figure S5. **Top**: A description of lanthanide coordination in the SAP and TSAP geometry isomers of the macrocyclic ring protons in the LnDOTMA-(gly)$_4$ complexes. **Bottom**: Assignment of the peaks in the ^{1}H NMR spectrum of YbDOTMA-(gly)$_4$. The spectrum was recorded in H_2O at 298 K (pH 7.4). The notation scheme H^1 and H^4 for *axial* protons, H^2 and H^3 for *equatorial* protons of macrocyclic ring and H^{ac} for an acetamide proton was used.

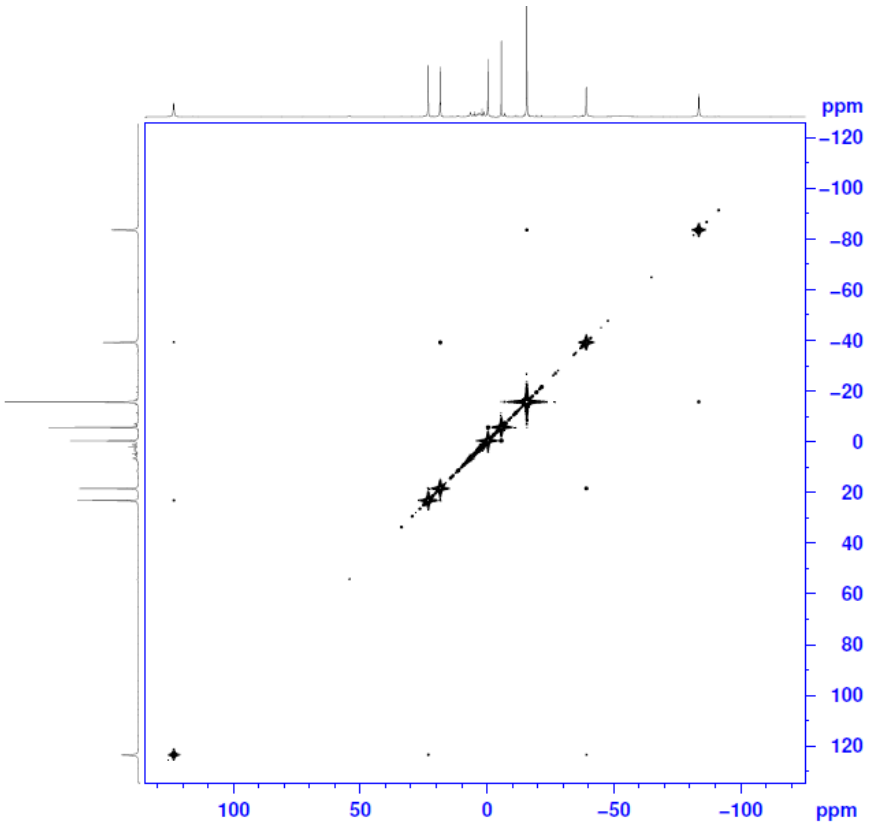

Figure S6. ^{1}H COSY spectrum of YbDOTMA-(gly)$_4$ recorded at 400 MHz and 298 K in D_2O (pD 7.4).

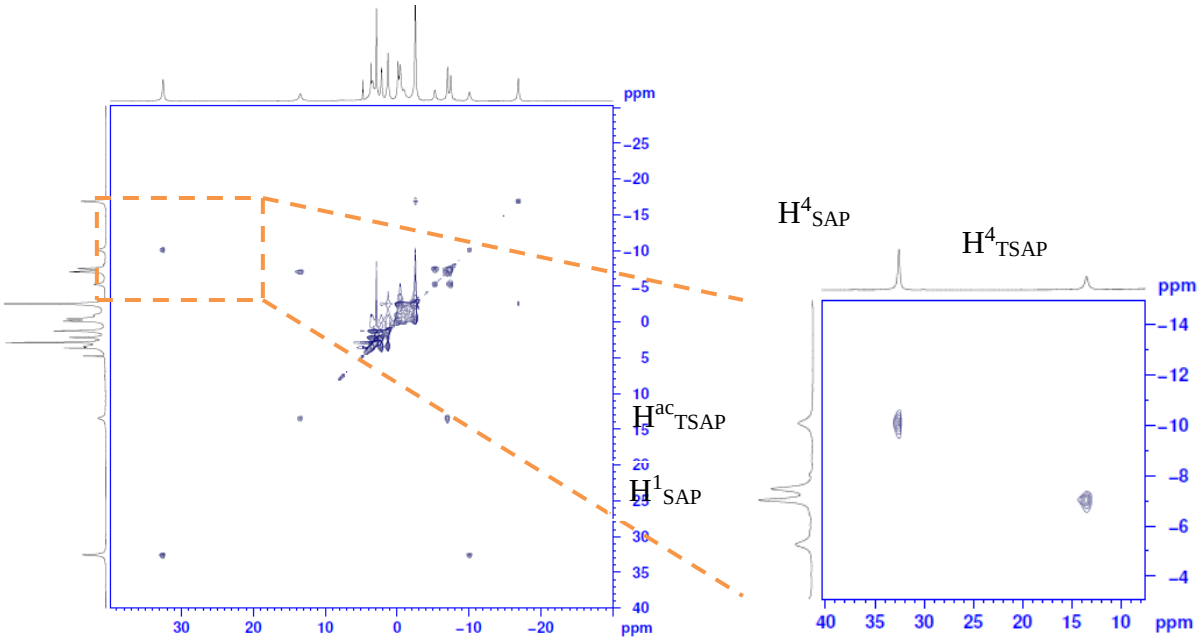

Figure S7. ^{1}H EXSY spectrum of EuDOTMA-(gly)$_4$ recorded at 400 MHz and 298 K in D_2O (pD 7.4). EXSY spectrum shows the SAP and TSAP transition of cyclen protons.

CH_3

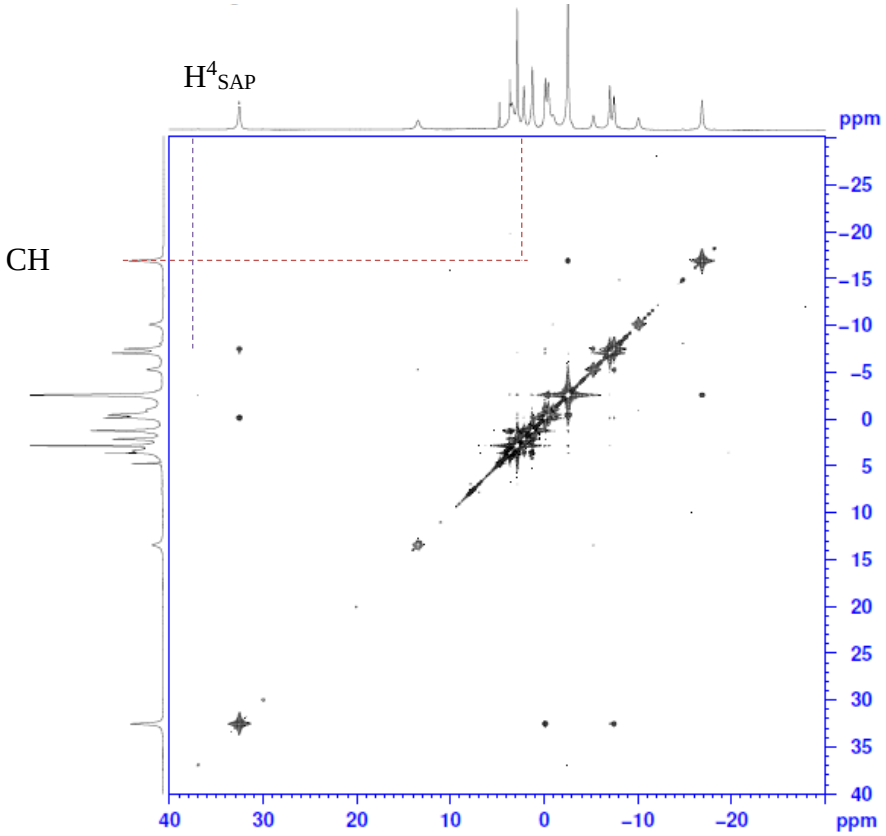

Figure S8. ^{1}H COSY spectrum of EuDOTMA-(gly)$_4$ recorded at 400 MHz and 298 K in D_2O (pD 7.4).

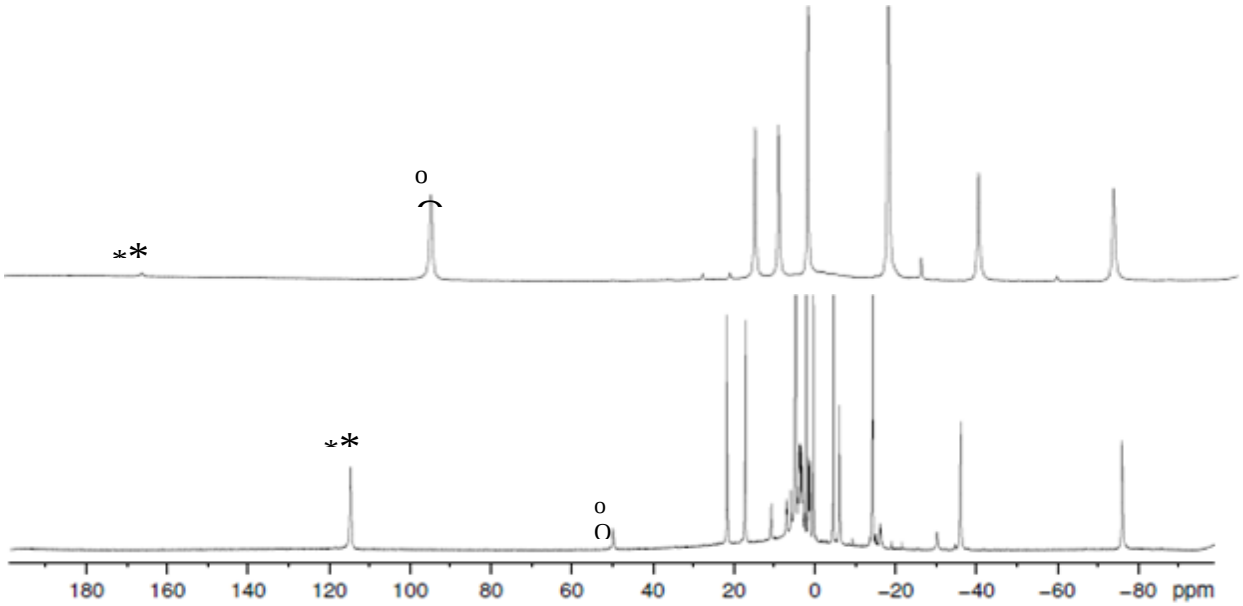

Figure S9. ^{1}H NMR spectra of YbDOTMA (Top) and YbDOTMA-(gly)$_4$ (bottom). All spectra were recorded in D_2O at 298 K and 400 MHz. (*) is denoted for the axial H^4 protons of the SAP isomer and (o) is for the H^4 protons of the TSAP isomer.

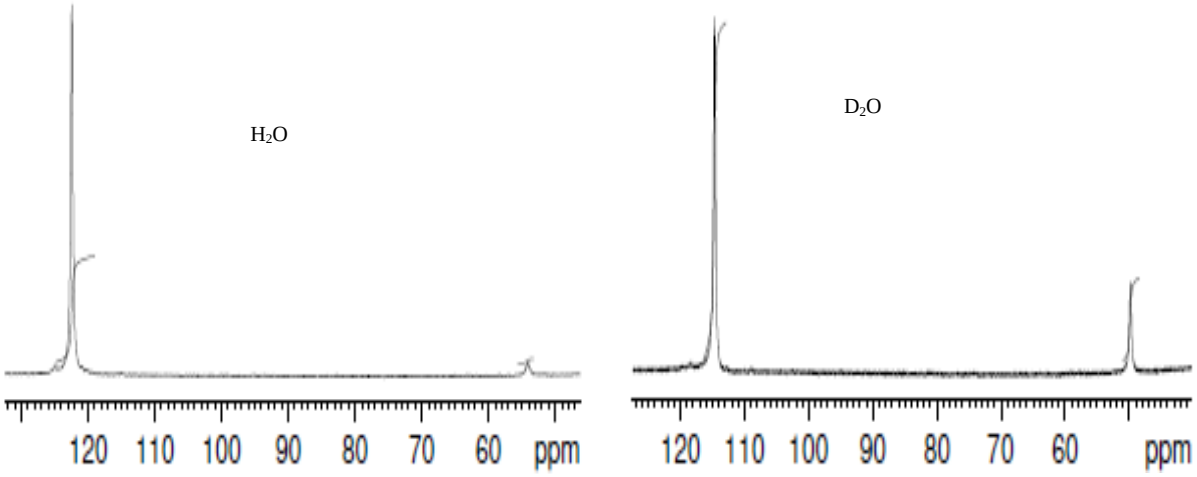

Figure S10. ^{1}H NMR spectra of YbDOTMA-(gly)$_4$ in H_2O and D_2O, focusing on the highly shifted H^4 resonances. The most highly shifted downfield resonance in each spectrum corresponds to the SAP isomer. The smaller resonance near 50-55 ppm reflects the TSAP isomer.

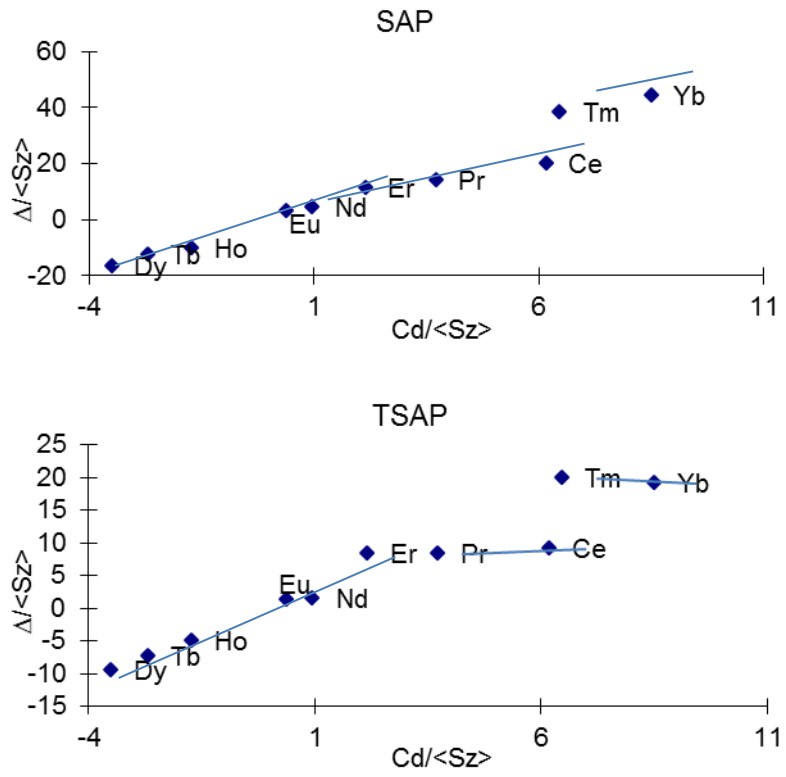

Figure S11. Plot of $\langle S_Z \rangle$ vs $C_d/\langle S_Z \rangle$ for the axial proton (H^4) of the SAP (left) isomeric forms in DOTMA-(gly)$_4$ chelates.

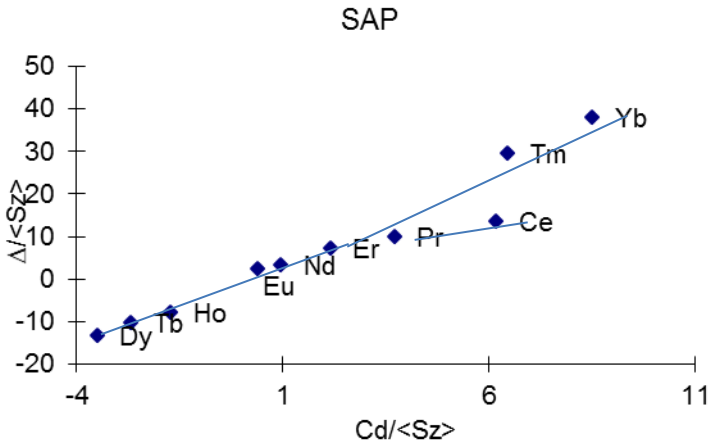

Figure S12. Plot of $\langle S_Z \rangle$ vs $C_d/\langle S_Z \rangle$ for the axial proton (H^4) of the DOTA-$(gly)_4$ chelates.

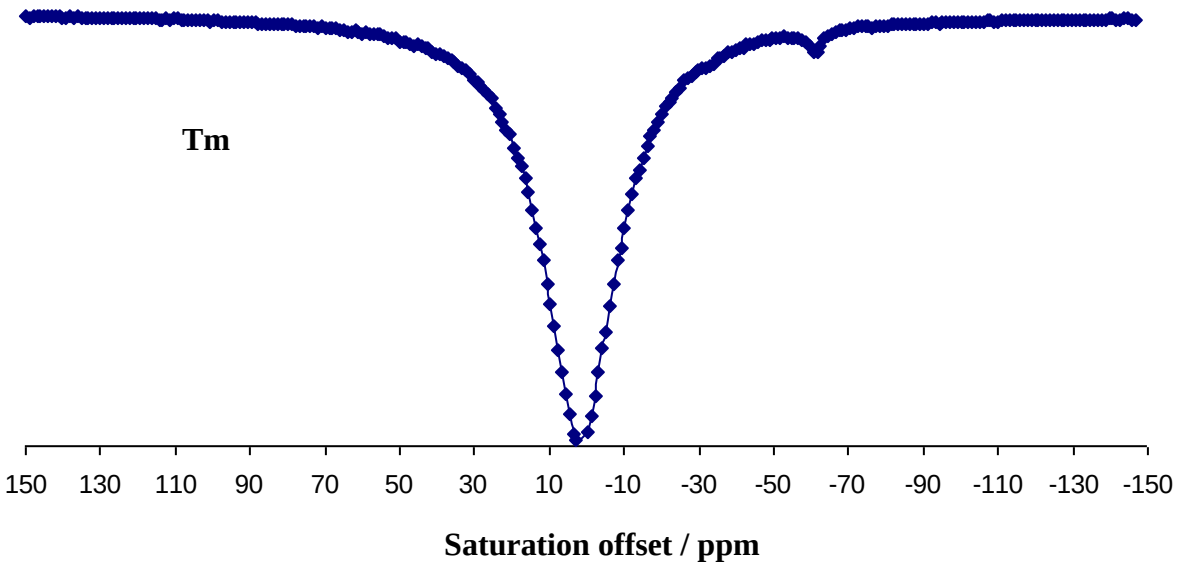

Figure S13. CEST spectra of 30 mM TmDOTMA-$(gly)_4$ complex at B_1= 800 Hz (90% D_2O+10% H_2O, 37ºC)

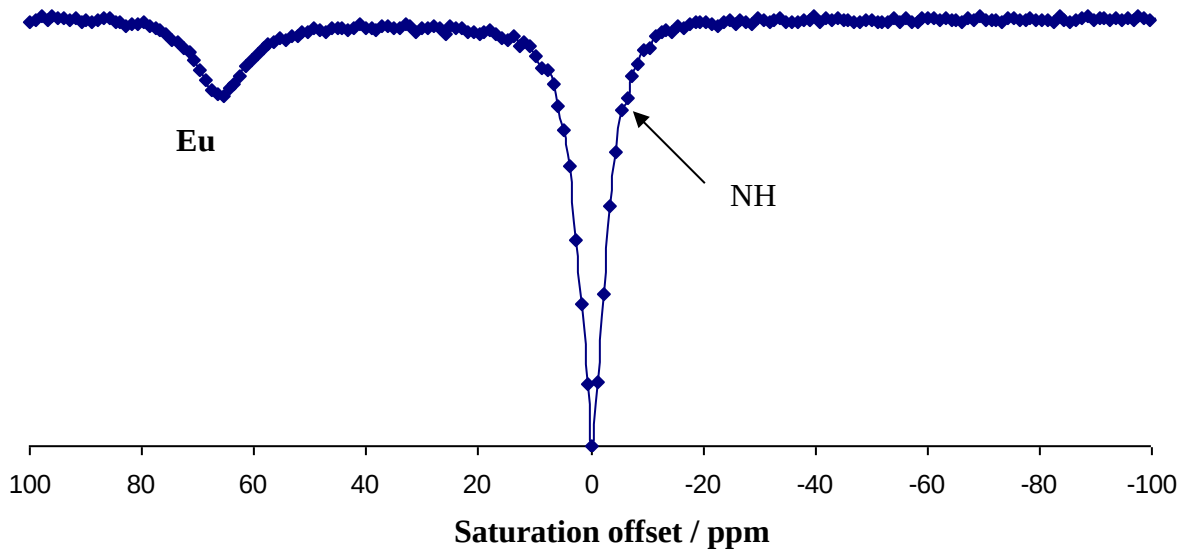

Figure S14. CEST spectra of 40 mM EuDOTMA-$(gly)_4$ complex at B_1= 400 Hz

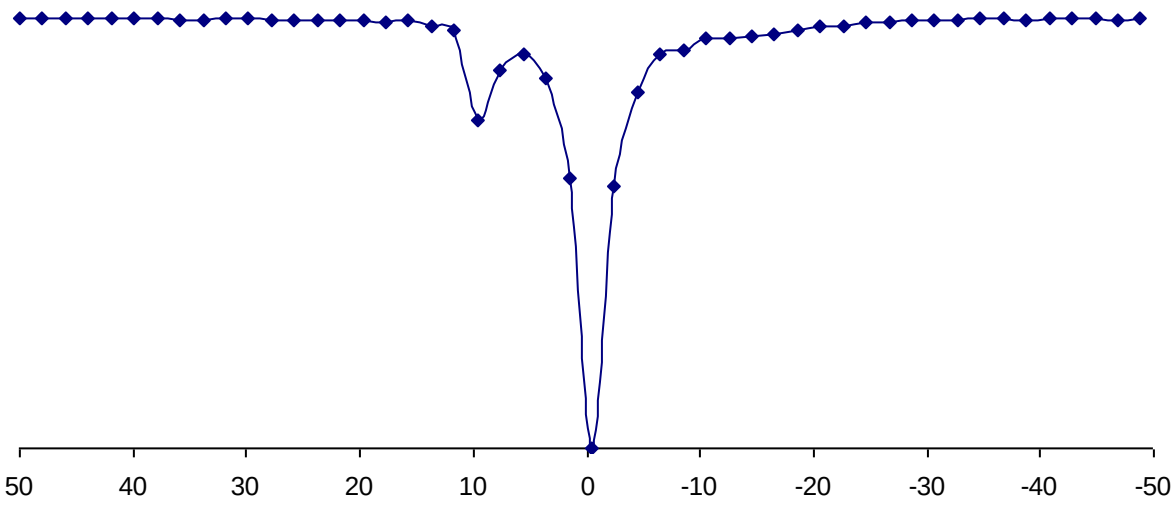

Figure S15. CEST spectra of 80mM SmDOTMA-$(gly)_4$complex at B_1= 100 Hz

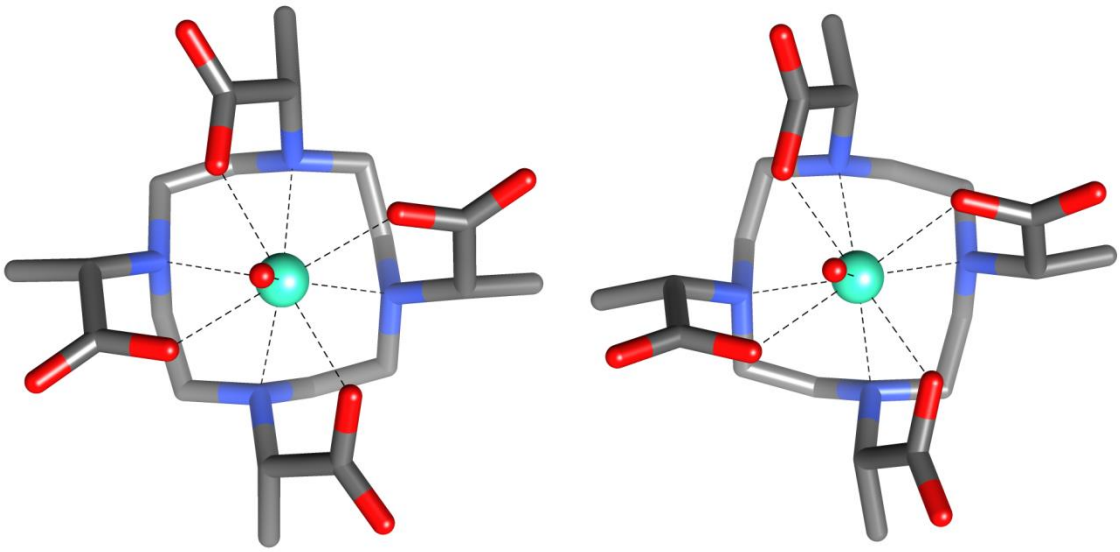

Figure S16. Optimized geometry of the SAP (left) and TSAP (right) isomers of $[Gd(DOTMA)(H_2O)]^-$ obtained with DFT calculations performed in aqueous solution at the TPSSh/LCRECP/6-31G(d,p) level. Hydrogen atoms were omitted for simplicity.

Optimized geometries. Geometry optimizations of the $[Gd(DOTMA)(H_2O)]^-$ complex provide two energy minima that correspond to the capped square antiprismatic (SAP) and capped twisted-square antiprismatic diastereoisomers (TSAP, Figure S14). The structural parameters calculated both in the gas-phase and in aqueous solution are shown in Table S3. The distances between the Gd^{3+} ion and the donor atoms of the ligand calculated for the two isomers are quite similar. As observed previously for related systems, the inclusion of solvent effects (water) provokes an important shortening of the Ln-N distances,[i] which is also reflected in a shortening of the distance between the Gd^{3+} ion and the plane defined by the four N atoms of the macrocycle (Ln-P_N) and a lengthening of the distance to the plane defined by the four O atoms of the ligand coordinated to the metal ion (Ln-P_O). The structural parameters calculated in aqueous solution for the TSAP isomer present an excellent agreement with the values observed in the X-ray crystal structure reported by Woods et al.[ii] The main discrepancy between

the experimental and calculated geometries concerns the distance between the metal ion and the oxygen atom of the inner-sphere water molecule, which is overestimated by calculations by ~0.17 Å. However, this is not surprising considering that the Gd-O_{water} distances observed in the solid state have been shown to be very sensitive the environment of the coordinated water molecule. For instance, the Gd-O_{water} distances observed for different salts of the $[Gd(DOTAM)(H_2O)]^{3+}$ complex were shown to differ up to 0.12 Å depending upon the anions present in the crystal lattice.[iii] Furthermore, it has been recently shown that the accurate calculation of accurate Gd-O_{water} distances and ^{17}O hyperfine coupling constants of coordinated water molecules requires the explicit inclusion of at least two second-sphere water molecules.[iv] The Gd-O_{water} distances calculated for the SAP isomer are shorter than those obtained for the TSAP one, in line with the faster water exchange rates usually observed for SAP isomers in DOTA-like complexes.[i]

Table S1. Geometrical parameters of calculated structures for the two isomers of $[Gd(DOTMA)(H_2O)]^-$ and $[Gd(DOTMA\text{-}gly_4)(H_2O)]^-$.[a]

	DOTMA					DOTMA-gly_4	
	SAP		TSAP		TSAP	SAP	TSAP
	Gas-phase	Water	Gas-phase	Water	exptl	Water	Water
Gd-N	2.736(25)	2.686(10)	2.756(25)	2.684(11)	2.681(23)	2.670(4)	2.689(7)
Gd-O	2.363(35)	2.374(23)	2.367(36)	2.382(26)	2.373(18)	2.399(11)	2.402(15)
Gd-O_W	2.565	2.616	2.556	2.671	2.500	2.520	2.546
ω^b	39.6(4)	38.4(4)	-28.0(4)	-26.8(3)	-26.2(8)	38.3(6)	-25.9(8)
Gd-$P_O{}^c$	0.648	0.769	0.700	0.847	0.794	0.776	0.871
Gd-$P_N{}^d$	1.702	1.611	1.755	1.630	1.663	1.572	1.618
P_O-$P_N{}^e$	2.350	2.381	2.456	2.477	2.457	2.348	2.489

[a] Distances (Å); The average values of bond distances to the oxygen atoms of the P_O (Gd-O) and P_N (Gd-N) planes are reported with standard deviations within parentheses; O_W, oxygen atom of the inner-sphere water molecule. [b] Mean twist angle (º) of the upper and lower planes. [c] Distance between the lanthanide and the least-squares plane defined by the coordinated oxygen atoms, P_O. [d] Distance between the lanthanide and the least-squares plane defined by the nitrogen atoms, P_N. [e] Distance between the centroids of the P_O and P_N planes.

The calculations performed for the Gd^{3+} complex were extended to several Ln^{3+} ions across the lanthanide series. The average Ln-O and Ln-N distances involving the eight donor atoms of the macrocycle decrease quadratically along the series as a consequence of the lanthanide contraction, in agreement with previous experimental [ii]and theoretical[iii, iv,v] studies.

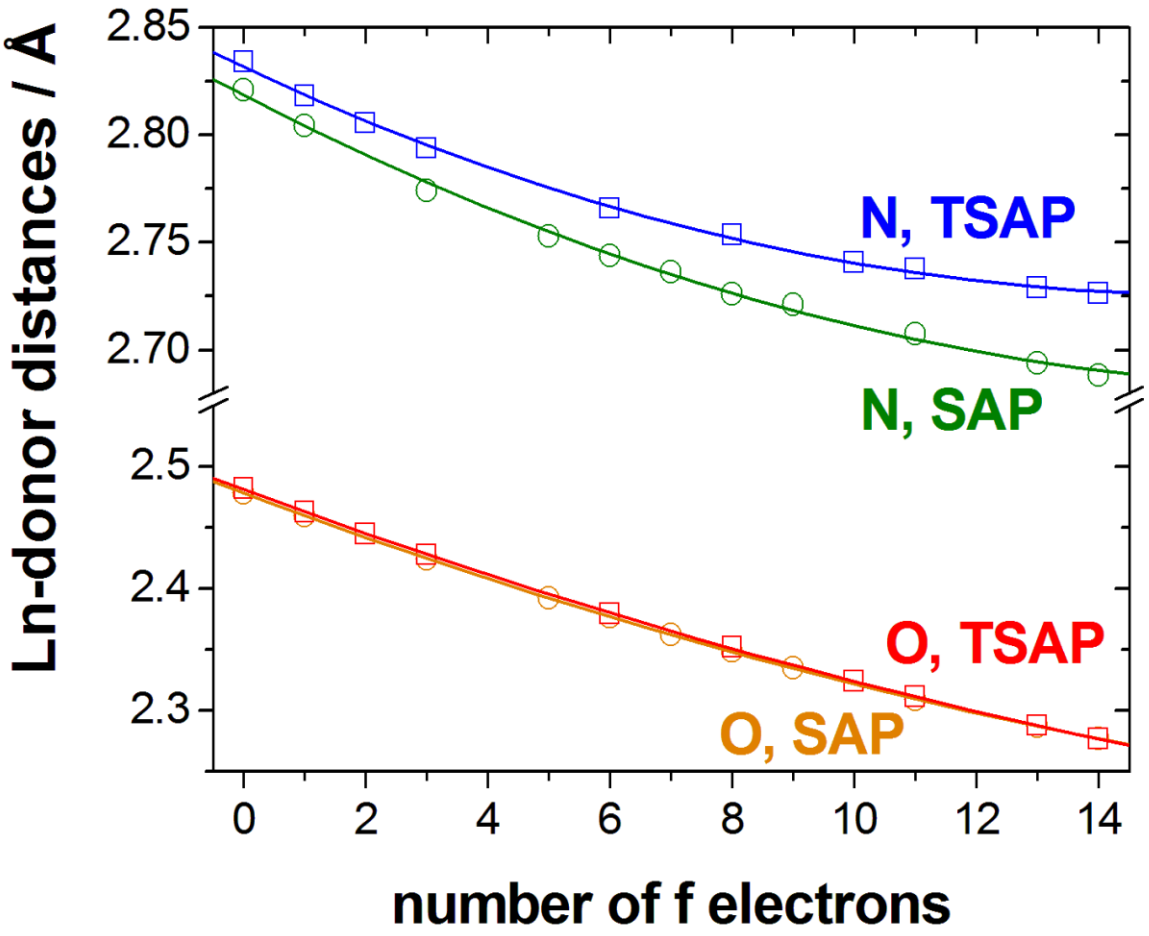

Figure S17. Variation of the average Ln-O_{amide} and Ln-N distances of the metal coordination environments for the $[Ln(DOTMA)(H_2O)]^-$ complexes at the TPSSh/LCRECP/6-31G(d,p) level (results obtained in the gas-phase). The solid lines represent quadratic fits of the data to $y = a + bx + cx^2$ with $R^2 > 0.99$.

The relative energies of the SAP and TSAP isomers of $[Ln(DOTMA)(H_2O)]^-$ were first investigated in the gas-phase (Figure S16, see also Table S4). According to our calculations the TSAP isomer is

more stable than the SAP one only for the light Ln^{3+} ions (Ln = La, Ce), which is in contrast with the experimental evidence. A stabilization of the SAP isomer occurs across the whole lanthanide series from La^{3+} to Lu^{3+}. To understand the reasons for this trend we calculated the counterpoise-corrected binding energies (BEs) of the ligands, as well as the relative strain energies (SEs) of the macrocyclic ligand across the series (Figure 17, see also Table S4). Our calculations provide positive relative binding energies (R_{BE}) calculated as $R_{BE} = BE_{TSAP} - BE_{SAP}$. The actual binding energies are negative, and therefore a positive binding energy indicates that the binding energy of the ligand is higher in the case of the SAP form, an effect that is magnified across the lanthanide series as the positive charge density of the metal ion increases. On the other hand, the relative strain energies $R_{SE} = LE_{TSAP} - LE_{SAP}$ (LE_{TSAP} and LE_{SAP} are the ligand energies with the ligand adopting the geometry observed for the TSAP and SAP isomer, respectively) are negative, which indicates that the in the SAP isomer the ligand is more strained than in the TSAP form. The R_{SE} values do not experience an important variation along the lanthanide series (< 0.5 kcal mol^{-1}), while the calculated R_{BEs} present a more important variation (~1.76 kcal mol^{-1}). The sum of the R_{SE} and R_{BE} values results in an increased stability of the SAP isomer across the series, as reflected by the relative free energies of the two isomers. Thus, the stabilization of the SAP form across the series is mainly the result of the higher binding energy of the ligand to the metal ion for this isomer, which further increases by increasing metal ion density.[vi, vii]

Table S2. Relative free energies (ΔG^o) for the SAP ⇆ TSAP equilibrium, relative binding energies (BE), relative strain energies of the ligands (SE), and hydration free energies (ΔG^o_{hyd}) (kcal·mol^{-1}).

Ln	$\Delta G^{oa,b}$	$\Delta G^{oa,c}$	$\Delta G^{oa,d}$	R_{SE}	R_{BE}	$\Delta G^o_{hyd(SAP)}$	$\Delta G^o_{hyd(TSAP)}$
La	-0.30	-3.66	-3.16	-4.24	4.44	-80.03	-83.39
Ce	-0.15	-3.54	-3.32	-4.19	4.60	-80.63	-84.03
Nd	0.14	-3.60	-4.03	-4.32	5.08	-79.80	-83.54
Eu	0.34	-3.11	-2.97	-3.84	5.11	-79.97	-83.42
Tb	0.73	-2.88	-2.61	-4.00	5.67	-79.48	-83.09
Er	1.48	-1.86	-2.55	-3.73	5.89	-79.23	-82.57
Yb	1.80	-1.60	-3.35	-3.83	6.05	-79.56	-82.96
Lu	1.80	-1.37	-3.71	-3.78	6.20	-79.49	-82.67

[a] Relative free energies are defined as $\Delta G^o = G^o_{TSAP} - G^o_{SAP}$; Note that the actual values of the complex free energies are negative, and therefore a positive relative free energy indicates that the SAP isomer is more stable than the TSAP one. The

same holds for the relative strain energies, as the actual ligand energies are negative. [b] Values obtained from calculations in the gas-phase. [c] Values calculated in aqueous solution using geometries optimized in the gas phase. [d] Values obtained in solution using geometries optimized in solution. [e] Obtained using the geometries optimized in the gas phase.

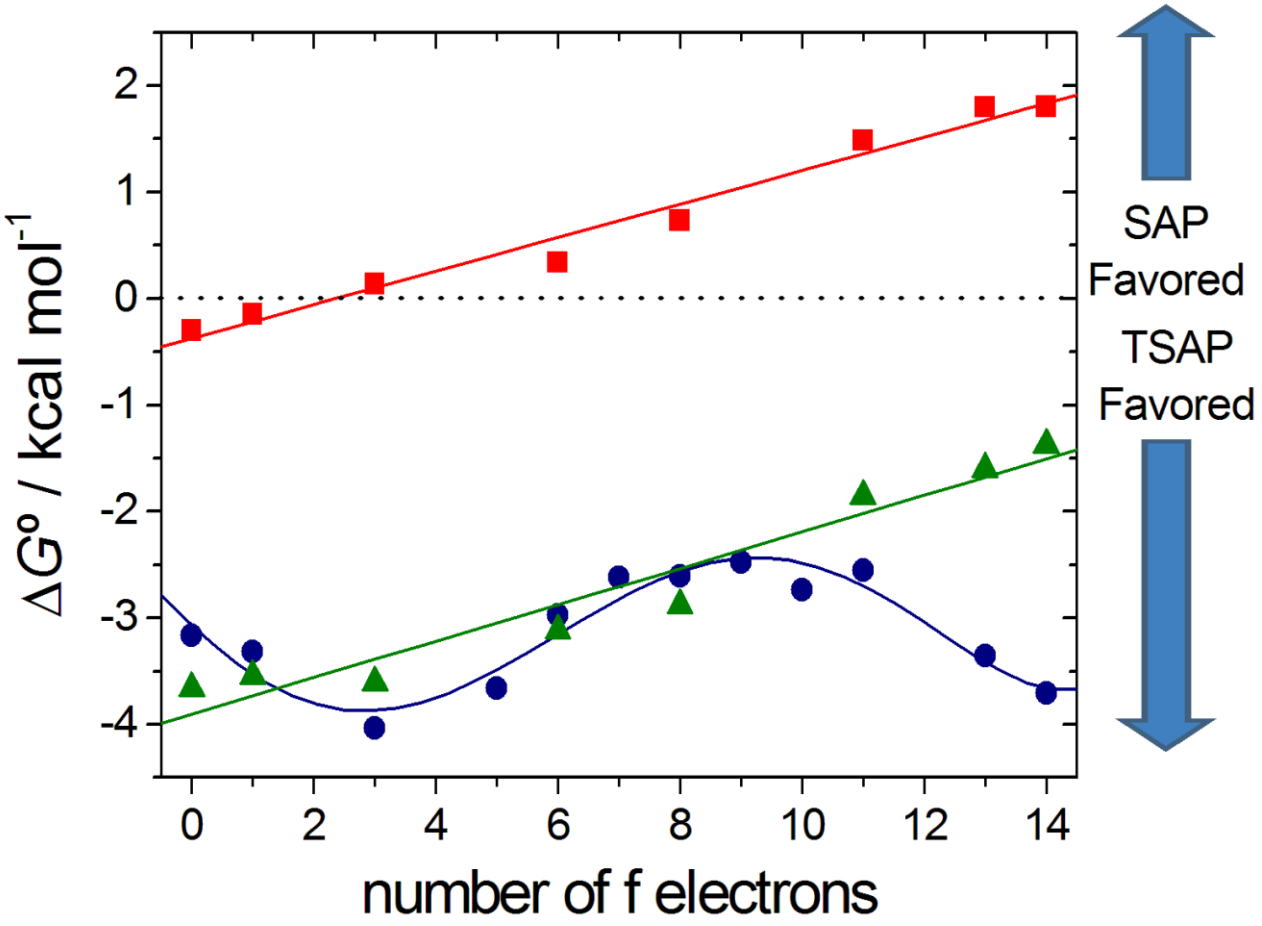

Figure S18. Relative energies of the SAP and TSAP isomers of $[Ln(DOTMA)(H_2O)]^-$ complexes calculated in the gas-phase (red squares), in aqueous solution using geometries optimized in the gas phase (green triangles) and in aqueous solution using geometries optimized in water (blue circles). The solid lines are used only as a guide for the eye.

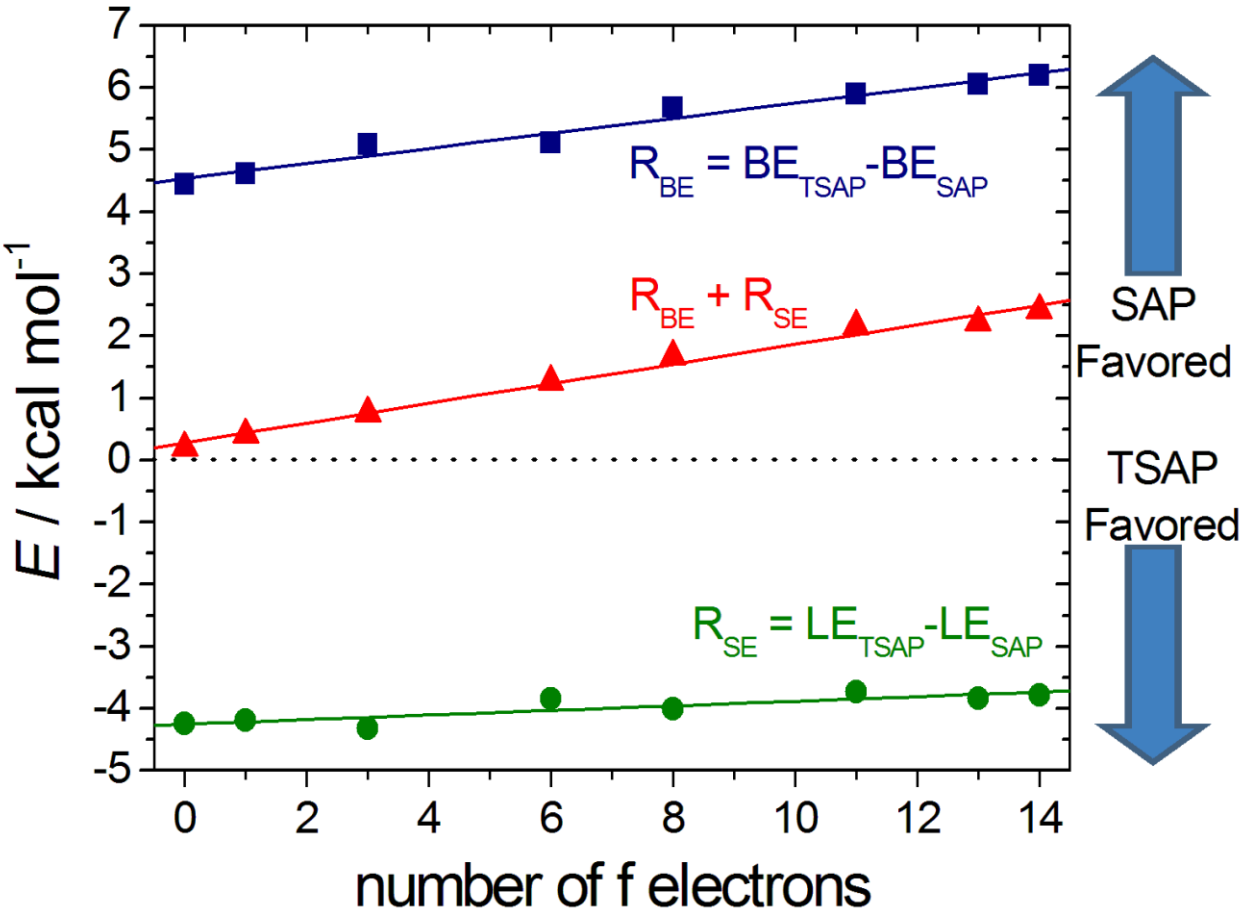

Figure S19. Relative energies binding energies (R_{BE}) and strain energies (R_{SE}) of the ligands in the SAP and TSAP isomers of $[Ln(DOTMA)(H_2O)]^-$ complexes calculated in the gas-phase. The solid lines are used only as a guide for the eye.

The effects of the solvent on the relative energies of the SAP and TSAP isomers of $[Ln(DOTMA)(H_2O)]^-$ were first evaluated by calculating the hydration free energies of the two isomers using the structures optimized in the gas-phase. The hydration free energies of the TSAP isomer are considerably more negative than those of the SAP form (3.2 – 3.7 kcal mol^{-1}, Table 2), and therefore the inclusion of solvent effects provokes an important stabilization of the TSAP form, which becomes the most stable one along the whole lanthanide series from La^{3+} to Lu^{3+}. This is related to the higher dipole moment of the TSAP complexes (26.3-26.8 *D* in aqueous solution at the TPSSh/LCRECP/6-31G(d,p)

level) compared with the SAP isomers (21.1-22.6 *D*). However, the hydration free energies of a given isomer do not change significantly across the series, and thus the SAP isomer becomes steadily more stable across the series. This is in contrast to the experimental trend, which shows a stabilization of the SAP isomer on proceeding to the right across the series with a maximum around Tb, with a stabilization of the TSAP isomer for the heaviest Ln^{3+} ions. Thus, we conclude that a structural change results in a significant change of the contributions related to the geometry relaxation passing from vacuo to solution. This is confirmed by the relative free energies of the two isomers obtained from geometry optimizations in aqueous solution, which show a bell-like shape with a maximum population of the SAP isomer around Tb (Figure 3).

Table S3. Yb^{3+}-induced paramagnetic shifts in Ybdota-$(gly)_4$ versus the geometric factors obtained from DFT calculations.[a]

	θ (°)	**r (Å)**	**$(3\cos^2\theta-1)/r^3$**	**δ_{para} (ppm)**
H1ax	22.93	3.744	0.0294	95.8
H1eq	45.57	4.459	0.00531	20.9
H2ax	67.14	3.721	-0.0106	-35.3
H2eq	47.11	4.467	0.00437	17.9
H3ax	87.49	3.588	-0.0215	-62.4
H3eq	72.88	4.382	-0.00880	-29.8
H4a	123.47	5.129	-0.000649	-3.1
H4b	108.68	5.207	-0.00490	-7.1

[a] The diamagnetic contribution was assumed to be 3.0 ppm for all nuclei. Negative paramagnetic shifts correspond to shifts to higher field.

Table S4. Yb^{3+}-induced paramagnetic shifts in Ybdotma-$(gly)_4$ versus the geometric factors obtained from DFT calculations.[a]

	θ (°)	r (Å)	$(3\cos^2\theta-1)/r^3$	δ_{para} (ppm)
H1ax	23.27	3.685	0.0306	119.5
H1eq	45.75	4.438	0.00527	19.9
H2ax	67.69	3.726	-0.0110	-41.9
H2eq	47.23	4.442	0.00432	15.2
H3	89.71	3.456	-0.0242	-85.9
H4a	123.66	5.089	-0.000595	-3.4
H4b	108.15	5.149	-0.00519	-8.6
Me	73.36	5.346	-0.00455	-18.3

[a] The diamagnetic contribution was assumed to be 3.0 ppm for all nuclei. Negative paramagnetic shifts correspond to shifts to higher field.

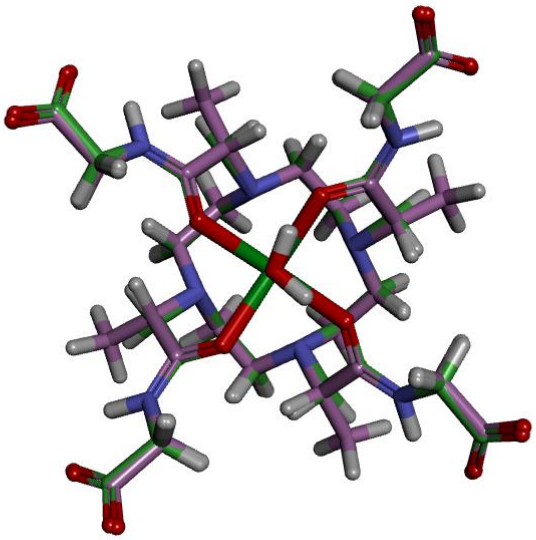

Figure S20. Overlay of the DFT structures obtained for the SAP isomers of YbDOTMA-$(gly)_4$ and YbDOTA-$(gly)_4$ complexes obtained with DFT calculations.

Table S5. [La(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-2.120791	0.740775	0.880792
2	6	-1.627660	1.632164	1.964225
3	6	-0.584166	2.661298	1.527786
4	7	0.662987	2.059009	0.973243
5	6	1.483959	1.513698	2.086956
6	6	2.548637	0.501996	1.664653
7	7	1.985587	-0.727388	1.035993
8	6	1.412882	-1.608097	2.089454
9	6	0.407326	-2.643277	1.583605
10	7	-0.796726	-2.047073	0.937116
11	6	-1.701962	-1.497433	1.981745
12	6	-2.729420	-0.482793	1.476852
13	6	-3.109174	1.406743	-0.035114
14	6	-2.405057	2.402984	-0.995534
15	8	-1.296792	1.977483	-1.513231
16	6	1.386499	3.083926	0.145325
17	6	2.413259	2.436396	-0.818781
18	8	2.028239	1.323854	-1.366927
19	6	3.047284	-1.399530	0.210651
20	6	2.436027	-2.415341	-0.787983
21	8	1.363175	-2.008639	-1.398776
22	6	-1.453695	-3.071778	0.054915
23	6	-2.417745	-2.404630	-0.961795
24	8	-1.972618	-1.318862	-1.508323
25	1	-1.205907	0.992647	2.743460
26	1	-2.458523	2.172021	2.439016
27	1	-1.003258	3.320058	0.764875
28	1	-0.358641	3.288493	2.403395
29	1	0.795825	1.042605	2.793120
30	1	1.978000	2.322366	2.642787
31	1	3.237947	0.955804	0.949361
32	1	3.136499	0.249888	2.559872
33	1	0.930824	-0.960972	2.826065
34	1	2.207186	-2.139353	2.631353
35	1	0.883085	-3.302336	0.854625
36	1	0.121716	-3.268863	2.442463
37	1	-1.071405	-1.028270	2.740561
38	1	-2.242114	-2.303428	2.497564
39	1	-3.367797	-0.940872	0.719325
40	1	-3.377594	-0.223361	2.327227
41	1	-3.444232	0.592951	-0.690886
42	1	0.608382	3.470595	-0.525495
43	1	3.427352	-0.592842	-0.429542
44	1	-0.629039	-3.449412	-0.563221
45	8	0.957783	-0.152082	-3.334395
46	1	1.629438	0.496976	-3.031057
47	1	1.324989	-1.016927	-3.050957
48	6	-2.104100	-4.251291	0.784524
49	1	-3.015267	-3.957550	1.311985
50	1	-2.388681	-5.004012	0.046116
51	1	-1.413830	-4.714093	1.496851
52	6	4.217384	-1.997825	0.998155
53	1	4.992318	-2.303850	0.292139
54	1	4.652020	-1.268280	1.688422

55	1	3.924071	-2.888112	1.560518
56	6	1.990156	4.257026	0.925097
57	1	2.859616	3.956469	1.515885
58	1	2.329852	5.011127	0.212320
59	1	1.253238	4.718970	1.589286
60	6	-4.330317	2.025135	0.652269
61	1	-4.075312	2.929673	1.210395
62	1	-5.054481	2.313163	-0.112941
63	1	-4.813135	1.313324	1.329366
64	8	3.005246	-3.496707	-0.987818
65	8	3.479774	3.018940	-1.055123
66	8	-2.944695	3.491688	-1.245300
67	8	-3.500482	-2.950864	-1.222691
68	57	-0.012777	0.001113	-0.757818

E(RTPSSh) = -1710.8181719 Hartree
Zero-point correction = 0.560676 Hartree/particle
Sum of electronic and thermal Energies = -1710.221705 Hartree
Sum of electronic and thermal Enthalpies = -1710.220761 Hartree
Sum of electronic and thermal Free Energies = -1710.322691 Hartree

Table S6. [Ce(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	2.062752	0.849897	0.895351
2	6	2.273546	-0.139765	1.983869
3	6	2.189932	-1.602921	1.546503
4	7	0.862772	-1.979824	0.979457
5	6	-0.119485	-2.151905	2.083254
6	6	-1.584558	-2.078405	1.651245
7	7	-1.962541	-0.773624	1.034601
8	6	-2.120319	0.254243	2.098726
9	6	-2.057369	1.699152	1.600922
10	7	-0.762388	2.047831	0.948142
11	6	0.276750	2.261285	1.990431
12	6	1.717701	2.175186	1.484691
13	6	3.245972	0.992000	-0.020618
14	6	3.368113	-0.219528	-0.984181
15	8	2.250867	-0.642037	-1.485527
16	6	1.011565	-3.205713	0.120705
17	6	-0.190916	-3.394050	-0.840170
18	8	-0.650675	-2.300889	-1.371498
19	6	-3.201767	-0.955117	0.200335
20	6	-3.397686	0.210668	-0.802587
21	8	-2.316074	0.608344	-1.404700
22	6	-0.952467	3.236935	0.046189
23	6	0.207878	3.365882	-0.977385
24	8	0.588834	2.254414	-1.523574
25	1	1.515101	0.053097	2.746972
26	1	3.242813	0.016409	2.477184
27	1	2.948210	-1.813484	0.789784
28	1	2.431929	-2.221813	2.423315
29	1	0.081223	-1.371926	2.822280
30	1	0.037170	-3.106094	2.604407
31	1	-1.801538	-2.863099	0.923939
32	1	-2.199898	-2.285542	2.539302
33	1	-1.327336	0.087729	2.832523
34	1	-3.065533	0.116820	2.640934

35	1	-2.854614	1.882842	0.877896
36	1	-2.250617	2.354031	2.463427
37	1	0.118820	1.505777	2.764506
38	1	0.143048	3.232206	2.486486
39	1	1.894581	2.935691	0.721413
40	1	2.378166	2.412175	2.332187
41	1	2.966736	1.826526	-0.676068
42	1	1.842379	-2.950475	-0.548961
43	1	-2.959058	-1.814149	-0.437613
44	1	-1.819919	2.960523	-0.566050
45	8	-0.715099	-0.442579	-3.310775
46	1	-0.801489	-1.384621	-3.048620
47	1	-1.576922	-0.050853	-3.051153
48	6	-1.249780	4.561216	0.755711
49	1	-0.377031	4.951824	1.285473
50	1	-1.528206	5.303407	0.004533
51	1	-2.079140	4.461362	1.463062
52	6	-4.483825	-1.265907	0.979691
53	1	-5.265622	-1.536398	0.266918
54	1	-4.342834	-2.104674	1.668397
55	1	-4.846501	-0.401852	1.542574
56	6	1.357988	-4.498176	0.867172
57	1	0.518578	-4.872527	1.459173
58	1	1.601986	-5.267927	0.132229
59	1	2.223032	-4.363289	1.523837
60	6	4.574590	1.323872	0.665482
61	1	4.967152	0.478784	1.237140
62	1	5.313202	1.563189	-0.102477
63	1	4.478780	2.187926	1.330610
64	8	-4.535478	0.649355	-1.016593
65	8	-0.598770	-4.535596	-1.091215
66	8	4.492259	-0.668773	-1.254584
67	8	0.657420	4.490007	-1.247340
68	58	0.007030	0.006151	-0.723895

E(RTPSSh) = -1711.4699447 Hartree
Zero-point correction = 0.560549 Hartree/particle
Sum of electronic and thermal Energies = -1710.873630 Hartree
Sum of electronic and thermal Enthalpies = -1710.872686 Hartree
Sum of electronic and thermal Free Energies = -1710.974039 Hartree

Table S7. [Pr(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	1.984962	0.996533	0.902548
2	6	2.273994	0.027021	1.991593
3	6	2.297336	-1.437485	1.553055
4	7	1.001592	-1.908122	0.983648
5	6	0.034717	-2.155125	2.085704
6	6	-1.429716	-2.192214	1.649122
7	7	-1.900101	-0.918369	1.031863
8	6	-2.143604	0.091022	2.096841
9	6	-2.185093	1.536643	1.601005
10	7	-0.916471	1.978471	0.953844
11	6	0.100571	2.267463	1.999290
12	6	1.543539	2.292013	1.493860
13	6	3.152329	1.224968	-0.016509
14	6	3.353138	0.030009	-0.986614

15	8	2.264842	-0.473445	-1.477422
16	6	1.240101	-3.116055	0.120278
17	6	0.061183	-3.379714	-0.851012
18	8	-0.475400	-2.316588	-1.371255
19	6	-3.114301	-1.190623	0.186595
20	6	-3.383804	-0.044251	-0.820948
21	8	-2.326229	0.439536	-1.401961
22	6	-1.188766	3.150533	0.051157
23	6	-0.043953	3.353859	-0.975860
24	8	0.421413	2.266784	-1.505446
25	1	1.508136	0.164569	2.759350
26	1	3.231884	0.254876	2.478806
27	1	3.069974	-1.591523	0.797166
28	1	2.582157	-2.038961	2.429061
29	1	0.174981	-1.362503	2.825238
30	1	0.261850	-3.094964	2.607064
31	1	-1.584591	-2.990654	0.920742
32	1	-2.031673	-2.445118	2.534452
33	1	-1.347124	-0.017903	2.837520
34	1	-3.080476	-0.117584	2.630935
35	1	-2.990471	1.661556	0.874440
36	1	-2.430244	2.174750	2.463025
37	1	-0.001171	1.500594	2.771530
38	1	-0.106808	3.224395	2.497204
39	1	1.662439	3.065133	0.732042
40	1	2.185030	2.576351	2.341338
41	1	2.814739	2.041916	-0.666852
42	1	2.056547	-2.801245	-0.542091
43	1	-2.803571	-2.030292	-0.448010
44	1	-2.037269	2.816268	-0.558935
45	8	-0.609973	-0.445569	-3.284512
46	1	-0.636131	-1.396781	-3.043241
47	1	-1.507614	-0.124297	-3.051910
48	6	-1.571916	4.453368	0.759642
49	1	-0.723460	4.906760	1.278787
50	1	-1.910574	5.171050	0.009472
51	1	-2.384573	4.297508	1.476266
52	6	-4.377510	-1.592660	0.954564
53	1	-5.127410	-1.932192	0.237093
54	1	-4.178854	-2.409864	1.654897
55	1	-4.813109	-0.753348	1.502998
56	6	1.668406	-4.385527	0.863600
57	1	0.850123	-4.822794	1.441965
58	1	1.977400	-5.131180	0.128322
59	1	2.512383	-4.192748	1.533152
60	6	4.457555	1.643130	0.667392
61	1	4.910905	0.822654	1.229879
62	1	5.174027	1.940337	-0.101242
63	1	4.304250	2.492618	1.340553
64	8	-4.548914	0.300886	-1.058271
65	8	-0.259253	-4.544926	-1.119596
66	8	4.504408	-0.332805	-1.271465
67	8	0.316057	4.505056	-1.264672
68	59	0.007925	0.007684	-0.703798

E(RTPSSh) = -1712.105256 Hartree
Zero-point correction = 0.560304 Hartree/particle
Sum of electronic and thermal Energies = -1711.509237 Hartree
Sum of electronic and thermal Enthalpies = -1711.508293 Hartree
Sum of electronic and thermal Free Energies = -1711.609495 Hartree

Table S8. [Nd(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	1.927246	1.090421	0.906949
2	6	2.268969	0.139211	1.996101
3	6	2.364393	-1.321178	1.556331
4	7	1.093261	-1.851540	0.984405
5	6	0.141948	-2.151266	2.086467
6	6	-1.318242	-2.260370	1.649101
7	7	-1.849075	-1.010657	1.031564
8	6	-2.145060	-0.016603	2.097923
9	6	-2.257345	1.425126	1.602727
10	7	-1.010756	1.926598	0.956892
11	6	-0.012333	2.269575	2.003516
12	6	1.427077	2.363952	1.498287
13	6	3.079647	1.372522	-0.016802
14	6	3.327286	0.188714	-0.988456
15	8	2.259016	-0.364352	-1.469460
16	6	1.390468	-3.042680	0.116017
17	6	0.223978	-3.356967	-0.853422
18	8	-0.366272	-2.317070	-1.362605
19	6	-3.045902	-1.340624	0.182993
20	6	-3.365541	-0.203745	-0.818515
21	8	-2.328154	0.332652	-1.389859
22	6	-1.336561	3.081512	0.049777
23	6	-0.201112	3.329969	-0.976070
24	8	0.315385	2.260949	-1.494578
25	1	1.499106	0.239337	2.765712
26	1	3.215578	0.415485	2.480175
27	1	3.144713	-1.436390	0.801544
28	1	2.676919	-1.910050	2.431480
29	1	0.243293	-1.354183	2.827630
30	1	0.416230	-3.079923	2.605212
31	1	-1.433423	-3.065504	0.920752
32	1	-1.907965	-2.542608	2.533872
33	1	-1.344706	-0.087301	2.839080
34	1	-3.070952	-0.272125	2.630839
35	1	-3.067314	1.510740	0.875601
36	1	-2.534310	2.050549	2.464525
37	1	-0.077394	1.500453	2.777501
38	1	-0.267759	3.216331	2.498521
39	1	1.508381	3.141843	0.736491
40	1	2.055104	2.678832	2.345126
41	1	2.703820	2.174428	-0.664891
42	1	2.190088	-2.685652	-0.545696
43	1	-2.692525	-2.161433	-0.454057
44	1	-2.168600	2.707270	-0.559748
45	8	-0.581005	-0.460649	-3.263210
46	1	-0.565042	-1.411972	-3.020984
47	1	-1.491599	-0.178762	-3.029606
48	6	-1.778789	4.368861	0.751927
49	1	-0.951623	4.864372	1.266920
50	1	-2.152229	5.065625	-0.001392
51	1	-2.581864	4.178909	1.471123
52	6	-4.290347	-1.806829	0.945195
53	1	-5.018490	-2.186046	0.225205
54	1	-4.052366	-2.611617	1.647602
55	1	-4.771384	-0.990519	1.490272
56	6	1.879925	-4.293578	0.852878

57	1	1.082619	-4.775107	1.425291
58	1	2.228747	-5.018313	0.114553
59	1	2.710798	-4.062576	1.526665
60	6	4.368662	1.847039	0.660907
61	1	4.861215	1.046568	1.219250
62	1	5.066832	2.176754	-0.111253
63	1	4.181630	2.687908	1.336344
64	8	-4.544239	0.081482	-1.067811
65	8	-0.040108	-4.534028	-1.131153
66	8	4.491138	-0.123420	-1.282580
67	8	0.107764	4.493691	-1.274088
68	60	0.006747	0.009684	-0.692115

E(RTPSSh) = -1712.72839 Hartree
Zero-point correction = 0.561232 Hartree/particle
Sum of electronic and thermal Energies = -1712.131757 Hartree
Sum of electronic and thermal Enthalpies = -1712.130813 Hartree
Sum of electronic and thermal Free Energies = -1712.230381 Hartree

Table S9. [Sm(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.813819	2.038407	0.924143
2	6	1.672061	1.508939	2.016329
3	6	2.664692	0.435144	1.572398
4	7	2.006556	-0.768439	0.988873
5	6	1.462209	-1.614667	2.083155
6	6	0.387993	-2.604402	1.635856
7	7	-0.803948	-1.947172	1.028515
8	6	-1.647237	-1.359547	2.102395
9	6	-2.646086	-0.313390	1.609208
10	7	-1.991758	0.859257	0.963018
11	6	-1.442694	1.758597	2.011428
12	6	-0.370758	2.725964	1.511323
13	6	1.542305	2.975968	0.002424
14	6	2.462259	2.202688	-0.976915
15	8	1.962941	1.106132	-1.454027
16	6	2.981146	-1.499147	0.108488
17	6	2.262632	-2.449554	-0.879541
18	8	1.145491	-1.995662	-1.364644
19	6	-1.542966	-2.940503	0.176226
20	6	-2.479023	-2.239309	-0.837969
21	8	-1.993936	-1.161018	-1.377744
22	6	-2.964568	1.545956	0.045832
23	6	-2.227745	2.448800	-0.976121
24	8	-1.141305	1.944757	-1.471288
25	1	1.008017	1.097691	2.780515
26	1	2.231759	2.316970	2.507020
27	1	3.347681	0.841244	0.823148
28	1	3.275166	0.164010	2.446390
29	1	1.048602	-0.944042	2.840355
30	1	2.262390	-2.175715	2.584507
31	1	0.798503	-3.296822	0.897805
32	1	0.101705	-3.203895	2.512658
33	1	-0.974508	-0.904623	2.834583
34	1	-2.198412	-2.140589	2.643527
35	1	-3.330067	-0.758323	0.883357
36	1	-3.254574	-0.000027	2.470265

37	1	-1.026065	1.123954	2.797118
38	1	-2.240463	2.342958	2.489741
39	1	-0.787958	3.385380	0.747580
40	1	-0.076099	3.363316	2.358198
41	1	0.748868	3.376483	-0.641026
42	1	3.384685	-0.711661	-0.540617
43	1	-0.757543	-3.376379	-0.453926
44	1	-3.367987	0.728789	-0.565001
45	8	-0.137089	-0.600217	-3.224853
46	1	0.463471	-1.346119	-3.007928
47	1	-1.028545	-0.960085	-3.030592
48	6	-4.124559	2.270482	0.735223
49	1	-3.799830	3.183249	1.241411
50	1	-4.851941	2.565558	-0.024028
51	1	-4.629246	1.622982	1.459433
52	6	-2.250378	-4.068498	0.934478
53	1	-2.585826	-4.816488	0.213066
54	1	-1.577216	-4.556432	1.646295
55	1	-3.136502	-3.716207	1.469216
56	6	4.142513	-2.187018	0.833591
57	1	3.820635	-3.077396	1.380350
58	1	4.873776	-2.512045	0.090698
59	1	4.640287	-1.504413	1.529442
60	6	2.266252	4.141208	0.683804
61	1	3.161891	3.814397	1.218634
62	1	2.589539	4.848082	-0.083169
63	1	1.609037	4.669997	1.381592
64	8	-3.573048	-2.746597	-1.117320
65	8	2.792448	-3.522769	-1.194374
66	8	3.564491	2.681250	-1.283807
67	8	-2.721558	3.540859	-1.294580
68	62	0.000134	0.014221	-0.666469

E(RTPSSh) = -1713.9504937 Hartree
Zero-point correction = 0.561384 Hartree/particle
Sum of electronic and thermal Energies = -1713.353788 Hartree
Sum of electronic and thermal Enthalpies = -1713.352843 Hartree
Sum of electronic and thermal Free Energies = -1713.452667 Hartree

Table S10. [Eu(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.788212	2.043332	0.927382
2	6	1.656354	1.528114	2.018732
3	6	2.659115	0.464346	1.574191
4	7	2.011290	-0.744771	0.990679
5	6	1.477786	-1.596627	2.085860
6	6	0.417159	-2.600216	1.637262
7	7	-0.780585	-1.956145	1.027179
8	6	-1.634509	-1.384628	2.101102
9	6	-2.643383	-0.348391	1.608543
10	7	-1.998505	0.831092	0.965688
11	6	-1.460073	1.732663	2.017678
12	6	-0.402967	2.716154	1.517939
13	6	1.502114	2.987339	0.001362
14	6	2.424669	2.221163	-0.980364
15	8	1.938121	1.113251	-1.443940
16	6	2.990625	-1.465114	0.107609

17	6	2.277993	-2.415466	-0.883760
18	8	1.151721	-1.972040	-1.357324
19	6	-1.503338	-2.954429	0.167295
20	6	-2.441605	-2.258918	-0.848007
21	8	-1.969549	-1.167326	-1.372503
22	6	-2.975507	1.511176	0.048534
23	6	-2.243477	2.414415	-0.975707
24	8	-1.149127	1.918696	-1.461269
25	1	0.999079	1.112645	2.786536
26	1	2.208289	2.344366	2.504495
27	1	3.337389	0.877195	0.824459
28	1	3.273098	0.198419	2.447321
29	1	1.054358	-0.929670	2.841033
30	1	2.284865	-2.145964	2.589159
31	1	0.837600	-3.287935	0.900494
32	1	0.135565	-3.203013	2.513291
33	1	-0.968941	-0.925169	2.836677
34	1	-2.177585	-2.174731	2.637268
35	1	-3.321174	-0.798946	0.880474
36	1	-3.256774	-0.042071	2.468655
37	1	-1.031543	1.099404	2.798271
38	1	-2.264799	2.302884	2.501369
39	1	-0.830265	3.371661	0.756515
40	1	-0.114143	3.355707	2.365106
41	1	0.701867	3.377842	-0.639555
42	1	3.387980	-0.672590	-0.539044
43	1	-0.709792	-3.377449	-0.461558
44	1	-3.374997	0.691106	-0.560697
45	8	-0.123777	-0.565176	-3.206952
46	1	0.483805	-1.307681	-2.998864
47	1	-1.011518	-0.937127	-3.018284
48	6	-4.138457	2.231305	0.737329
49	1	-3.817892	3.146602	1.241644
50	1	-4.867747	2.521295	-0.022020
51	1	-4.639293	1.582647	1.463196
52	6	-2.201277	-4.093674	0.917152
53	1	-2.525421	-4.841952	0.190901
54	1	-1.525667	-4.577212	1.629655
55	1	-3.093358	-3.753577	1.449885
56	6	4.156951	-2.147695	0.829559
57	1	3.841210	-3.041513	1.374311
58	1	4.889445	-2.466039	0.085005
59	1	4.650976	-1.463853	1.526885
60	6	2.216752	4.161262	0.677342
61	1	3.118135	3.844986	1.208847
62	1	2.529080	4.870021	-0.092425
63	1	1.556972	4.684262	1.377088
64	8	-3.523544	-2.782202	-1.144272
65	8	2.818686	-3.479557	-1.210913
66	8	3.516482	2.714132	-1.301447
67	8	-2.747460	3.498662	-1.304999
68	63	-0.000058	0.014392	-0.650342

E(RTPSSh) = -1714.5481586 Hartree
Zero-point correction = 0.561636 Hartree/particle
Sum of electronic and thermal Energies = -1713.951299 Hartree
Sum of electronic and thermal Enthalpies = -1713.950354 Hartree
Sum of electronic and thermal Free Energies = -1714.049723 Hartree

Table S11. [Gd(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.818456	2.026390	0.931962
2	6	1.681101	1.498938	2.021370
3	6	2.667188	0.421088	1.574199
4	7	1.998000	-0.775430	0.989585
5	6	1.452546	-1.620490	2.084331
6	6	0.377239	-2.608025	1.635787
7	7	-0.809704	-1.944559	1.025782
8	6	-1.658320	-1.364438	2.099998
9	6	-2.648992	-0.309997	1.609380
10	7	-1.983672	0.859189	0.968818
11	6	-1.434559	1.753441	2.021310
12	6	-0.360597	2.719210	1.523560
13	6	1.545049	2.956343	0.002173
14	6	2.449238	2.172609	-0.981879
15	8	1.946568	1.067220	-1.433766
16	6	2.961469	-1.510935	0.102693
17	6	2.227582	-2.442803	-0.889822
18	8	1.104782	-1.979261	-1.352480
19	6	-1.545467	-2.927822	0.160957
20	6	-2.466253	-2.213067	-0.855945
21	8	-1.979318	-1.119906	-1.363473
22	6	-2.945486	1.553899	0.047490
23	6	-2.193653	2.439721	-0.976641
24	8	-1.103719	1.925453	-1.452752
25	1	1.020332	1.090569	2.789729
26	1	2.244701	2.307261	2.507080
27	1	3.351362	0.823890	0.824319
28	1	3.277629	0.143666	2.446246
29	1	1.037524	-0.948839	2.839917
30	1	2.252009	-2.181099	2.587442
31	1	0.786756	-3.302400	0.899057
32	1	0.086370	-3.206466	2.511751
33	1	-0.988822	-0.918543	2.840557
34	1	-2.215560	-2.148440	2.630485
35	1	-3.334448	-0.747078	0.880194
36	1	-3.257361	0.005084	2.469913
37	1	-1.018730	1.115608	2.804767
38	1	-2.231800	2.337176	2.501387
39	1	-0.776170	3.383299	0.762887
40	1	-0.060724	3.352594	2.371519
41	1	0.749450	3.359574	-0.636642
42	1	3.372386	-0.724139	-0.542414
43	1	-0.757091	-3.362417	-0.466453
44	1	-3.357545	0.739667	-0.561164
45	8	-0.131439	-0.526152	-3.188131
46	1	0.461176	-1.283367	-2.991053
47	1	-1.026207	-0.883251	-3.003716
48	6	-4.098012	2.296574	0.729940
49	1	-3.763673	3.207472	1.233316
50	1	-4.819249	2.597038	-0.032981
51	1	-4.612971	1.658815	1.455659
52	6	-2.264959	-4.058177	0.904129
53	1	-2.597952	-4.798552	0.173895
54	1	-1.599750	-4.554919	1.617426
55	1	-3.153542	-3.706238	1.435095
56	6	4.116445	-2.218137	0.819283
57	1	3.785562	-3.107363	1.362612
58	1	4.840580	-2.547940	0.071636

59	1	4.625444	-1.545847	1.517152
60	6	2.282691	4.119268	0.672521
61	1	3.179302	3.788867	1.203589
62	1	2.606185	4.819052	-0.100768
63	1	1.634160	4.657025	1.371655
64	8	-3.549921	-2.723038	-1.169168
65	8	2.749894	-3.512461	-1.229186
66	8	3.544215	2.650775	-1.315357
67	8	-2.679141	3.529933	-1.314841
68	64	0.000285	0.015272	-0.632233

E(RTPSSh) = -1715.1389338 Hartree
Zero-point correction = 0.561731 Hartree/particle
Sum of electronic and thermal Energies = -1714.542048 Hartree
Sum of electronic and thermal Enthalpies = -1714.541104 Hartree
Sum of electronic and thermal Free Energies = -1714.640142 Hartree

Table S12. [Tb(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.786927	2.030056	0.936306
2	6	1.657366	1.518721	2.027339
3	6	2.657781	0.455073	1.579733
4	7	2.002851	-0.748502	0.994054
5	6	1.475587	-1.603993	2.088852
6	6	0.414616	-2.604657	1.637274
7	7	-0.778670	-1.953122	1.027592
8	6	-1.635359	-1.390976	2.103766
9	6	-2.641447	-0.352375	1.612971
10	7	-1.989793	0.824413	0.973053
11	6	-1.457162	1.725305	2.028098
12	6	-0.399987	2.707850	1.529019
13	6	1.500433	2.969672	0.005981
14	6	2.412024	2.197318	-0.979366
15	8	1.924635	1.081575	-1.422733
16	6	2.973629	-1.467997	0.101722
17	6	2.249504	-2.402600	-0.894704
18	8	1.118570	-1.950468	-1.349521
19	6	-1.499967	-2.942487	0.157072
20	6	-2.425815	-2.236497	-0.860663
21	8	-1.952520	-1.132670	-1.358253
22	6	-2.960329	1.507246	0.051552
23	6	-2.219874	2.400283	-0.973840
24	8	-1.118949	1.902006	-1.441620
25	1	1.002192	1.103794	2.797253
26	1	2.209396	2.336577	2.510147
27	1	3.336026	0.868007	0.830040
28	1	3.272498	0.184238	2.450790
29	1	1.053819	-0.940296	2.847867
30	1	2.285135	-2.154125	2.587263
31	1	0.834593	-3.291305	0.899199
32	1	0.129451	-3.209710	2.510522
33	1	-0.971949	-0.935540	2.843879
34	1	-2.179071	-2.185079	2.633303
35	1	-3.319626	-0.799656	0.883219
36	1	-3.255377	-0.044766	2.472170
37	1	-1.030143	1.092374	2.809859
38	1	-2.264235	2.294306	2.509226

39	1	-0.826784	3.364964	0.768609
40	1	-0.107388	3.346285	2.375627
41	1	0.698754	3.362838	-0.631150
42	1	3.373544	-0.672939	-0.539829
43	1	-0.704279	-3.364720	-0.469435
44	1	-3.362786	0.687631	-0.555887
45	8	-0.123015	-0.494951	-3.172075
46	1	0.477049	-1.247459	-2.980545
47	1	-1.013537	-0.863429	-2.989602
48	65	0.000516	0.016652	-0.627476
49	6	-4.120940	2.237435	0.733635
50	1	-3.796950	3.153323	1.234740
51	1	-4.846091	2.528050	-0.029384
52	1	-4.627993	1.595318	1.461069
53	6	-2.208646	-4.083488	0.894048
54	1	-2.530081	-4.825582	0.160376
55	1	-1.540880	-4.574500	1.608897
56	1	-3.103544	-3.743320	1.422066
57	6	4.137942	-2.165594	0.812397
58	1	3.818732	-3.063049	1.349115
59	1	4.865764	-2.480747	0.061973
60	1	4.638927	-1.492255	1.515056
61	6	2.224960	4.141090	0.675680
62	1	3.127816	3.821708	1.202897
63	1	2.536526	4.846367	-0.097524
64	1	1.572043	4.669213	1.378071
65	8	-3.499139	-2.761029	-1.184449
66	8	2.784609	-3.462248	-1.244565
67	8	3.496414	2.691934	-1.322687
68	8	-2.723129	3.479003	-1.322141

E(RTPSSh) = -1715.7276619 Hartree
Zero-point correction = 0.561876 Hartree/particle
Sum of electronic and thermal Energies = -1715.130694 Hartree
Sum of electronic and thermal Enthalpies = -1715.129750 Hartree
Sum of electronic and thermal Free Energies = -1715.228536 Hartree

Table S13. [Dy(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.993739	1.930999	0.936948
2	6	1.811140	1.335179	2.026046
3	6	2.692359	0.174085	1.574010
4	7	1.907287	-0.951344	0.992778
5	6	1.291749	-1.739312	2.092047
6	6	0.138682	-2.630499	1.640159
7	7	-0.978439	-1.859129	1.025867
8	6	-1.781988	-1.219209	2.100305
9	6	-2.666973	-0.077124	1.609370
10	7	-1.886761	1.021782	0.975074
11	6	-1.262133	1.856755	2.034085
12	6	-0.113732	2.729009	1.533899
13	6	1.797747	2.790470	0.002928
14	6	2.615737	1.926768	-0.987715
15	8	2.020678	0.855540	-1.407489
16	6	2.791198	-1.776201	0.101246
17	6	1.967740	-2.620198	-0.898566
18	8	0.888617	-2.048721	-1.344905

19	6	-1.791500	-2.765374	0.146154
20	6	-2.631292	-1.964088	-0.874730
21	8	-2.051983	-0.901570	-1.350439
22	6	-2.775027	1.811093	0.055351
23	6	-1.941627	2.612424	-0.973088
24	8	-0.891039	2.003334	-1.423874
25	1	1.120137	0.989921	2.799325
26	1	2.445987	2.093022	2.504577
27	1	3.403646	0.513593	0.818383
28	1	3.280406	-0.161360	2.440816
29	1	0.933021	-1.028269	2.840658
30	1	2.039663	-2.362413	2.600773
31	1	0.489802	-3.356118	0.903562
32	1	-0.209445	-3.202270	2.512802
33	1	-1.081960	-0.843474	2.850945
34	1	-2.411866	-1.956076	2.616702
35	1	-3.385012	-0.446259	0.874176
36	1	-3.247996	0.294115	2.466282
37	1	-0.896132	1.179073	2.809640
38	1	-2.006512	2.500273	2.522124
39	1	-0.473609	3.426100	0.774318
40	1	0.246412	3.333167	2.379421
41	1	1.038062	3.265947	-0.629191
42	1	3.280236	-1.030552	-0.537434
43	1	-1.038761	-3.264452	-0.476461
44	1	-3.269876	1.042046	-0.549777
45	8	-0.170863	-0.446943	-3.155723
46	1	0.337979	-1.265145	-2.970394
47	1	-1.097529	-0.710264	-2.970119
48	66	0.003389	0.016353	-0.606321
49	6	-3.844556	2.668485	0.739292
50	1	-3.419688	3.543849	1.238068
51	1	-4.535195	3.036069	-0.022444
52	1	-4.417594	2.088171	1.469695
53	6	-2.619280	-3.831138	0.871820
54	1	-3.006401	-4.534440	0.131792
55	1	-2.010977	-4.389760	1.590133
56	1	-3.480467	-3.405058	1.393707
57	6	3.867771	-2.602672	0.812351
58	1	3.448725	-3.456250	1.352321
59	1	4.553012	-3.000923	0.061445
60	1	4.444371	-1.989994	1.512556
61	6	2.644440	3.880482	0.667558
62	1	3.507643	3.470446	1.198966
63	1	3.030191	4.543084	-0.109849
64	1	2.051109	4.479746	1.365744
65	8	-3.741956	-2.385401	-1.222815
66	8	2.387861	-3.727116	-1.259516
67	8	3.733542	2.317415	-1.357483
68	8	-2.334675	3.730387	-1.339738

E(RTPSSh) = -1716.3144319 Hartree
Zero-point correction = 0.561910 Hartree/particle
Sum of electronic and thermal Energies = -1715.717432 Hartree
Sum of electronic and thermal Enthalpies = -1715.716487 Hartree
Sum of electronic and thermal Free Energies = -1715.815096 Hartree

Table S14. [Ho(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center	Atomic	Coordinates (Angstroms)

Number	Number	X	Y	Z
1	7	0.760296	2.030523	0.942174
2	6	1.639990	1.534634	2.032958
3	6	2.651186	0.483403	1.583010
4	7	2.004313	-0.723868	0.996950
5	6	1.489491	-1.584248	2.093478
6	6	0.444574	-2.599720	1.639834
7	7	-0.753035	-1.959583	1.027442
8	6	-1.620476	-1.416470	2.104668
9	6	-2.636504	-0.388394	1.614977
10	7	-1.992490	0.793723	0.978183
11	6	-1.472779	1.697399	2.037356
12	6	-0.431664	2.695729	1.537988
13	6	1.460097	2.975374	0.007397
14	6	2.369224	2.207954	-0.982490
15	8	1.895002	1.078368	-1.403965
16	6	2.978015	-1.433860	0.101070
17	6	2.256415	-2.362502	-0.901026
18	8	1.114973	-1.918446	-1.337447
19	6	-1.458852	-2.951936	0.148679
20	6	-2.381676	-2.248450	-0.871799
21	8	-1.921693	-1.128134	-1.344744
22	6	-2.966267	1.469936	0.055841
23	6	-2.229153	2.359308	-0.973411
24	8	-1.116260	1.871437	-1.422897
25	1	0.992155	1.112358	2.805242
26	1	2.181849	2.361137	2.512361
27	1	3.323652	0.904075	0.832584
28	1	3.270343	0.216298	2.452036
29	1	1.055136	-0.924268	2.848790
30	1	2.306466	-2.120086	2.595370
31	1	0.875328	-3.280760	0.902841
32	1	0.163455	-3.208825	2.511513
33	1	-0.965090	-0.956458	2.848762
34	1	-2.154896	-2.220800	2.628161
35	1	-3.309017	-0.840984	0.883379
36	1	-3.254872	-0.086911	2.473143
37	1	-1.033036	1.067361	2.814596
38	1	-2.287660	2.251059	2.523095
39	1	-0.868684	3.347974	0.779308
40	1	-0.143972	3.337119	2.383946
41	1	0.651648	3.361578	-0.624982
42	1	3.374898	-0.633549	-0.535449
43	1	-0.655418	-3.364134	-0.474317
44	1	-3.366704	0.647208	-0.548284
45	8	-0.113686	-0.454144	-3.146686
46	1	0.487584	-1.206489	-2.958964
47	1	-1.003024	-0.826096	-2.965677
48	67	0.000817	0.017723	-0.606462
49	6	-4.128783	2.199204	0.735648
50	1	-3.807796	3.118040	1.233388
51	1	-4.854958	2.484443	-0.028365
52	1	-4.633473	1.558248	1.465800
53	6	-2.162942	-4.102211	0.875378
54	1	-2.473097	-4.843428	0.136004
55	1	-1.495778	-4.590876	1.592442
56	1	-3.064099	-3.772165	1.399157
57	6	4.144801	-2.132085	0.806860
58	1	3.828672	-3.032812	1.339963
59	1	4.872258	-2.441722	0.053812
60	1	4.645003	-1.460819	1.512099

61	6	2.179960	4.153273	0.670618
62	1	3.088156	3.842052	1.193602
63	1	2.481834	4.858872	-0.106104
64	1	1.527130	4.678204	1.375527
65	8	-3.439564	-2.787611	-1.221249
66	8	2.801109	-3.409760	-1.272750
67	8	3.438852	2.717404	-1.349787
68	8	-2.745016	3.425150	-1.342631

E(RTPSSh) = -1716.8982644 Hartree
Zero-point correction = 0.562182 Hartree/particle
Sum of electronic and thermal Energies = -1716.301106 Hartree
Sum of electronic and thermal Enthalpies = -1716.300161 Hartree
Sum of electronic and thermal Free Energies = -1716.398395 Hartree

Table S15. [Er(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.744789	2.031318	0.944726
2	6	1.628339	1.543236	2.036048
3	6	2.646772	0.500297	1.584827
4	7	2.005618	-0.709413	0.998168
5	6	1.497730	-1.572846	2.095561
6	6	0.462180	-2.596672	1.641005
7	7	-0.738145	-1.963152	1.027822
8	6	-1.610333	-1.428904	2.105788
9	6	-2.633156	-0.408358	1.616254
10	7	-1.994722	0.776846	0.980275
11	6	-1.482693	1.683459	2.040937
12	6	-0.450262	2.689870	1.541116
13	6	1.438089	2.979494	0.008668
14	6	2.346969	2.215131	-0.983000
15	8	1.878959	1.079772	-1.395425
16	6	2.982007	-1.413357	0.100835
17	6	2.263053	-2.340024	-0.904250
18	8	1.116127	-1.901551	-1.331756
19	6	-1.436252	-2.957843	0.146042
20	6	-2.357711	-2.256796	-0.876615
21	8	-1.904857	-1.128985	-1.338425
22	6	-2.971265	1.447716	0.057202
23	6	-2.236915	2.336711	-0.973515
24	8	-1.117606	1.855460	-1.413787
25	1	0.983961	1.115112	2.808072
26	1	2.162889	2.374370	2.515530
27	1	3.315762	0.926092	0.834269
28	1	3.268405	0.235739	2.452842
29	1	1.056056	-0.915411	2.848797
30	1	2.319022	-2.100161	2.599401
31	1	0.898722	-3.274164	0.904191
32	1	0.184043	-3.208263	2.511878
33	1	-0.958750	-0.964223	2.850167
34	1	-2.137980	-2.238315	2.628292
35	1	-3.302231	-0.865006	0.884085
36	1	-3.253976	-0.110472	2.473874
37	1	-1.036494	1.056276	2.816861
38	1	-2.302220	2.229031	2.527893
39	1	-0.892388	3.338797	0.782640
40	1	-0.165858	3.333410	2.386504

41	1	0.626537	3.362092	-0.621735
42	1	3.376528	-0.609858	-0.532949
43	1	-0.628910	-3.365654	-0.474602
44	1	-3.368732	0.622500	-0.545284
45	8	-0.107835	-0.435634	-3.135498
46	1	0.495960	-1.186018	-2.948469
47	1	-0.995596	-0.811626	-2.955547
48	6	-4.136415	2.174134	0.735484
49	1	-3.818839	3.094758	1.232126
50	1	-4.863093	2.455774	-0.029376
51	1	-4.639299	1.532529	1.466333
52	6	-2.137838	-4.111988	0.868892
53	1	-2.442851	-4.853283	0.127464
54	1	-1.470771	-4.599018	1.587181
55	1	-3.041773	-3.786353	1.390629
56	6	4.150708	-2.110337	0.804566
57	1	3.836962	-3.012894	1.335985
58	1	4.878466	-2.416621	0.050440
59	1	4.649645	-1.439389	1.511020
60	6	2.154628	4.160499	0.669838
61	1	3.065083	3.853321	1.191300
62	1	2.452141	4.866698	-0.108011
63	1	1.501096	4.683329	1.375673
64	8	-3.407610	-2.803545	-1.238047
65	8	2.813687	-3.380394	-1.286305
66	8	3.409604	2.731562	-1.360640
67	8	-2.760071	3.395396	-1.352859
68	68	0.000834	0.018096	-0.597155

E(RTPSSh) = -1717.4814698 Hartree
Zero-point correction = 0.562281 Hartree/particle
Sum of electronic and thermal Energies = -1716.884223 Hartree
Sum of electronic and thermal Enthalpies = -1716.883278 Hartree
Sum of electronic and thermal Free Energies = -1716.981486 Hartree

Table S16. [Tm(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.560163	2.081800	0.944585
2	6	1.482452	1.678422	2.038103
3	6	2.590083	0.734248	1.583883
4	7	2.054163	-0.525793	0.997452
5	6	1.627843	-1.429993	2.096642
6	6	0.690368	-2.542788	1.642169
7	7	-0.558918	-2.016031	1.025954
8	6	-1.479260	-1.566900	2.103512
9	6	-2.587223	-0.641438	1.614058
10	7	-2.052460	0.594403	0.979733
11	6	-1.624358	1.542676	2.041654
12	6	-0.687119	2.636219	1.540053
13	6	1.170842	3.085920	0.008370
14	6	2.133250	2.401099	-0.990438
15	8	1.770664	1.222912	-1.386398
16	6	3.087916	-1.145024	0.100998
17	6	2.451920	-2.124850	-0.908985
18	8	1.265745	-1.794703	-1.324513
19	6	-1.166394	-3.067573	0.142490
20	6	-2.136868	-2.450832	-0.888210

21	8	-1.793516	-1.279467	-1.334048
22	6	-3.083190	1.181829	0.058287
23	6	-2.425398	2.123653	-0.976078
24	8	-1.262819	1.744465	-1.401010
25	1	0.880846	1.191936	2.809758
26	1	1.937367	2.555438	2.517660
27	1	3.213544	1.219417	0.830524
28	1	3.236596	0.524170	2.448562
29	1	1.129090	-0.816280	2.850841
30	1	2.493619	-1.880294	2.600728
31	1	1.186883	-3.178201	0.906321
32	1	0.464852	-3.176325	2.512603
33	1	-0.875912	-1.050040	2.853985
34	1	-1.933892	-2.422588	2.620869
35	1	-3.211747	-1.153913	0.879785
36	1	-3.233702	-0.398983	2.470218
37	1	-1.123869	0.958922	2.818278
38	1	-2.489970	2.012000	2.528050
39	1	-1.184671	3.240767	0.779474
40	1	-0.458788	3.304706	2.382972
41	1	0.327950	3.405248	-0.615606
42	1	3.417788	-0.309837	-0.527792
43	1	-0.323418	-3.408260	-0.470227
44	1	-3.416066	0.325067	-0.538743
45	8	-0.066518	-0.415015	-3.120031
46	1	0.593179	-1.117530	-2.937495
47	1	-0.922626	-0.860492	-2.946794
48	69	-0.001898	0.017853	-0.580528
49	6	-4.300442	1.815824	0.738197
50	1	-4.057123	2.764568	1.224430
51	1	-5.052375	2.029523	-0.024353
52	1	-4.743954	1.141496	1.478012
53	6	-1.775254	-4.274393	0.863652
54	1	-2.006674	-5.041819	0.122363
55	1	-1.078247	-4.700223	1.592378
56	1	-2.710414	-4.025115	1.372598
57	6	4.307674	-1.747303	0.805914
58	1	4.067413	-2.674521	1.333607
59	1	5.058311	-1.991919	0.051757
60	1	4.750367	-1.040798	1.515201
61	6	1.794224	4.319013	0.669625
62	1	2.728401	4.084881	1.187713
63	1	2.032664	5.047033	-0.108545
64	1	1.103455	4.787141	1.378440
65	8	-3.120719	-3.097850	-1.270101
66	8	3.097441	-3.104296	-1.304197
67	8	3.136962	3.013006	-1.386570
68	8	-3.041609	3.125382	-1.370457

E(RTPSSh) = -1718.0612266 Hartree
Zero-point correction = 0.562381 Hartree/particle
Sum of electronic and thermal Energies = -1717.463864 Hartree
Sum of electronic and thermal Enthalpies = -1717.462920 Hartree
Sum of electronic and thermal Free Energies = -1717.561077 Hartree

Table S17. [Yb(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z

1	7	0.519596	2.088931	0.949207
2	6	1.450587	1.704343	2.042247
3	6	2.575231	0.779717	1.588399
4	7	2.061915	-0.488921	1.001127
5	6	1.654901	-1.402141	2.100198
6	6	0.738033	-2.531525	1.644158
7	7	-0.520128	-2.025509	1.028914
8	6	-1.449428	-1.597473	2.107541
9	6	-2.574019	-0.691357	1.618513
10	7	-2.060971	0.553163	0.983244
11	6	-1.654362	1.510227	2.045809
12	6	-0.736614	2.621029	1.545363
13	6	1.111490	3.101826	0.010997
14	6	2.083050	2.430503	-0.987047
15	8	1.742474	1.242526	-1.372400
16	6	3.103956	-1.087818	0.101235
17	6	2.482252	-2.068895	-0.915683
18	8	1.285833	-1.760899	-1.319472
19	6	-1.107005	-3.083700	0.139387
20	6	-2.086571	-2.478131	-0.888918
21	8	-1.762986	-1.297702	-1.326506
22	6	-3.099322	1.120586	0.058293
23	6	-2.455414	2.069389	-0.978162
24	8	-1.280626	1.715012	-1.391285
25	1	0.858934	1.208269	2.815352
26	1	1.890364	2.590113	2.519899
27	1	3.191505	1.276200	0.836560
28	1	3.224313	0.580291	2.453761
29	1	1.144945	-0.799069	2.855342
30	1	2.530093	-1.836054	2.602248
31	1	1.246225	-3.156423	0.907208
32	1	0.523372	-3.171143	2.512833
33	1	-0.856759	-1.070561	2.859483
34	1	-1.887996	-2.463329	2.621683
35	1	-3.190229	-1.215280	0.885419
36	1	-3.223916	-0.459124	2.474891
37	1	-1.144369	0.936902	2.823745
38	1	-2.529888	1.963223	2.529850
39	1	-1.245045	3.217901	0.786039
40	1	-0.519692	3.292410	2.389051
41	1	0.263024	3.405469	-0.613069
42	1	3.422482	-0.244803	-0.522942
43	1	-0.257554	-3.405203	-0.474858
44	1	-3.416409	0.257434	-0.537934
45	8	-0.059055	-0.399397	-3.112482
46	1	0.613507	-1.089548	-2.929972
47	1	-0.906768	-0.860763	-2.938896
48	6	-4.328833	1.735888	0.733148
49	1	-4.102053	2.687384	1.221646
50	1	-5.079903	1.939340	-0.032954
51	1	-4.765973	1.054460	1.470234
52	6	-1.694805	-4.305414	0.852573
53	1	-1.915204	-5.070859	0.105818
54	1	-0.989648	-4.724843	1.577052
55	1	-2.632856	-4.075029	1.364855
56	6	4.332195	-1.677207	0.802145
57	1	4.104883	-2.610746	1.324164
58	1	5.086577	-1.905857	0.046745
59	1	4.763739	-0.968841	1.516410
60	6	1.715139	4.346172	0.668960
61	1	2.654248	4.128588	1.185018
62	1	1.939022	5.077062	-0.110741

63	1	1.018469	4.803638	1.378957
64	8	-3.058462	-3.139033	-1.277497
65	8	3.146569	-3.027921	-1.329091
66	8	3.071069	3.060324	-1.394590
67	8	-3.091097	3.053883	-1.385212
68	70	-0.001812	0.018067	-0.572957

E(RTPSSh) = -1718.6447604 Hartree
Zero-point correction = 0.562353 Hartree/particle
Sum of electronic and thermal Energies = -1718.047409 Hartree
Sum of electronic and thermal Enthalpies = -1718.046465 Hartree
Sum of electronic and thermal Free Energies = -1718.144744 Hartree

Table S18. [Lu(DOTMA)(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.581246	2.066652	0.953409
2	6	1.498922	1.657242	2.047717
3	6	2.595586	0.701879	1.592238
4	7	2.040383	-0.548232	1.003543
5	6	1.610220	-1.449768	2.103190
6	6	0.661136	-2.550550	1.645454
7	7	-0.579552	-2.003351	1.030157
8	6	-1.495338	-1.551348	2.110230
9	6	-2.592188	-0.613462	1.620154
10	7	-2.037979	0.613757	0.986019
11	6	-1.606012	1.557308	2.050048
12	6	-0.657013	2.639997	1.548678
13	6	1.204641	3.059273	0.013243
14	6	2.150748	2.355873	-0.987335
15	8	1.771263	1.178814	-1.370833
16	6	3.063037	-1.178571	0.102037
17	6	2.409605	-2.139339	-0.913919
18	8	1.221778	-1.795122	-1.314664
19	6	-1.199946	-3.043331	0.141722
20	6	-2.155752	-2.406636	-0.889393
21	8	-1.795876	-1.234987	-1.322553
22	6	-3.057196	1.213860	0.059702
23	6	-2.381801	2.139280	-0.977540
24	8	-1.218676	1.746592	-1.389243
25	1	0.892774	1.176759	2.819830
26	1	1.961981	2.530567	2.526200
27	1	3.225701	1.180844	0.840386
28	1	3.239380	0.480158	2.455888
29	1	1.117474	-0.832435	2.858503
30	1	2.473276	-1.908206	2.604286
31	1	1.150454	-3.189454	0.907698
32	1	0.425774	-3.184547	2.512727
33	1	-0.887059	-1.040939	2.861240
34	1	-1.957334	-2.404753	2.624433
35	1	-3.222784	-1.118426	0.885854
36	1	-3.236209	-0.360943	2.475097
37	1	-1.112112	0.968998	2.827602
38	1	-2.468832	2.034027	2.533971
39	1	-1.147334	3.250869	0.788440
40	1	-0.418780	3.305802	2.390900
41	1	0.365407	3.390465	-0.609259
42	1	3.406273	-0.345444	-0.522162

43	1	-0.360923	-3.395394	-0.469848
44	1	-3.402314	0.360743	-0.535338
45	8	-0.069545	-0.391308	-3.109447
46	1	0.579747	-1.103168	-2.925751
47	1	-0.931719	-0.824762	-2.936522
48	71	-0.001621	0.017545	-0.570682
49	6	-4.265710	1.870756	0.733294
50	1	-4.008004	2.816107	1.218460
51	1	-5.010445	2.095701	-0.033002
52	1	-4.724102	1.206109	1.472803
53	6	-1.830728	-4.242474	0.856588
54	1	-2.072413	-5.003146	0.111551
55	1	-1.142836	-4.682225	1.585665
56	1	-2.763135	-3.979906	1.363744
57	6	4.273113	-1.806385	0.801154
58	1	4.017795	-2.733046	1.322571
59	1	5.019139	-2.057435	0.044564
60	1	4.727453	-1.112412	1.515421
61	6	1.851086	4.282821	0.669456
62	1	2.783207	4.034028	1.184206
63	1	2.098288	5.005224	-0.111138
64	1	1.171289	4.763953	1.380137
65	8	-3.145357	-3.037089	-1.283476
66	8	3.042815	-3.118602	-1.328602
67	8	3.158216	2.952301	-1.397415
68	8	-2.984758	3.143183	-1.386636

E(RTPSSh) = -1719.1985484 Hartree
Zero-point correction = 0.562662 Hartree/particle
Sum of electronic and thermal Energies = -1718.601020 Hartree
Sum of electronic and thermal Enthalpies = -1718.600076 Hartree
Sum of electronic and thermal Free Energies = -1718.697968 Hartree

Table S19. [La(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.914280	-0.886111	1.144210
2	6	-1.272339	-1.674306	2.228033
3	6	-0.163612	-2.607231	1.740963
4	7	0.982366	-1.893400	1.113559
5	6	1.826187	-1.253980	2.156474
6	6	2.732021	-0.142390	1.627298
7	7	1.987124	0.994450	1.019820
8	6	1.396892	1.855364	2.074949
9	6	0.262909	2.754388	1.580525
10	7	-0.904754	2.004004	1.040965
11	6	-1.706917	1.423268	2.147488
12	6	-2.633365	0.288267	1.711331
13	6	-2.843004	-1.721479	0.317403
14	6	-3.245923	-0.963228	-0.977100
15	8	-2.394092	-0.078404	-1.405030
16	6	1.771129	-2.824257	0.244204
17	6	0.961370	-3.212490	-1.024063
18	8	0.004057	-2.399794	-1.361479
19	6	2.870681	1.759415	0.082470
20	6	3.208381	0.883956	-1.159921
21	8	2.348048	-0.039701	-1.454716
22	6	-1.725309	2.881216	0.145140

23	6	-0.935965	3.202181	-1.157345
24	8	-0.039885	2.333393	-1.506414
25	1	-0.867673	-0.967073	2.956456
26	1	-2.011940	-2.276781	2.773447
27	1	-0.562755	-3.302152	0.997623
28	1	0.169713	-3.208285	2.599293
29	1	1.157909	-0.851409	2.921799
30	1	2.455892	-1.995342	2.667852
31	1	3.400674	-0.543225	0.861178
32	1	3.364345	0.200929	2.458883
33	1	1.027543	1.198214	2.866518
34	1	2.159812	2.492449	2.544011
35	1	0.628334	3.407442	0.783653
36	1	-0.039562	3.402905	2.415440
37	1	-1.008149	1.058260	2.904463
38	1	-2.315796	2.191269	2.644665
39	1	-3.327532	0.648425	0.947227
40	1	-3.237371	-0.003885	2.582429
41	1	-2.240844	-2.567129	-0.039899
42	1	2.607599	-2.228058	-0.141717
43	1	2.250481	2.575416	-0.310346
44	1	-2.568091	2.260601	-0.185915
45	8	-0.528723	-0.599708	-3.274954
46	1	-0.360031	-1.525791	-2.988754
47	1	-1.465625	-0.446480	-3.020160
48	57	0.010043	0.012492	-0.642959
49	6	4.137574	2.360129	0.704540
50	1	4.798845	1.586074	1.105022
51	1	4.683506	2.888121	-0.078937
52	1	3.906344	3.070828	1.503514
53	6	-2.281489	4.157002	0.790372
54	1	-1.481729	4.825807	1.121866
55	1	-2.866783	4.689515	0.039067
56	1	-2.931109	3.938296	1.643070
57	6	-4.075727	-2.270124	1.047968
58	1	-4.727705	-1.466645	1.403079
59	1	-4.649075	-2.873307	0.342288
60	1	-3.803899	-2.901845	1.898453
61	6	2.335874	-4.069594	0.941913
62	1	1.540991	-4.713726	1.329547
63	1	2.899004	-4.644659	0.205183
64	1	3.008528	-3.810956	1.764845
65	8	-4.300667	-1.270585	-1.546152
66	8	-1.234792	4.221934	-1.795038
67	8	4.234175	1.142518	-1.805133
68	8	1.294915	-4.223289	-1.655111

E(RTPSSh) = -1710.8237971 Hartree
Zero-point correction = 0.560730 Hartree/particle
Sum of electronic and thermal Energies = -1710.227515 Hartree
Sum of electronic and thermal Enthalpies = -1710.226571 Hartree
Sum of electronic and thermal Free Energies = -1710.327730 Hartree

Table S20. [Ce(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.878037	-0.955826	1.141257
2	6	-1.216939	-1.728383	2.224614

3	6	-0.074024	-2.618430	1.737579
4	7	1.043804	-1.858464	1.113599
5	6	1.862890	-1.194878	2.160211
6	6	2.729755	-0.051541	1.634720
7	7	1.943976	1.057380	1.028347
8	6	1.327249	1.897321	2.085287
9	6	0.162272	2.754935	1.591866
10	7	-0.974704	1.960979	1.049751
11	6	-1.760305	1.354066	2.153517
12	6	-2.643215	0.187071	1.712149
13	6	-2.771469	-1.818003	0.302714
14	6	-3.189079	-1.066395	-0.990140
15	8	-2.366098	-0.146697	-1.402006
16	6	1.865623	-2.753361	0.237553
17	6	1.067324	-3.159771	-1.031522
18	8	0.088671	-2.371127	-1.365606
19	6	2.798656	1.852544	0.089439
20	6	3.147761	0.998594	-1.164046
21	8	2.314040	0.050289	-1.456588
22	6	-1.821878	2.806998	0.148262
23	6	-1.038647	3.148243	-1.151790
24	8	-0.116927	2.304344	-1.495425
25	1	-0.838182	-1.010873	2.957004
26	1	-1.938910	-2.357454	2.763362
27	1	-0.445970	-3.327171	0.993177
28	1	0.281292	-3.207960	2.595030
29	1	1.177869	-0.815410	2.922573
30	1	2.515663	-1.915537	2.672211
31	1	3.414602	-0.426322	0.869679
32	1	3.347530	0.313411	2.467947
33	1	0.981648	1.227563	2.876918
34	1	2.068377	2.560665	2.552974
35	1	0.504385	3.422545	0.796674
36	1	-0.165255	3.390465	2.427147
37	1	-1.052537	1.013280	2.913517
38	1	-2.398321	2.100196	2.647396
39	1	-3.349472	0.523798	0.948434
40	1	-3.237042	-0.131940	2.580746
41	1	-2.137999	-2.639553	-0.055673
42	1	2.678137	-2.124033	-0.146690
43	1	2.153778	2.654167	-0.292379
44	1	-2.641279	2.157263	-0.184876
45	8	-0.462670	-0.560607	-3.254586
46	1	-0.266500	-1.486134	-2.984752
47	1	-1.408254	-0.446316	-3.012363
48	6	4.054283	2.482629	0.705294
49	1	4.741974	1.723665	1.089975
50	1	4.576417	3.035019	-0.077561
51	1	3.811053	3.176623	1.515204
52	6	-2.422533	4.065342	0.787277
53	1	-1.646619	4.759531	1.123183
54	1	-3.019214	4.577988	0.031109
55	1	-3.070268	3.827163	1.636094
56	6	-3.989614	-2.412194	1.021533
57	1	-4.662285	-1.632664	1.391137
58	1	-4.546275	-3.016215	0.303276
59	1	-3.701459	-3.051949	1.860560
60	6	2.476340	-3.981480	0.926223
61	1	1.705846	-4.652291	1.317750
62	1	3.051932	-4.535446	0.182971
63	1	3.145961	-3.703265	1.745175
64	8	-4.223683	-1.410078	-1.575110

65	8	-1.363437	4.157662	-1.792901
66	8	4.157467	1.293921	-1.819002
67	8	1.428804	-4.157012	-1.668286
68	58	0.006691	0.012471	-0.624241

E(RTPSSh) = -1711.4756591 Hartree
Zero-point correction = 0.561012 Hartree/particle
Sum of electronic and thermal Energies = -1710.879121 Hartree
Sum of electronic and thermal Enthalpies = -1710.878177 Hartree
Sum of electronic and thermal Free Energies = -1710.979323 Hartree

Table S21. [Pr(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.886907	-0.925272	1.140815
2	6	-1.244894	-1.712864	2.224052
3	6	-0.115570	-2.618456	1.736798
4	7	1.009644	-1.870060	1.113187
5	6	1.842124	-1.227090	2.161654
6	6	2.725269	-0.096644	1.637309
7	7	1.951140	1.020827	1.032449
8	6	1.353952	1.869736	2.093138
9	6	0.203400	2.745952	1.601868
10	7	-0.939969	1.965389	1.055216
11	6	-1.737596	1.375086	2.159033
12	6	-2.638596	0.223815	1.716038
13	6	-2.788533	-1.770041	0.293194
14	6	-3.191086	-1.000407	-0.993033
15	8	-2.349364	-0.093265	-1.395843
16	6	1.815610	-2.771825	0.229125
17	6	1.001800	-3.162364	-1.034378
18	8	0.041534	-2.349368	-1.365401
19	6	2.811595	1.803292	0.087808
20	6	3.148787	0.935530	-1.158641
21	8	2.294388	0.004762	-1.449816
22	6	-1.770997	2.820720	0.147421
23	6	-0.974129	3.146032	-1.147998
24	8	-0.065337	2.284772	-1.484943
25	1	-0.857480	-1.005639	2.961790
26	1	-1.980169	-2.331720	2.756416
27	1	-0.496944	-3.322230	0.992381
28	1	0.233676	-3.213295	2.592999
29	1	1.166135	-0.839510	2.928088
30	1	2.483413	-1.961940	2.667760
31	1	3.405153	-0.479905	0.871926
32	1	3.348401	0.261143	2.469546
33	1	0.998958	1.205043	2.885028
34	1	2.108647	2.518885	2.558922
35	1	0.555877	3.412547	0.810336
36	1	-0.119662	3.382501	2.438054
37	1	-1.036924	1.023202	2.920598
38	1	-2.362718	2.133661	2.650387
39	1	-3.341487	0.573297	0.955023
40	1	-3.235496	-0.091345	2.583882
41	1	-2.163736	-2.596203	-0.069795
42	1	2.632369	-2.150232	-0.158127
43	1	2.172337	2.607249	-0.298555
44	1	-2.597357	2.181490	-0.188469

45	8	-0.451411	-0.511595	-3.240152
46	1	-0.278528	-1.445097	-2.982797
47	1	-1.396484	-0.380069	-3.004841
48	6	4.073870	2.425699	0.696845
49	1	4.753908	1.663834	1.089351
50	1	4.601112	2.963572	-0.092739
51	1	3.838652	3.131492	1.498917
52	6	-2.356751	4.089424	0.779327
53	1	-1.573327	4.773551	1.118492
54	1	-2.940745	4.608518	0.017604
55	1	-3.014012	3.863411	1.624202
56	6	-4.017305	-2.353816	1.002283
57	1	-4.683671	-1.568772	1.371559
58	1	-4.576522	-2.948607	0.278253
59	1	-3.741744	-3.001233	1.839597
60	6	2.416311	-4.009592	0.908691
61	1	1.641311	-4.672385	1.304910
62	1	2.978209	-4.568294	0.158470
63	1	3.097227	-3.742690	1.722113
64	8	-4.233684	-1.314709	-1.579961
65	8	-1.275465	4.160077	-1.793099
66	8	4.167218	1.203703	-1.811563
67	8	1.332082	-4.170869	-1.670014
68	59	0.007136	0.011837	-0.613218

E(RTPSSh) = -1712.1107368 Hartree
Zero-point correction = 0.561138 Hartree/particle
Sum of electronic and thermal Energies = -1711.514132 Hartree
Sum of electronic and thermal Enthalpies = -1711.513188 Hartree
Sum of electronic and thermal Free Energies = -1711.614117 Hartree

Table S22. [Nd(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.893867	-0.913947	1.138262
2	6	-1.264478	-1.710181	2.222805
3	6	-0.140357	-2.621549	1.734679
4	7	0.990293	-1.878438	1.114055
5	6	1.829867	-1.247932	2.164529
6	6	2.717593	-0.120828	1.642069
7	7	1.948152	1.001069	1.037843
8	6	1.358781	1.853748	2.099793
9	6	0.215392	2.738522	1.608553
10	7	-0.930371	1.964241	1.057189
11	6	-1.739649	1.383785	2.157936
12	6	-2.644066	0.237593	1.710520
13	6	-2.795667	-1.747373	0.279920
14	6	-3.174182	-0.969554	-1.008380
15	8	-2.324048	-0.061321	-1.390516
16	6	1.786849	-2.783877	0.225602
17	6	0.972434	-3.149073	-1.044550
18	8	0.015810	-2.326283	-1.362227
19	6	2.814264	1.777734	0.093653
20	6	3.137882	0.907380	-1.153633
21	8	2.275840	-0.018983	-1.436892
22	6	-1.748537	2.824162	0.142208
23	6	-0.937586	3.136345	-1.146855
24	8	-0.034648	2.264090	-1.471153

25	1	-0.875113	-1.007913	2.964735
26	1	-2.007352	-2.325536	2.748832
27	1	-0.526218	-3.319084	0.986845
28	1	0.204634	-3.222408	2.588301
29	1	1.158189	-0.861165	2.935373
30	1	2.468191	-1.990095	2.663714
31	1	3.394925	-0.507087	0.876120
32	1	3.342625	0.233632	2.474221
33	1	0.996872	1.190191	2.889545
34	1	2.119349	2.495033	2.566713
35	1	0.573974	3.404912	0.819740
36	1	-0.106153	3.374969	2.445353
37	1	-1.045869	1.027723	2.924078
38	1	-2.361804	2.148509	2.643381
39	1	-3.339785	0.591047	0.944964
40	1	-3.247876	-0.075508	2.574260
41	1	-2.177539	-2.580076	-0.078496
42	1	2.615573	-2.172678	-0.152204
43	1	2.182973	2.588755	-0.290626
44	1	-2.577884	2.191298	-0.197875
45	8	-0.414342	-0.461612	-3.217634
46	1	-0.257004	-1.401003	-2.972525
47	1	-1.361019	-0.323148	-2.993124
48	60	0.009473	0.012587	-0.592141
49	6	4.084169	2.384502	0.702398
50	1	4.756217	1.613870	1.091450
51	1	4.616267	2.920014	-0.085587
52	1	3.857836	3.090042	1.507337
53	6	-2.325497	4.100040	0.767473
54	1	-1.537073	4.777053	1.109274
55	1	-2.900924	4.623155	0.001935
56	1	-2.989233	3.882289	1.609441
57	6	-4.038960	-2.318123	0.973858
58	1	-4.703149	-1.526527	1.332927
59	1	-4.593923	-2.909362	0.243607
60	1	-3.779697	-2.966197	1.815877
61	6	2.363924	-4.037605	0.896266
62	1	1.576173	-4.696404	1.273526
63	1	2.929758	-4.592942	0.146410
64	1	3.036925	-3.789268	1.722040
65	8	-4.204958	-1.280592	-1.617373
66	8	-1.219458	4.152682	-1.797158
67	8	4.151707	1.170299	-1.815841
68	8	1.298630	-4.148039	-1.696906

E(RTPSSh) = -1712.7334131 Hartree
Zero-point correction = 0.561120 Hartree/particle
Sum of electronic and thermal Energies = -1712.136829 Hartree
Sum of electronic and thermal Enthalpies = -1712.135885 Hartree
Sum of electronic and thermal Free Energies = -1712.236808 Hartree

Table S23. [Sm(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.721883	-1.203526	1.133408
2	6	-0.983300	-1.898035	2.217766
3	6	0.271966	-2.614938	1.727580
4	7	1.266120	-1.693267	1.112139

5	6	1.994608	-0.947062	2.169036
6	6	2.693408	0.306556	1.650245
7	7	1.752152	1.287617	1.042403
8	6	1.042355	2.041510	2.105699
9	6	-0.226273	2.731768	1.612294
10	7	-1.227364	1.778568	1.057706
11	6	-1.939378	1.082895	2.158473
12	6	-2.650186	-0.190313	1.707278
13	6	-2.475634	-2.161745	0.262639
14	6	-2.955033	-1.437188	-1.021370
15	8	-2.251885	-0.401023	-1.375410
16	6	2.194598	-2.453942	0.215455
17	6	1.437468	-2.930052	-1.051961
18	8	0.369105	-2.253342	-1.358174
19	6	2.481761	2.185719	0.090828
20	6	2.940139	1.358505	-1.141640
21	8	2.215740	0.319367	-1.420210
22	6	-2.166356	2.490487	0.131823
23	6	-1.393661	2.930204	-1.141378
24	8	-0.373175	2.195131	-1.457327
25	1	-0.712200	-1.149405	2.966689
26	1	-1.622758	-2.627012	2.734433
27	1	0.002687	-3.360967	0.975779
28	1	0.711066	-3.157206	2.577066
29	1	1.269207	-0.670990	2.938691
30	1	2.739804	-1.582810	2.666964
31	1	3.427504	0.033318	0.888239
32	1	3.249339	0.758862	2.483779
33	1	0.788565	1.332405	2.897884
34	1	1.695465	2.795105	2.567008
35	1	0.022719	3.448004	0.825177
36	1	-0.650286	3.307037	2.447590
37	1	-1.201404	0.838709	2.926873
38	1	-2.674626	1.742392	2.639903
39	1	-3.391588	0.050771	0.941301
40	1	-3.197904	-0.600135	2.567792
41	1	-1.733696	-2.885693	-0.096660
42	1	2.913551	-1.716926	-0.163065
43	1	1.728011	2.877864	-0.304826
44	1	-2.871447	1.727931	-0.221686
45	8	-0.304017	-0.441112	-3.177978
46	1	-0.010332	-1.352214	-2.954315
47	1	-1.262857	-0.453539	-2.963093
48	6	3.635592	2.995710	0.692698
49	1	4.419098	2.347522	1.096291
50	1	4.080602	3.597557	-0.101517
51	1	3.296384	3.669023	1.485536
52	6	-2.955336	3.651512	0.747419
53	1	-2.294346	4.445831	1.106326
54	1	-3.594785	4.078368	-0.027130
55	1	-3.590571	3.324328	1.575748
56	6	-3.621660	-2.922629	0.940181
57	1	-4.401313	-2.245682	1.301363
58	1	-4.074502	-3.584282	0.199799
59	1	-3.271836	-3.532245	1.778026
60	6	2.964635	-3.605304	0.874312
61	1	2.291859	-4.381032	1.251608
62	1	3.606383	-4.061404	0.118603
63	1	3.595595	-3.259271	1.697941
64	8	-3.917250	-1.893574	-1.650077
65	8	-1.804585	3.903570	-1.788363
66	8	3.919088	1.746748	-1.794327

67	8	1.899317	-3.868707	-1.711490
68	62	0.007717	0.014865	-0.565502

E(RTPSSh) = -1713.9543252 Hartree
Zero-point correction = 0.560421 Hartree/particle
Sum of electronic and thermal Energies = -1713.358334 Hartree
Sum of electronic and thermal Enthalpies = -1713.357390 Hartree
Sum of electronic and thermal Free Energies = -1713.458503 Hartree

Table S24. [Eu(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.749346	-1.154337	1.135006
2	6	-1.035055	-1.874909	2.218637
3	6	0.201087	-2.622808	1.728416
4	7	1.216681	-1.725360	1.112354
5	6	1.970032	-1.003875	2.168986
6	6	2.701046	0.230416	1.649631
7	7	1.784183	1.236021	1.044953
8	6	1.099390	2.009247	2.110450
9	6	-0.149931	2.733114	1.617829
10	7	-1.174202	1.805537	1.062854
11	6	-1.906155	1.132156	2.164164
12	6	-2.651391	-0.119945	1.712009
13	6	-2.527921	-2.086439	0.258448
14	6	-2.977403	-1.343254	-1.024678
15	8	-2.245843	-0.322291	-1.366527
16	6	2.119659	-2.508913	0.209777
17	6	1.341040	-2.958857	-1.053079
18	8	0.290263	-2.250112	-1.349473
19	6	2.534574	2.113359	0.090514
20	6	2.957789	1.274508	-1.145751
21	8	2.209571	0.248590	-1.410218
22	6	-2.092069	2.542110	0.135619
23	6	-1.309016	2.948033	-1.141544
24	8	-0.300920	2.190367	-1.444358
25	1	-0.745899	-1.138194	2.972833
26	1	-1.697125	-2.588211	2.728719
27	1	-0.087180	-3.361996	0.976820
28	1	0.627344	-3.175740	2.577489
29	1	1.255613	-0.709777	2.942440
30	1	2.698555	-1.662616	2.661640
31	1	3.425074	-0.062238	0.885162
32	1	3.271319	0.667467	2.481527
33	1	0.827219	1.307481	2.903175
34	1	1.774825	2.743986	2.570130
35	1	0.118077	3.442441	0.830722
36	1	-0.559720	3.319086	2.452717
37	1	-1.175695	0.867023	2.933004
38	1	-2.622027	1.813358	2.644784
39	1	-3.387292	0.142108	0.947612
40	1	-3.208404	-0.517734	2.572114
41	1	-1.807301	-2.831563	-0.099867
42	1	2.856583	-1.791513	-0.171021
43	1	1.799938	2.828370	-0.299857
44	1	-2.822307	1.801564	-0.212706
45	8	-0.305470	-0.420102	-3.168989
46	1	-0.044624	-1.339876	-2.940087

47	1	-1.263567	-0.397321	-2.952064
48	6	3.716912	2.886489	0.685273
49	1	4.479531	2.214341	1.089710
50	1	4.178950	3.468336	-0.114143
51	1	3.402772	3.574694	1.475733
52	6	-2.840747	3.732492	0.745669
53	1	-2.153053	4.505647	1.100661
54	1	-3.464556	4.176798	-0.031832
55	1	-3.486973	3.431352	1.575479
56	6	-3.701286	-2.812586	0.927826
57	1	-4.462726	-2.112563	1.283909
58	1	-4.168872	-3.460174	0.184156
59	1	-3.376035	-3.432933	1.767764
60	6	2.860033	-3.683851	0.861052
61	1	2.167703	-4.440615	1.241475
62	1	3.482677	-4.156829	0.099696
63	1	3.506956	-3.358347	1.680768
64	8	-3.945307	-1.771035	-1.664709
65	8	-1.701154	3.918870	-1.803894
66	8	3.933717	1.642086	-1.814824
67	8	1.769609	-3.908734	-1.718958
68	63	0.009837	0.015256	-0.552773

E(RTPSSh) = -1714.5519019 Hartree
Zero-point correction = 0.561011 Hartree/particle
Sum of electronic and thermal Energies = -1713.955534 Hartree
Sum of electronic and thermal Enthalpies = -1713.954590 Hartree
Sum of electronic and thermal Free Energies = -1714.054459 Hartree

Table S25. [Gd(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.438246	-1.523052	1.136097
2	6	-0.580698	-2.063353	2.220228
3	6	0.794155	-2.510273	1.731851
4	7	1.576212	-1.405143	1.114999
5	6	2.146940	-0.531847	2.171008
6	6	2.577352	0.836941	1.651743
7	7	1.455061	1.607016	1.050048
8	6	0.612866	2.204701	2.114624
9	6	-0.769510	2.623679	1.622319
10	7	-1.556110	1.488002	1.068210
11	6	-2.114815	0.664777	2.168260
12	6	-2.553017	-0.724774	1.714525
13	6	-1.977062	-2.603424	0.252155
14	6	-2.574906	-1.976907	-1.032534
15	8	-2.099943	-0.810452	-1.362594
16	6	2.630913	-1.957487	0.207455
17	6	1.970826	-2.561800	-1.057761
18	8	0.783964	-2.110761	-1.343783
19	6	1.983113	2.631152	0.092644
20	6	2.569585	1.907821	-1.150000
21	8	2.082030	0.731850	-1.395768
22	6	-2.616893	1.995605	0.140856
23	6	-1.944967	2.554715	-1.141623
24	8	-0.785505	2.051606	-1.430245
25	1	-0.467835	-1.278638	2.972937
26	1	-1.064458	-2.907872	2.730588

27	1	0.684807	-3.297848	0.981776
28	1	1.335152	-2.948892	2.582406
29	1	1.384324	-0.409172	2.944644
30	1	3.006579	-1.007928	2.663197
31	1	3.346257	0.717625	0.884229
32	1	3.035377	1.392602	2.482317
33	1	0.511673	1.461766	2.910320
34	1	1.101967	3.076090	2.572138
35	1	-0.670598	3.374075	0.833554
36	1	-1.302087	3.102007	2.456637
37	1	-1.343348	0.574876	2.937675
38	1	-2.968491	1.162596	2.649111
39	1	-3.330946	-0.638730	0.951304
40	1	-3.002677	-1.241035	2.574634
41	1	-1.102525	-3.162619	-0.100780
42	1	3.184626	-1.088507	-0.166451
43	1	1.103785	3.165769	-0.286494
44	1	-3.165849	1.108721	-0.197260
45	8	-0.183992	-0.448430	-3.160994
46	1	0.273628	-1.288412	-2.934810
47	1	-1.123728	-0.642358	-2.951480
48	64	0.005629	0.017452	-0.537450
49	6	2.969293	3.648007	0.678646
50	1	3.869806	3.164264	1.068483
51	1	3.276630	4.322470	-0.122517
52	1	2.518337	4.243792	1.477714
53	6	-3.611524	2.994154	0.743882
54	1	-3.115579	3.906934	1.086929
55	1	-4.321746	3.276874	-0.035061
56	1	-4.169981	2.563859	1.580543
57	6	-2.956911	-3.584094	0.908715
58	1	-3.862799	-3.081993	1.261033
59	1	-3.255607	-4.317468	0.157789
60	1	-2.503087	-4.117413	1.749154
61	6	3.622801	-2.937091	0.847572
62	1	3.125362	-3.836616	1.222172
63	1	4.334223	-3.246952	0.080196
64	1	4.180287	-2.477857	1.669197
65	8	-3.411168	-2.614723	-1.683876
66	8	-2.551558	3.395289	-1.820905
67	8	3.416228	2.491992	-1.840697
68	8	2.601769	-3.382838	-1.733870

E(RTPSSh) = -1715.1427197 Hartree
Zero-point correction = 0.561633 Hartree/particle
Sum of electronic and thermal Energies = -1714.545835 Hartree
Sum of electronic and thermal Enthalpies = -1714.544891 Hartree
Sum of electronic and thermal Free Energies = -1714.644311 Hartree

Table S26. [Tb(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.361218	-1.586820	1.136151
2	6	-0.484508	-2.093792	2.221305
3	6	0.910044	-2.471343	1.732446
4	7	1.635212	-1.326966	1.117275
5	6	2.168522	-0.432224	2.174607
6	6	2.530760	0.955655	1.656149

7	7	1.371165	1.667874	1.052924
8	6	0.505472	2.230473	2.117449
9	6	-0.895156	2.579881	1.625241
10	7	-1.621795	1.405013	1.070372
11	6	-2.148253	0.560344	2.170070
12	6	-2.516427	-0.847763	1.714163
13	6	-1.846516	-2.687785	0.245469
14	6	-2.473443	-2.081615	-1.034446
15	8	-2.047392	-0.894742	-1.358957
16	6	2.713085	-1.827328	0.206278
17	6	2.077764	-2.459740	-1.057222
18	8	0.872165	-2.057951	-1.340804
19	6	1.851594	2.713332	0.092780
20	6	2.476507	2.011531	-1.142842
21	8	2.036593	0.816394	-1.387849
22	6	-2.702705	1.861607	0.138720
23	6	-2.051148	2.455490	-1.137759
24	8	-0.870092	2.002678	-1.423132
25	1	-0.410909	-1.310154	2.980210
26	1	-0.930252	-2.963558	2.723701
27	1	0.838560	-3.261453	0.980519
28	1	1.473971	-2.884882	2.580560
29	1	1.404138	-0.347477	2.951795
30	1	3.051493	-0.868909	2.661953
31	1	3.305764	0.874438	0.889789
32	1	2.959416	1.534800	2.486216
33	1	0.440378	1.486891	2.916436
34	1	0.954442	3.125574	2.570204
35	1	-0.833165	3.334464	0.836715
36	1	-1.452852	3.030607	2.458417
37	1	-1.378433	0.507538	2.944731
38	1	-3.027090	1.019052	2.644316
39	1	-3.295922	-0.798263	0.949375
40	1	-2.941604	-1.388979	2.571260
41	1	-0.945902	-3.202358	-0.109982
42	1	3.224837	-0.932860	-0.167540
43	1	0.948763	3.203423	-0.291518
44	1	-3.206615	0.949912	-0.203842
45	8	-0.154321	-0.429207	-3.144265
46	1	0.335956	-1.251027	-2.919767
47	1	-1.086267	-0.664166	-2.943496
48	65	0.004773	0.018586	-0.534929
49	6	2.787040	3.777209	0.677806
50	1	3.705342	3.336677	1.077208
51	1	3.069917	4.458566	-0.126530
52	1	2.305007	4.358699	1.469289
53	6	-3.745404	2.811061	0.739238
54	1	-3.293927	3.745532	1.085164
55	1	-4.465105	3.061582	-0.042010
56	1	-4.286803	2.354423	1.573128
57	6	-2.780135	-3.716667	0.894922
58	1	-3.710181	-3.259689	1.245602
59	1	-3.041224	-4.460319	0.140069
60	1	-2.304813	-4.231709	1.734813
61	6	3.749636	-2.762260	0.841880
62	1	3.294466	-3.685390	1.212700
63	1	4.474156	-3.035886	0.072969
64	1	4.286448	-2.281781	1.665134
65	8	-3.284531	-2.749030	-1.687248
66	8	-2.687610	3.275736	-1.813908
67	8	3.308126	2.624191	-1.826994
68	8	2.740043	-3.254507	-1.734565

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---------------------------------------------------------------------
 E(RTPSSh) = -1715.7308748 Hartree
 Zero-point correction = 0.561381 Hartree/particle
 Sum of electronic and thermal Energies = -1715.134223 Hartree
 Sum of electronic and thermal Enthalpies = -1715.133279 Hartree
 Sum of electronic and thermal Free Energies = -1715.232692 Hartree
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Table S27. [Dy(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.338908	-1.602055	1.136612
2	6	-0.458725	-2.101230	2.222242
3	6	0.940048	-2.460045	1.732829
4	7	1.648180	-1.304681	1.118397
5	6	2.172765	-0.406445	2.176761
6	6	2.516969	0.985433	1.658256
7	7	1.347094	1.680698	1.055060
8	6	0.477001	2.234750	2.120274
9	6	-0.927267	2.566191	1.627559
10	7	-1.636488	1.381172	1.071778
11	6	-2.155270	0.531865	2.171410
12	6	-2.504979	-0.880023	1.714273
13	6	-1.808999	-2.706486	0.242341
14	6	-2.436419	-2.102672	-1.037664
15	8	-2.026442	-0.907048	-1.351170
16	6	2.729929	-1.790029	0.204125
17	6	2.097043	-2.423696	-1.059223
18	8	0.884017	-2.037053	-1.332904
19	6	1.813852	2.729764	0.092195
20	6	2.441916	2.030020	-1.142323
21	8	2.014712	0.828678	-1.379332
22	6	-2.720461	1.823964	0.136993
23	6	-2.069952	2.421212	-1.137815
24	8	-0.882399	1.980519	-1.415213
25	1	-0.395164	-1.318666	2.983176
26	1	-0.894996	-2.977544	2.721603
27	1	0.878486	-3.249977	0.979886
28	1	1.510839	-2.866711	2.579607
29	1	1.408490	-0.331354	2.955056
30	1	3.061539	-0.833750	2.661900
31	1	3.292699	0.914066	0.891665
32	1	2.937821	1.571330	2.487502
33	1	0.420966	1.491310	2.920060
34	1	0.916234	3.135584	2.571202
35	1	-0.874446	3.321618	0.839190
36	1	-1.492336	3.008997	2.459939
37	1	-1.386402	0.488323	2.947583
38	1	-3.040431	0.980765	2.643294
39	1	-3.283952	-0.839647	0.948477
40	1	-2.923745	-1.428462	2.569875
41	1	-0.902046	-3.210951	-0.111277
42	1	3.231476	-0.889227	-0.168141
43	1	0.905350	3.209792	-0.291211
44	1	-3.213791	0.906517	-0.205442
45	8	-0.146493	-0.416752	-3.135998
46	1	0.349555	-1.233699	-2.906560
47	1	-1.076054	-0.657088	-2.930754
48	6	2.740287	3.803819	0.672732

49	1	3.663475	3.373158	1.071596
50	1	3.015103	4.485850	-0.133855
51	1	2.254194	4.382670	1.463683
52	6	-3.773998	2.764060	0.733022
53	1	-3.332861	3.703746	1.078112
54	1	-4.494456	3.005688	-0.050357
55	1	-4.312673	2.303683	1.566642
56	6	-2.734753	-3.746054	0.885876
57	1	-3.670475	-3.299209	1.234465
58	1	-2.985665	-4.490358	0.128190
59	1	-2.257344	-4.258464	1.726207
60	6	3.777138	-2.716591	0.834307
61	1	3.331969	-3.644898	1.204316
62	1	4.501925	-2.981299	0.062497
63	1	4.311784	-2.232973	1.657152
64	8	-3.232512	-2.777806	-1.700774
65	8	-2.711029	3.233187	-1.819541
66	8	3.263515	2.648477	-1.833238
67	8	2.765585	-3.205558	-1.745240
68	66	0.004203	0.019019	-0.524394

E(RTPSSh) = -1716.3170192 Hartree
Zero-point correction = 0.561261 Hartree/particle
Sum of electronic and thermal Energies = -1715.720433 Hartree
Sum of electronic and thermal Enthalpies = -1715.719489 Hartree
Sum of electronic and thermal Free Energies = -1715.819036 Hartree

Table S28. [Ho(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.318696	-1.615109	1.135821
2	6	-0.435704	-2.106650	2.221913
3	6	0.966111	-2.449715	1.732095
4	7	1.659184	-1.284856	1.118297
5	6	2.174800	-0.384256	2.178439
6	6	2.504129	1.010536	1.660051
7	7	1.325660	1.691078	1.056469
8	6	0.452582	2.237119	2.123266
9	6	-0.954441	2.553044	1.630188
10	7	-1.648112	1.359314	1.073261
11	6	-2.158281	0.505940	2.173435
12	6	-2.493483	-0.908221	1.714519
13	6	-1.776854	-2.722285	0.238940
14	6	-2.405623	-2.119181	-1.039964
15	8	-2.007148	-0.917457	-1.344966
16	6	2.744467	-1.757978	0.202121
17	6	2.112993	-2.393309	-1.060344
18	8	0.895620	-2.015869	-1.328104
19	6	1.781807	2.743278	0.092102
20	6	2.411909	2.045206	-1.141823
21	8	1.993778	0.839570	-1.373269
22	6	-2.735266	1.790999	0.137115
23	6	-2.086056	2.391538	-1.136239
24	8	-0.893758	1.960363	-1.408094
25	1	-0.380325	-1.324498	2.983850
26	1	-0.863732	-2.988039	2.719558
27	1	0.912788	-3.239627	0.978592
28	1	1.542833	-2.850245	2.577705

29	1	1.409682	-0.317227	2.956555
30	1	3.067819	-0.803549	2.662804
31	1	3.280934	0.947322	0.893881
32	1	2.917726	1.602167	2.488774
33	1	0.404569	1.493294	2.923094
34	1	0.883677	3.142583	2.572778
35	1	-0.909384	3.309326	0.842240
36	1	-1.526027	2.988502	2.461893
37	1	-1.388762	0.469501	2.949204
38	1	-3.047754	0.946484	2.645196
39	1	-3.272654	-0.874505	0.948709
40	1	-2.906020	-1.463550	2.568627
41	1	-0.865288	-3.218739	-0.114180
42	1	3.238046	-0.852403	-0.169228
43	1	0.869313	3.216431	-0.290062
44	1	-3.220718	0.869393	-0.205191
45	8	-0.139669	-0.401692	-3.128041
46	1	0.361066	-1.215665	-2.898938
47	1	-1.067392	-0.648076	-2.922611
48	67	0.003629	0.019551	-0.516276
49	6	2.702245	3.824140	0.669379
50	1	3.629321	3.400462	1.066642
51	1	2.970545	4.507024	-0.138702
52	1	2.214076	4.400656	1.460802
53	6	-3.796623	2.723778	0.730533
54	1	-3.363731	3.668407	1.072459
55	1	-4.519637	2.956635	-0.053188
56	1	-4.330890	2.261403	1.565913
57	6	-2.696501	-3.769495	0.878613
58	1	-3.636206	-3.329802	1.225491
59	1	-2.940209	-4.514771	0.119504
60	1	-2.217494	-4.279293	1.719621
61	6	3.799166	-2.678181	0.828890
62	1	3.361376	-3.610944	1.196403
63	1	4.525757	-2.935062	0.056109
64	1	4.330344	-2.192913	1.653020
65	8	-3.192747	-2.798438	-1.709514
66	8	-2.731182	3.196872	-1.822019
67	8	3.227623	2.667138	-1.836577
68	8	2.784816	-3.168165	-1.751058

E(RTPSSh) = -1716.9009134 Hartree
Zero-point correction = 0.561363 Hartree/particle
Sum of electronic and thermal Energies = -1716.304245 Hartree
Sum of electronic and thermal Enthalpies = -1716.303301 Hartree
Sum of electronic and thermal Free Energies = -1716.402755 Hartree

Table S29. [Er(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.315479	-1.614070	1.136947
2	6	-0.434131	-2.108161	2.222816
3	6	0.967398	-2.449556	1.732032
4	7	1.657484	-1.282838	1.118826
5	6	2.175545	-0.385002	2.179684
6	6	2.503190	1.010004	1.661813
7	7	1.322812	1.687619	1.059115
8	6	0.451794	2.234635	2.126630

9	6	-0.954853	2.550251	1.633308
10	7	-1.645728	1.355559	1.075502
11	6	-2.157864	0.503595	2.175474
12	6	-2.491722	-0.910324	1.716014
13	6	-1.771906	-2.719100	0.236831
14	6	-2.397430	-2.110078	-1.040185
15	8	-1.997936	-0.906430	-1.336706
16	6	2.740281	-1.753719	0.198698
17	6	2.102303	-2.385429	-1.061747
18	8	0.884042	-2.005089	-1.321782
19	6	1.776452	2.738406	0.092169
20	6	2.403242	2.035833	-1.140228
21	8	1.982499	0.830011	-1.366010
22	6	-2.730702	1.786103	0.136475
23	6	-2.075887	2.383083	-1.135204
24	8	-0.882871	1.949679	-1.400855
25	1	-0.379250	-1.327816	2.986635
26	1	-0.862715	-2.990680	2.718043
27	1	0.914168	-3.238728	0.977714
28	1	1.545769	-2.850140	2.576451
29	1	1.412030	-0.319051	2.959487
30	1	3.069464	-0.805415	2.661503
31	1	3.279422	0.947802	0.894958
32	1	2.916417	1.602667	2.489940
33	1	0.404044	1.491407	2.927069
34	1	0.883780	3.140181	2.575198
35	1	-0.909800	3.306856	0.845639
36	1	-1.527946	2.984692	2.464469
37	1	-1.389596	0.467485	2.952540
38	1	-3.048023	0.944691	2.645451
39	1	-3.270727	-0.876859	0.950014
40	1	-2.903569	-1.467219	2.569404
41	1	-0.859917	-3.214445	-0.116331
42	1	3.232355	-0.847514	-0.172735
43	1	0.863172	3.210010	-0.289634
44	1	-3.215425	0.864282	-0.205862
45	8	-0.140150	-0.387644	-3.121958
46	1	0.357548	-1.203067	-2.892717
47	1	-1.068139	-0.629972	-2.913545
48	6	2.697861	3.820587	0.665080
49	1	3.624992	3.397946	1.063348
50	1	2.966041	4.500598	-0.145518
51	1	2.210250	4.399990	1.454701
52	6	-3.792731	2.720755	0.725452
53	1	-3.359890	3.664859	1.068960
54	1	-4.512380	2.954519	-0.061147
55	1	-4.330851	2.259177	1.558799
56	6	-2.693030	-3.767969	0.871349
57	1	-3.632456	-3.329134	1.220113
58	1	-2.937215	-4.509337	0.108517
59	1	-2.214730	-4.282361	1.709984
60	6	3.797035	-2.675325	0.819546
61	1	3.360581	-3.608014	1.188850
62	1	4.519392	-2.932334	0.042784
63	1	4.332836	-2.190798	1.641117
64	8	-3.182920	-2.784731	-1.716124
65	8	-2.716297	3.188234	-1.825534
66	8	3.218783	2.653267	-1.839121
67	8	2.768375	-3.160194	-1.757992
68	68	0.003522	0.019610	-0.508740

E(RTPSSh) = -1717.4839164 Hartree

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Zero-point correction = 0.561501 Hartree/particle
Sum of electronic and thermal Energies = -1716.887141 Hartree
Sum of electronic and thermal Enthalpies = -1716.886196 Hartree
Sum of electronic and thermal Free Energies = -1716.985548 Hartree
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Table S30. [Tm(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.435419	-1.506241	1.139962
2	6	-0.595459	-2.067634	2.226046
3	6	0.775564	-2.515930	1.735651
4	7	1.552001	-1.405990	1.122076
5	6	2.139874	-0.551257	2.181882
6	6	2.573263	0.814369	1.663637
7	7	1.447971	1.579352	1.062145
8	6	0.622942	2.192939	2.129444
9	6	-0.755806	2.614623	1.637581
10	7	-1.533875	1.476051	1.079965
11	6	-2.112535	0.666858	2.179170
12	6	-2.554443	-0.716357	1.719078
13	6	-1.973037	-2.571801	0.237902
14	6	-2.545527	-1.913223	-1.038521
15	8	-2.051837	-0.743491	-1.329281
16	6	2.593514	-1.957349	0.199687
17	6	1.906086	-2.531355	-1.061875
18	8	0.718895	-2.058531	-1.314645
19	6	1.979126	2.592007	0.094473
20	6	2.544626	1.842782	-1.139295
21	8	2.027661	0.674426	-1.362659
22	6	-2.581776	1.986297	0.139325
23	6	-1.883342	2.524866	-1.134213
24	8	-0.729111	1.996949	-1.398405
25	1	-0.480186	-1.292079	2.988392
26	1	-1.091475	-2.913662	2.722059
27	1	0.662298	-3.298987	0.981484
28	1	1.321265	-2.959318	2.580439
29	1	1.383688	-0.427235	2.962036
30	1	2.999312	-1.039286	2.662760
31	1	3.341677	0.693107	0.895872
32	1	3.031565	1.373397	2.491451
33	1	0.519352	1.455817	2.930375
34	1	1.123957	3.062486	2.577410
35	1	-0.655064	3.366505	0.850368
36	1	-1.293570	3.090951	2.469495
37	1	-1.349218	0.570836	2.956222
38	1	-2.965404	1.176561	2.648859
39	1	-3.328716	-0.623326	0.953073
40	1	-3.007968	-1.240219	2.572333
41	1	-1.101116	-3.135360	-0.113685
42	1	3.156436	-1.091945	-0.168026
43	1	1.104385	3.133610	-0.284569
44	1	-3.137201	1.103948	-0.199179
45	8	-0.173298	-0.370627	-3.118958
46	1	0.258269	-1.223182	-2.891864
47	1	-1.115159	-0.537945	-2.897366
48	69	0.007194	0.018833	-0.506423
49	6	2.984283	3.599261	0.663268
50	1	3.878437	3.105774	1.055304

51	1	3.299311	4.258118	-0.147956
52	1	2.547236	4.212962	1.456471
53	6	-3.568501	3.003001	0.723888
54	1	-3.064594	3.914032	1.059535
55	1	-4.270834	3.285106	-0.062488
56	1	-4.137194	2.588938	1.561865
57	6	-2.973115	-3.548909	0.867396
58	1	-3.877213	-3.041237	1.216026
59	1	-3.271641	-4.267404	0.101989
60	1	-2.536427	-4.100288	1.705224
61	6	3.575303	-2.961905	0.814987
62	1	3.067886	-3.859892	1.179418
63	1	4.276039	-3.270074	0.037023
64	1	4.146334	-2.524196	1.639089
65	8	-3.379743	-2.521293	-1.719043
66	8	-2.458542	3.374673	-1.828461
67	8	3.405761	2.391464	-1.840400
68	8	2.508733	-3.350557	-1.764694

E(RTPSSh) = -1718.0634798 Hartree
Zero-point correction = 0.562082 Hartree/particle
Sum of electronic and thermal Energies = -1717.466376 Hartree
Sum of electronic and thermal Enthalpies = -1717.465432 Hartree
Sum of electronic and thermal Free Energies = -1717.563984 Hartree

Table S31. [Yb(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	1.991233	-0.095141	-1.261722
2	6	1.721010	-1.065938	-2.350449
3	6	1.041816	-2.333180	-1.842216
4	7	-0.256999	-2.054336	-1.169276
5	6	-1.321373	-1.820671	-2.177031
6	6	-2.541882	-1.122834	-1.586157
7	7	-2.205036	0.193658	-0.978216
8	6	-2.079305	1.231455	-2.032126
9	6	-1.332599	2.469700	-1.547019
10	7	0.038717	2.155457	-1.059435
11	6	0.967840	1.991786	-2.206263
12	6	2.249052	1.259776	-1.823180
13	6	3.134502	-0.516754	-0.391229
14	6	3.127904	0.339650	0.897788
15	8	1.956951	0.770918	1.276301
16	6	-0.600415	-3.167988	-0.227608
17	6	0.315228	-3.096307	1.018805
18	8	0.853576	-1.933318	1.255127
19	6	-3.220597	0.558343	0.062707
20	6	-3.026838	-0.353308	1.301685
21	8	-1.837783	-0.855450	1.447212
22	6	0.501526	3.204036	-0.094533
23	6	-0.273799	3.048292	1.239473
24	8	-0.749103	1.863069	1.477446
25	1	1.082479	-0.571233	-3.087327
26	1	2.643122	-1.343424	-2.878911
27	1	1.688685	-2.835394	-1.117885
28	1	0.908003	-3.022922	-2.686737
29	1	-0.892750	-1.204738	-2.972021
30	1	-1.640964	-2.759177	-2.650316

31	1	-2.984608	-1.746937	-0.805629
32	1	-3.297938	-1.011480	-2.375412
33	1	-1.546497	0.784324	-2.876175
34	1	-3.062273	1.536253	-2.416498
35	1	-1.879926	2.934723	-0.722825
36	1	-1.302020	3.202641	-2.364906
37	1	0.438718	1.434719	-2.984264
38	1	1.233103	2.961514	-2.649467
39	1	2.791639	1.834118	-1.067346
40	1	2.897798	1.203671	-2.707922
41	1	2.889045	-1.526766	-0.043332
42	1	-1.597442	-2.933229	0.160372
43	1	-2.942122	1.557616	0.415690
44	1	1.534188	2.940656	0.160043
45	8	1.398795	-0.626214	3.683040
46	1	1.221342	-1.316548	3.009728
47	1	1.715227	0.094083	3.103374
48	6	-4.678617	0.585814	-0.408597
49	1	-5.018205	-0.398039	-0.744914
50	1	-5.303030	0.879875	0.436718
51	1	-4.833820	1.305091	-1.218195
52	6	0.472380	4.646256	-0.612313
53	1	-0.542717	4.973505	-0.854813
54	1	0.850258	5.298563	0.176894
55	1	1.102453	4.775724	-1.497296
56	6	4.512206	-0.538317	-1.061004
57	1	4.812804	0.454265	-1.408767
58	1	5.245709	-0.865465	-0.321969
59	1	4.544849	-1.230584	-1.907478
60	6	-0.624374	-4.575261	-0.835860
61	1	0.359518	-4.877954	-1.205808
62	1	-0.915320	-5.278483	-0.053584
63	1	-1.345123	-4.653767	-1.655002
64	8	4.183596	0.528286	1.511701
65	8	-0.363190	4.020344	2.000051
66	8	-3.973374	-0.518967	2.081394
67	8	0.454122	-4.099005	1.727010
68	70	-0.018290	0.004896	0.392415

E(RTPSSh) = -1718.6478812 Hartree
Zero-point correction = 0.561624 Hartree/particle
Sum of electronic and thermal Energies = -1718.050811 Hartree
Sum of electronic and thermal Enthalpies = -1718.049867 Hartree
Sum of electronic and thermal Free Energies = -1718.150084 Hartree

Table S32. [Lu(DOTMA)(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	1.888166	-0.686745	-1.237762
2	6	1.355764	-1.540933	-2.329287
3	6	0.326360	-2.547089	-1.827176
4	7	-0.837318	-1.892171	-1.168668
5	6	-1.781428	-1.369222	-2.187748
6	6	-2.740322	-0.332357	-1.611837
7	7	-2.028918	0.825078	-1.001846
8	6	-1.600305	1.778186	-2.056459
9	6	-0.522704	2.739047	-1.567659
10	7	0.688542	2.030668	-1.068427

11	6	1.540120	1.600175	-2.205109
12	6	2.543897	0.525007	-1.803343
13	6	2.846079	-1.423677	-0.352232
14	6	3.060557	-0.613688	0.948575
15	8	2.076606	0.172070	1.288958
16	6	-1.499335	-2.847353	-0.223111
17	6	-0.616085	-3.029068	1.034590
18	8	0.248810	-2.079051	1.257486
19	6	-2.895169	1.473375	0.036458
20	6	-2.988027	0.538421	1.269529
21	8	-1.992584	-0.282641	1.424820
22	6	1.430758	2.898914	-0.098013
23	6	0.627512	2.983505	1.224718
24	8	-0.180375	1.991692	1.453525
25	1	0.899445	-0.881172	-3.072712
26	1	2.160246	-2.080458	-2.847204
27	1	0.788937	-3.214816	-1.095789
28	1	0.002848	-3.170484	-2.671837
29	1	-1.189228	-0.922004	-2.990716
30	1	-2.365430	-2.177379	-2.649070
31	1	-3.358749	-0.789559	-0.835289
32	1	-3.420392	-0.001760	-2.408605
33	1	-1.222092	1.190637	-2.897336
34	1	-2.448982	2.359076	-2.442265
35	1	-0.913838	3.347290	-0.748107
36	1	-0.267529	3.427268	-2.384847
37	1	0.879720	1.219724	-2.989423
38	1	2.083758	2.447632	-2.644485
39	1	3.224971	0.919690	-1.044288
40	1	3.155377	0.269927	-2.679558
41	1	2.315826	-2.323493	-0.020420
42	1	-2.389653	-2.329212	0.148149
43	1	-2.331212	2.340808	0.397011
44	1	2.334848	2.341300	0.168735
45	8	0.933459	-0.818332	3.645704
46	1	0.624162	-1.487479	2.999184
47	1	1.519065	-0.285222	3.071819
48	71	-0.011869	0.011555	0.382104
49	6	-4.274723	1.938705	-0.441679
50	1	-4.890960	1.104227	-0.788832
51	1	-4.789823	2.401221	0.402118
52	1	-4.201446	2.677050	-1.245769
53	6	1.835006	4.283541	-0.616064
54	1	0.964387	4.900352	-0.856647
55	1	2.393117	4.792360	0.171709
56	1	2.473277	4.218299	-1.502171
57	6	4.173867	-1.834503	-0.997130
58	1	4.758490	-0.967090	-1.316605
59	1	4.761643	-2.373465	-0.252021
60	1	4.023401	-2.491995	-1.858186
61	6	-1.923775	-4.194284	-0.820285
62	1	-1.065757	-4.777835	-1.166140
63	1	-2.423689	-4.769536	-0.039156
64	1	-2.619940	-4.068398	-1.654423
65	8	4.094238	-0.776640	1.604885
66	8	0.817410	3.939926	1.986098
67	8	-3.952952	0.643311	2.036837
68	8	-0.792574	-4.010826	1.762971

E(RTPSSh) = -1719.2010682 Hartree
Zero-point correction = 0.561134 Hartree/particle
Sum of electronic and thermal Energies = -1718.604389 Hartree

Sum of electronic and thermal Enthalpies = -1718.603445 Hartree
Sum of electronic and thermal Free Energies = -1718.703875 Hartree

Table S33. [La(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	2.080002	-0.641511	1.574643
2	6	1.581235	-1.549085	2.648928
3	6	0.591251	-2.616757	2.187004
4	7	-0.655953	-2.059508	1.576956
5	6	-1.548282	-1.549096	2.659037
6	6	-2.627190	-0.566315	2.205463
7	7	-2.080892	0.676364	1.576644
8	6	-1.581737	1.587943	2.647469
9	6	-0.592758	2.655619	2.182920
10	7	0.653336	2.098381	1.572004
11	6	1.545860	1.587730	2.652752
12	6	2.624329	0.605431	2.197977
13	6	3.153569	-1.263475	0.720193
14	6	2.524055	-2.252403	-0.267106
15	8	1.414339	-1.953231	-0.814899
16	6	-1.296829	-3.127055	0.729188
17	6	-2.297290	-2.483907	-0.238156
18	8	-2.009138	-1.364455	-0.765596
19	6	-3.149241	1.297105	0.715641
20	6	-2.509291	2.275464	-0.276975
21	8	-1.393557	1.974783	-0.807918
22	6	1.294052	3.162813	0.722149
23	6	2.281429	2.514148	-0.255328
24	8	1.984780	1.394858	-0.777814
25	1	1.112635	-0.920649	3.409736
26	1	2.414287	-2.053519	3.154453
27	1	1.055865	-3.275979	1.451505
28	1	0.352442	-3.235832	3.062521
29	1	-0.907635	-1.067592	3.401422
30	1	-2.040146	-2.377729	3.184041
31	1	-3.299335	-1.038849	1.487296
32	1	-3.230924	-0.320458	3.089839
33	1	-1.112312	0.961967	3.409865
34	1	-2.415132	2.092850	3.152135
35	1	-1.059046	3.314126	1.447918
36	1	-0.353534	3.275766	3.057819
37	1	0.906239	1.106349	3.396141
38	1	2.039243	2.415519	3.177927
39	1	3.292601	1.077198	1.475647
40	1	3.231910	0.363384	3.080678
41	1	3.485979	-0.440941	0.073887
42	1	-0.486469	-3.465329	0.070224
43	1	-3.485732	0.471572	0.075331
44	1	0.480770	3.508348	0.070851
45	8	0.076050	-0.415575	-2.720476
46	1	0.620737	-1.217608	-2.807191
47	1	-0.764254	-0.617966	-3.161475
48	7	3.381999	3.165178	-0.573212
49	7	-3.163626	3.371381	-0.601644
50	7	-3.398926	-3.138712	-0.543693
51	7	3.177038	-3.353120	-0.573663
52	1	3.678906	4.080683	-0.187919

53	1	-4.075591	3.673306	-0.210976
54	1	-3.688641	-4.056142	-0.156786
55	1	4.084409	-3.656300	-0.171537
56	6	2.796293	-4.345191	-1.571423
57	1	2.686672	-3.880986	-2.556852
58	6	4.375048	2.770252	-1.564114
59	1	3.910256	2.634821	-2.545958
60	6	-2.779205	4.352526	-1.608518
61	1	-1.832301	4.834165	-1.343126
62	6	-4.401873	-2.748576	-1.526562
63	1	-3.945263	-2.608800	-2.511654
64	1	4.852356	1.825279	-1.284186
65	1	1.841728	-4.814065	-1.311068
66	1	-4.883105	-1.807115	-1.241579
67	1	-2.652196	3.875427	-2.585544
68	6	-5.453564	-3.907268	-1.563643
69	8	-5.212815	-4.864051	-0.762822
70	8	-6.399084	-3.759039	-2.366624
71	6	3.942144	-5.411536	-1.586628
72	8	4.883295	-5.185182	-0.763232
73	8	3.800332	-6.351943	-2.396356
74	6	5.434108	3.921989	-1.607458
75	8	5.205908	4.879444	-0.803790
76	8	6.372946	3.768540	-2.417426
77	6	-3.933290	5.408892	-1.653935
78	8	-4.880998	5.190472	-0.835974
79	8	-3.791013	6.336281	-2.478852
80	6	1.850802	4.373208	1.484654
81	1	2.771410	4.144304	2.028021
82	1	2.061750	5.182616	0.780297
83	1	1.114021	4.754860	2.195391
84	6	-4.365692	1.853016	1.468695
85	1	-5.175979	2.046513	0.760336
86	1	-4.740848	1.121852	2.188711
87	1	-4.147183	2.783035	2.000196
88	6	-1.838288	-4.343051	1.493546
89	1	-2.753414	-4.120695	2.048673
90	1	-2.053794	-5.150262	0.788082
91	1	-1.091409	-4.724617	2.193557
92	6	4.372175	-1.806681	1.478823
93	1	4.157404	-2.731610	2.020601
94	1	5.182429	-2.004851	0.771777
95	1	4.745278	-1.066521	2.190497
96	57	-0.003425	0.012275	-0.137353

E(RTPSSh) = -2542.8414576 Hartree
Zero-point correction = 0.780436 Hartree/particle
Sum of electronic and thermal Energies = -2542.007278 Hartree
Sum of electronic and thermal Enthalpies = -2542.006334 Hartree
Sum of electronic and thermal Free Energies = -2542.153601 Hartree

Table S34. [Ce(DOTMA-(gly)$_4$(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	2.092702	-0.576430	1.597003
2	6	1.618153	-1.493096	2.674670
3	6	0.662110	-2.591513	2.214232
4	7	-0.597538	-2.070279	1.597441

5	6	-1.512774	-1.595274	2.676926
6	6	-2.613816	-0.639436	2.220724
7	7	-2.094821	0.615848	1.594011
8	6	-1.622250	1.539234	2.667065
9	6	-0.666495	2.636574	2.202935
10	7	0.592372	2.113306	1.587895
11	6	1.506316	1.641093	2.668701
12	6	2.608026	0.684632	2.216314
13	6	3.179568	-1.173571	0.742138
14	6	2.571573	-2.168709	-0.251171
15	8	1.447370	-1.897109	-0.784439
16	6	-1.199042	-3.151687	0.738265
17	6	-2.208102	-2.534498	-0.236338
18	8	-1.949011	-1.402145	-0.754272
19	6	-3.174547	1.210444	0.728880
20	6	-2.552940	2.194531	-0.268761
21	8	-1.423885	1.919089	-0.785305
22	6	1.195118	3.188220	0.723338
23	6	2.189987	2.559574	-0.259244
24	8	1.922265	1.425102	-0.765980
25	1	1.128065	-0.875076	3.430606
26	1	2.464494	-1.969415	3.185454
27	1	1.148014	-3.240997	1.483967
28	1	0.436882	-3.213101	3.091462
29	1	-0.892843	-1.104141	3.430598
30	1	-1.986464	-2.442263	3.189124
31	1	-3.271268	-1.127806	1.499661
32	1	-3.226945	-0.408853	3.102692
33	1	-1.132691	0.925578	3.426942
34	1	-2.470078	2.016998	3.174128
35	1	-1.152997	3.284078	1.471417
36	1	-0.441556	3.260931	3.078521
37	1	0.886246	1.152148	3.423722
38	1	1.980496	2.488836	3.179464
39	1	3.263448	1.170459	1.491597
40	1	3.222740	0.460306	3.098759
41	1	3.496347	-0.341560	0.100481
42	1	-0.372601	-3.460671	0.085187
43	1	-3.493448	0.374559	0.093499
44	1	0.366013	3.503977	0.077304
45	8	0.004945	-0.430417	-2.657388
46	1	0.635600	-1.168365	-2.737978
47	1	-0.853165	-0.796954	-2.928734
48	7	3.264289	3.241333	-0.600302
49	7	-3.235041	3.266910	-0.613802
50	7	-3.282731	-3.222124	-0.562469
51	7	3.255934	-3.243123	-0.580557
52	1	3.539538	4.169169	-0.227817
53	1	-4.158614	3.548613	-0.234998
54	1	-3.549747	-4.151350	-0.185981
55	1	4.176809	-3.523750	-0.192230
56	6	2.898420	-4.230733	-1.591323
57	1	2.762525	-3.754084	-2.567478
58	6	4.255602	2.866843	-1.600872
59	1	3.782466	2.710813	-2.575661
60	6	-2.869237	4.246061	-1.629549
61	1	-1.938375	4.757034	-1.362358
62	6	-4.282220	-2.859443	-1.559442
63	1	-3.814751	-2.699747	-2.536405
64	1	4.763078	1.938044	-1.320123
65	1	1.963392	-4.736496	-1.329217
66	1	-4.798518	-1.935879	-1.277589

67	1	-2.720962	3.760986	-2.599618
68	6	-5.293828	-4.052433	-1.618588
69	8	-5.032992	-5.004754	-0.818610
70	8	-6.231086	-3.931711	-2.435482
71	6	4.080032	-5.256446	-1.636189
72	8	5.020645	-5.012142	-0.817269
73	8	3.962783	-6.186589	-2.461427
74	6	5.280194	4.048291	-1.666383
75	8	5.032868	5.006043	-0.868685
76	8	6.213871	3.914828	-2.485682
77	6	-4.051741	5.269281	-1.697133
78	8	-5.000259	5.033725	-0.884864
79	8	-3.927905	6.190683	-2.531595
80	6	1.723541	4.420786	1.470179
81	1	2.658139	4.225742	2.002844
82	1	1.899019	5.231511	0.757691
83	1	0.984036	4.783704	2.187990
84	6	-4.402760	1.746237	1.477490
85	1	-5.218728	1.911213	0.768487
86	1	-4.759083	1.015118	2.207131
87	1	-4.205946	2.687828	1.997024
88	6	-1.709801	-4.388285	1.490491
89	1	-2.638983	-4.199794	2.034702
90	1	-1.888659	-5.199127	0.779015
91	1	-0.959326	-4.747552	2.198492
92	6	4.407537	-1.696428	1.500169
93	1	4.213267	-2.633931	2.027934
94	1	5.226529	-1.864116	0.795358
95	1	4.757969	-0.956908	2.224023
96	58	-0.003313	0.013412	-0.088462

E(RTPSSh) = -2543.4937071 Hartree
Zero-point correction = 0.780811 Hartree/particle
Sum of electronic and thermal Energies = -2542.659361 Hartree
Sum of electronic and thermal Enthalpies = -2542.658417 Hartree
Sum of electronic and thermal Free Energies = -2542.804370 Hartree

Table S35. [Pr(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	2.103071	-0.525558	1.608697
2	6	1.650112	-1.453832	2.688390
3	6	0.717247	-2.573561	2.229797
4	7	-0.554226	-2.082811	1.610134
5	6	-1.485734	-1.629798	2.687663
6	6	-2.605622	-0.697403	2.227982
7	7	-2.114134	0.568659	1.599142
8	6	-1.665546	1.509563	2.669990
9	6	-0.731818	2.625344	2.203072
10	7	0.539933	2.128921	1.589987
11	6	1.469790	1.686626	2.672619
12	6	2.590310	0.749453	2.224193
13	6	3.199493	-1.099844	0.752531
14	6	2.604810	-2.099315	-0.243504
15	8	1.469965	-1.851165	-0.763344
16	6	-1.128195	-3.175261	0.748161
17	6	-2.137810	-2.574794	-0.235201
18	8	-1.898949	-1.434292	-0.743958

19	6	-3.202285	1.135933	0.728389
20	6	-2.591761	2.121337	-0.273323
21	8	-1.454647	1.862833	-0.778942
22	6	1.116006	3.209612	0.716225
23	6	2.117021	2.590692	-0.265236
24	8	1.869342	1.447188	-0.760521
25	1	1.146834	-0.845417	3.442895
26	1	2.507517	-1.911424	3.197059
27	1	1.217364	-3.215488	1.502590
28	1	0.505446	-3.197210	3.108648
29	1	-0.878916	-1.125530	3.442723
30	1	-1.942843	-2.487963	3.195499
31	1	-3.252092	-1.200701	1.507333
32	1	-3.224963	-0.479309	3.108487
33	1	-1.163886	0.909585	3.432355
34	1	-2.525417	1.971145	3.170883
35	1	-1.230946	3.261403	1.470170
36	1	-0.521579	3.256567	3.077041
37	1	0.862360	1.190535	3.432656
38	1	1.927606	2.549245	3.172325
39	1	3.236834	1.245397	1.498448
40	1	3.208996	0.542250	3.107769
41	1	3.502718	-0.261183	0.113490
42	1	-0.291793	-3.469910	0.101814
43	1	-3.505137	0.290711	0.098186
44	1	0.278847	3.504339	0.071270
45	8	-0.012532	-0.380518	-2.602461
46	1	0.653707	-1.082416	-2.710755
47	1	-0.860987	-0.800881	-2.822973
48	7	3.179060	3.287196	-0.618854
49	7	-3.288374	3.180866	-0.633280
50	7	-3.194341	-3.282505	-0.580656
51	7	3.309822	-3.156317	-0.590122
52	1	3.434107	4.215906	-0.246124
53	1	-4.212319	3.443777	-0.253836
54	1	-3.440924	-4.212084	-0.202908
55	1	4.234532	-3.411081	-0.205032
56	6	2.953768	-4.142693	-1.602173
57	1	2.804795	-3.661709	-2.574349
58	6	4.173523	2.906194	-1.613431
59	1	3.702524	2.744535	-2.588441
60	6	-2.915943	4.160813	-1.645437
61	1	-1.983039	4.665184	-1.373338
62	6	-4.188463	-2.922220	-1.583481
63	1	-3.713735	-2.758394	-2.556345
64	1	4.676238	1.977341	-1.324776
65	1	2.024721	-4.656262	-1.334438
66	1	-4.707717	-2.000012	-1.303001
67	1	-2.765631	3.676234	-2.615578
68	6	-5.199241	-4.110230	-1.660650
69	8	-4.952984	-5.074127	-0.871583
70	8	-6.132285	-3.979776	-2.484184
71	6	4.135624	-5.161404	-1.670451
72	8	5.092872	-4.923885	-0.870651
73	8	4.007290	-6.090954	-2.498103
74	6	5.204243	4.076728	-1.691466
75	8	4.968625	5.050655	-0.911672
76	8	6.141136	3.924668	-2.507195
77	6	-4.085288	5.192852	-1.724508
78	8	-5.051322	4.969257	-0.931401
79	8	-3.940119	6.118900	-2.553673
80	6	1.622350	4.458363	1.451034

81	1	2.559709	4.286181	1.986307
82	1	1.781173	5.266390	0.731744
83	1	0.875495	4.812478	2.165507
84	6	-4.442088	1.655540	1.468966
85	1	-5.258946	1.800719	0.756807
86	1	-4.786920	0.922038	2.201605
87	1	-4.263787	2.602848	1.984422
88	6	-1.623071	-4.420376	1.496637
89	1	-2.559737	-4.248856	2.033171
90	1	-1.778796	-5.236169	0.785556
91	1	-0.871626	-4.762906	2.211763
92	6	4.436498	-1.605036	1.507713
93	1	4.258999	-2.547709	2.031753
94	1	5.259296	-1.753290	0.803154
95	1	4.772175	-0.862163	2.235021
96	59	-0.005672	0.010748	-0.039447

E(RTPSSh) = -2544.1515271 Hartree
Zero-point correction = 0.781745 Hartree/particle
Sum of electronic and thermal Energies = -2543.316451 Hartree
Sum of electronic and thermal Enthalpies = -2543.315506 Hartree
Sum of electronic and thermal Free Energies = -2543.460710 Hartree

Table S36. [Nd(DOTMA-$(gly)_4(H_2O)]^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	2.094268	-0.566499	1.603985
2	6	1.627905	-1.488125	2.683098
3	6	0.678206	-2.592062	2.220965
4	7	-0.582964	-2.075263	1.602125
5	6	-1.505246	-1.612106	2.682754
6	6	-2.610412	-0.661381	2.225279
7	7	-2.094900	0.596471	1.599614
8	6	-1.631467	1.525049	2.674357
9	6	-0.681050	2.627323	2.209004
10	7	0.580084	2.108278	1.593119
11	6	1.501816	1.649228	2.675395
12	6	2.607200	0.696286	2.222798
13	6	3.174133	-1.160978	0.740755
14	6	2.549034	-2.140896	-0.255936
15	8	1.420064	-1.858597	-0.770798
16	6	-1.177915	-3.150615	0.733047
17	6	-2.170566	-2.520133	-0.248395
18	8	-1.899889	-1.383972	-0.751338
19	6	-3.169632	1.187338	0.727826
20	6	-2.534040	2.159074	-0.271431
21	8	-1.402338	1.873044	-0.774222
22	6	1.176362	3.177223	0.718337
23	6	2.159468	2.535074	-0.266046
24	8	1.883588	1.396480	-0.757657
25	1	1.131717	-0.872454	3.436220
26	1	2.477861	-1.958196	3.192803
27	1	1.168225	-3.240669	1.492716
28	1	0.453718	-3.214808	3.097253
29	1	-0.889426	-1.117446	3.436693
30	1	-1.973797	-2.464321	3.190043
31	1	-3.265280	-1.152319	1.503536
32	1	-3.226629	-0.433530	3.105454

33	1	-1.136737	0.913935	3.432210
34	1	-2.483231	1.997094	3.179232
35	1	-1.170529	3.273602	1.478433
36	1	-0.457557	3.253177	3.083477
37	1	0.885980	1.158295	3.431829
38	1	1.971368	2.503018	3.179244
39	1	3.261626	1.183592	1.498188
40	1	3.223535	0.474028	3.104326
41	1	3.493703	-0.326931	0.103798
42	1	-0.346916	-3.459842	0.086686
43	1	-3.489875	0.349776	0.095999
44	1	0.344015	3.490798	0.076116
45	8	-0.029286	-0.297090	-2.580162
46	1	0.632358	-0.993362	-2.737870
47	1	-0.883850	-0.707871	-2.796001
48	7	3.236606	3.205404	-0.624278
49	7	-3.202692	3.236472	-0.631222
50	7	-3.247445	-3.195004	-0.596054
51	7	3.220198	-3.218683	-0.605970
52	1	3.514856	4.127696	-0.252770
53	1	-4.119422	3.522505	-0.251605
54	1	-3.522344	-4.116676	-0.219090
55	1	4.135117	-3.504141	-0.220076
56	6	2.824156	-4.197474	-1.610653
57	1	2.695594	-3.718462	-2.586713
58	6	4.221614	2.795949	-1.617135
59	1	3.747840	2.646285	-2.592713
60	6	-2.800161	4.210968	-1.637257
61	1	-1.853075	4.685833	-1.361173
62	6	-4.236985	-2.794963	-1.588474
63	1	-3.766200	-2.648064	-2.565940
64	1	4.697969	1.853907	-1.326679
65	1	1.874233	-4.669341	-1.339491
66	1	-4.716627	-1.853627	-1.301254
67	1	-2.663003	3.727892	-2.610086
68	6	-5.294553	-3.941951	-1.657126
69	8	-5.076955	-4.917829	-0.874556
70	8	-6.231420	-3.771397	-2.468953
71	6	3.961768	-5.265659	-1.670759
72	8	4.929252	-5.062055	-0.874137
73	8	3.792815	-6.196045	-2.490264
74	6	5.285109	3.936707	-1.694969
75	8	5.074799	4.919170	-0.918784
76	8	6.219926	3.755350	-2.507111
77	6	-3.937645	5.278231	-1.711768
78	8	-4.911256	5.079569	-0.921430
79	8	-3.762771	6.204177	-2.535343
80	6	1.713784	4.413453	1.451839
81	1	2.648318	4.219391	1.984580
82	1	1.890095	5.217228	0.731789
83	1	0.977348	4.785469	2.168041
84	6	-4.398697	1.734223	1.466295
85	1	-5.209189	1.901743	0.751692
86	1	-4.763901	1.006888	2.195223
87	1	-4.199646	2.675344	1.985480
88	6	-1.703239	-4.388235	1.472661
89	1	-2.634460	-4.197486	2.012239
90	1	-1.881243	-5.193817	0.755101
91	1	-0.959635	-4.755827	2.183560
92	6	4.402098	-1.699058	1.487429
93	1	4.203642	-2.636773	2.012920
94	1	5.215369	-1.869545	0.776755

95	1	4.762877	-0.965430	2.212136
96	60	-0.000923	0.011064	-0.026247

E(RTPSSh) = -2544.7743983 Hartree
Zero-point correction = 0.782050 Hartree/particle
Sum of electronic and thermal Energies = -2543.939091 Hartree
Sum of electronic and thermal Enthalpies = -2543.938147 Hartree
Sum of electronic and thermal Free Energies = -2544.082530 Hartree

Table S37. [Sm(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	2.092101	-0.545250	1.628448
2	6	1.643699	-1.474947	2.708247
3	6	0.704019	-2.585544	2.244955
4	7	-0.559142	-2.074125	1.626362
5	6	-1.488695	-1.629372	2.708560
6	6	-2.600760	-0.688470	2.251093
7	7	-2.091053	0.571940	1.626528
8	6	-1.643983	1.506911	2.702742
9	6	-0.704372	2.616496	2.236174
10	7	0.558419	2.103308	1.619359
11	6	1.488336	1.661725	2.702029
12	6	2.600194	0.718984	2.247514
13	6	3.170988	-1.127728	0.756542
14	6	2.544095	-2.098503	-0.246195
15	8	1.403728	-1.821181	-0.738893
16	6	-1.142379	-3.148999	0.749514
17	6	-2.126127	-2.517974	-0.238704
18	8	-1.860710	-1.370976	-0.720596
19	6	-3.166106	1.151996	0.748512
20	6	-2.530130	2.115638	-0.256662
21	8	-1.387897	1.835962	-0.738847
22	6	1.142246	3.172030	0.736324
23	6	2.116955	2.528958	-0.254214
24	8	1.847376	1.379644	-0.724172
25	1	1.145138	-0.867489	3.466594
26	1	2.502234	-1.936948	3.210770
27	1	1.199149	-3.228648	1.515327
28	1	0.482961	-3.212372	3.119088
29	1	-0.880477	-1.132807	3.467523
30	1	-1.948836	-2.490358	3.208531
31	1	-3.249950	-1.184076	1.527397
32	1	-3.221130	-0.464932	3.129332
33	1	-1.145687	0.902845	3.464028
34	1	-2.503571	1.970042	3.202440
35	1	-1.199586	3.257212	1.504583
36	1	-0.483765	3.246164	3.108482
37	1	0.880654	1.168165	3.463438
38	1	1.949752	2.523941	3.198820
39	1	3.248508	1.211685	1.521093
40	1	3.221125	0.499672	3.126412
41	1	3.485170	-0.287130	0.126094
42	1	-0.304879	-3.453582	0.109901
43	1	-3.482436	0.308685	0.122911
44	1	0.303070	3.480167	0.100898
45	8	-0.037856	-0.244829	-2.499944
46	1	0.629484	-0.933811	-2.665833

47	1	-0.891199	-0.670832	-2.691502
48	7	3.178315	3.208614	-0.640170
49	7	-3.208596	3.177317	-0.643243
50	7	-3.187274	-3.201831	-0.614405
51	7	3.224013	-3.160773	-0.624467
52	1	3.446969	4.141268	-0.287720
53	1	-4.136149	3.453160	-0.282690
54	1	-3.453430	-4.133745	-0.256249
55	1	4.149753	-3.437372	-0.258076
56	6	2.831382	-4.126198	-1.643267
57	1	2.676545	-3.629669	-2.606615
58	6	4.150070	2.803743	-1.647812
59	1	3.658772	2.628896	-2.610390
60	6	-2.811604	4.137835	-1.664741
61	1	-1.880604	4.641148	-1.384349
62	6	-4.163315	-2.806499	-1.621930
63	1	-3.675022	-2.635148	-2.586696
64	1	4.652855	1.876850	-1.353210
65	1	1.896541	-4.624177	-1.366500
66	1	-4.668822	-1.880027	-1.330884
67	1	-2.647399	3.636288	-2.623865
68	6	-5.195034	-3.974186	-1.729671
69	8	-4.973404	-4.956690	-0.956505
70	8	-6.118330	-3.811108	-2.558262
71	6	3.990166	-5.168597	-1.743997
72	8	4.965627	-4.961413	-0.958063
73	8	3.827775	-6.084891	-2.580429
74	6	5.187332	3.965291	-1.765799
75	8	4.973884	4.954168	-0.998636
76	8	6.107530	3.791768	-2.596077
77	6	-3.971975	5.176571	-1.781293
78	8	-4.952922	4.973734	-1.001125
79	8	-3.805530	6.087303	-2.623321
80	6	1.681260	4.413032	1.460606
81	1	2.625351	4.227205	1.979199
82	1	1.839971	5.217756	0.737512
83	1	0.952585	4.779729	2.187515
84	6	-4.398834	1.700616	1.479665
85	1	-5.211075	1.851632	0.763367
86	1	-4.758295	0.980858	2.218999
87	1	-4.206864	2.650567	1.985179
88	6	-1.670322	-4.390808	1.480346
89	1	-2.611864	-4.207853	2.004481
90	1	-1.829779	-5.197971	0.760201
91	1	-0.935391	-4.752375	2.203360
92	6	4.403979	-1.668086	1.493289
93	1	4.213721	-2.615139	2.004832
94	1	5.217749	-1.821405	0.779273
95	1	4.760153	-0.942493	2.228389
96	62	-0.000325	0.012526	0.028995

E(RTPSSh) = -2545.9974591 Hartree
Zero-point correction = 0.782634 Hartree/particle
Sum of electronic and thermal Energies = -2545.161819 Hartree
Sum of electronic and thermal Enthalpies = -2545.160875 Hartree
Sum of electronic and thermal Free Energies = -2545.304276 Hartree

Table S38. [Eu(DOTMA-(gly)$_4$(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center	Atomic	Coordinates (Angstroms)

Number	Number	X	Y	Z
1	7	2.092667	-0.530070	1.642756
2	6	1.654223	-1.464488	2.722497
3	6	0.722760	-2.580814	2.258120
4	7	-0.542619	-2.075341	1.639681
5	6	-1.476302	-1.641420	2.722629
6	6	-2.594378	-0.708453	2.265465
7	7	-2.091045	0.554766	1.641930
8	6	-1.653439	1.492997	2.719149
9	6	-0.722425	2.608941	2.252319
10	7	0.542496	2.102271	1.634954
11	6	1.476670	1.670282	2.717811
12	6	2.594561	0.736125	2.262294
13	6	3.172965	-1.104075	0.767563
14	6	2.548312	-2.071416	-0.238927
15	8	1.401256	-1.800550	-0.720064
16	6	-1.117685	-3.151205	0.759218
17	6	-2.098504	-2.522158	-0.232254
18	8	-1.839540	-1.368677	-0.702663
19	6	-3.167847	1.127498	0.761803
20	6	-2.534266	2.088070	-0.246942
21	8	-1.386131	1.814724	-0.718654
22	6	1.118030	3.172672	0.748944
23	6	2.089963	2.532000	-0.245082
24	8	1.826929	1.376813	-0.704351
25	1	1.152050	-0.861933	3.482413
26	1	2.517510	-1.920061	3.222713
27	1	1.222136	-3.219578	1.527622
28	1	0.505015	-3.210344	3.131095
29	1	-0.872581	-1.141816	3.483229
30	1	-1.929646	-2.507575	3.219845
31	1	-3.239341	-1.207761	1.540561
32	1	-3.217206	-0.489116	3.142977
33	1	-1.151111	0.892883	3.480944
34	1	-2.517560	1.948942	3.217594
35	1	-1.222129	3.245790	1.520423
36	1	-0.504944	3.240615	3.123906
37	1	0.873566	1.172758	3.480313
38	1	1.931217	2.537157	3.212782
39	1	3.238531	1.233493	1.535215
40	1	3.218039	0.519895	3.140097
41	1	3.483286	-0.259436	0.140891
42	1	-0.276377	-3.451593	0.122878
43	1	-3.481144	0.280777	0.139565
44	1	0.274955	3.476840	0.117003
45	8	-0.034577	-0.224708	-2.461376
46	1	0.637226	-0.910367	-2.623939
47	1	-0.885516	-0.658750	-2.646405
48	63	-0.000077	0.013113	0.056944
49	7	3.140924	3.219436	-0.645160
50	7	-3.220774	3.139090	-0.647950
51	7	-3.148943	-3.213742	-0.622872
52	7	3.236197	-3.122656	-0.632652
53	1	3.401271	4.158421	-0.303107
54	1	-4.155094	3.406276	-0.298275
55	1	-3.407348	-4.151956	-0.275116
56	1	4.168924	-3.391084	-0.277653
57	6	2.849765	-4.079855	-1.661502
58	1	2.677294	-3.572308	-2.616044
59	6	4.104832	2.821588	-1.663031
60	1	3.602852	2.629583	-2.616751

61	6	-2.831093	4.090638	-1.680536
62	1	-1.912360	4.615634	-1.399356
63	6	-4.118024	-2.824545	-1.639430
64	1	-3.620291	-2.637707	-2.596448
65	1	4.628049	1.906369	-1.367445
66	1	1.926407	-4.597931	-1.383065
67	1	-4.641788	-1.908660	-1.347195
68	1	-2.648517	3.577247	-2.629980
69	6	-5.129881	-4.007384	-1.769622
70	8	-4.903024	-4.992147	-1.000817
71	8	-6.044564	-3.852537	-2.609144
72	6	4.025241	-5.100720	-1.787800
73	8	5.003793	-4.888577	-1.007005
74	8	3.871117	-6.007185	-2.636331
75	6	5.120093	3.999759	-1.806243
76	8	4.901264	4.991180	-1.043854
77	8	6.030411	3.835242	-2.649093
78	6	-4.009116	5.105943	-1.824200
79	8	-4.992560	4.898770	-1.048312
80	8	-3.852183	6.005026	-2.680417
81	6	1.655033	4.416566	1.469788
82	1	2.604070	4.236360	1.981245
83	1	1.803314	5.222745	0.746109
84	1	0.929390	4.778367	2.202221
85	6	-4.402859	1.674642	1.490284
86	1	-5.216875	1.815770	0.773997
87	1	-4.757381	0.958051	2.235119
88	1	-4.215840	2.629279	1.988725
89	6	-1.643920	-4.396229	1.485899
90	1	-2.590560	-4.219123	2.002784
91	1	-1.792760	-5.204487	0.764722
92	1	-0.912234	-4.753209	2.214521
93	6	4.408925	-1.642987	1.500464
94	1	4.224315	-2.595162	2.004462
95	1	5.224053	-1.785603	0.785791
96	1	4.760321	-0.920885	2.241329

E(RTPSSh) = -2546.5947156 Hartree
Zero-point correction = 0.782749 Hartree/particle
Sum of electronic and thermal Energies = -2545.758986 Hartree
Sum of electronic and thermal Enthalpies = -2545.758042 Hartree
Sum of electronic and thermal Free Energies = -2545.901914 Hartree

Table S39. [Gd(DOTMA-(gly)$_4$(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	2.099857	-0.495023	1.667631
2	6	1.678419	-1.437343	2.747110
3	6	0.765489	-2.568641	2.281997
4	7	-0.507958	-2.083713	1.664008
5	6	-1.450038	-1.667888	2.746651
6	6	-2.582350	-0.752385	2.289118
7	7	-2.098931	0.518973	1.666656
8	6	-1.678519	1.465149	2.743572
9	6	-0.766045	2.596027	2.275909
10	7	0.506978	2.109708	1.659081
11	6	1.449466	1.695916	2.741742
12	6	2.581775	0.779264	2.286133

13	6	3.187044	-1.050847	0.790340
14	6	2.574422	-2.018929	-0.222087
15	8	1.418175	-1.766369	-0.691353
16	6	-1.064080	-3.166399	0.780853
17	6	-2.045458	-2.549517	-0.216962
18	8	-1.804581	-1.387178	-0.675445
19	6	-3.182649	1.073387	0.784310
20	6	-2.560964	2.034507	-0.230564
21	8	-1.404208	1.778897	-0.691002
22	6	1.063744	3.186735	0.770034
23	6	2.036868	2.557787	-0.229671
24	8	1.793155	1.393313	-0.675965
25	1	1.167085	-0.843851	3.508201
26	1	2.549789	-1.878834	3.245909
27	1	1.275176	-3.197727	1.550227
28	1	0.558670	-3.202983	3.154153
29	1	-0.855662	-1.160472	3.509549
30	1	-1.890037	-2.542556	3.240884
31	1	-3.217672	-1.261389	1.562449
32	1	-3.210104	-0.543961	3.165777
33	1	-1.167076	0.874167	3.506589
34	1	-2.550758	1.907019	3.240534
35	1	-1.276035	3.223069	1.542622
36	1	-0.559435	3.232589	3.146573
37	1	0.855603	1.190623	3.506484
38	1	1.890476	2.571447	3.233658
39	1	3.216273	1.286346	1.557440
40	1	3.209889	0.573990	3.163265
41	1	3.487988	-0.199312	0.168599
42	1	-0.215522	-3.457005	0.149796
43	1	-3.486519	0.219620	0.167206
44	1	0.213506	3.480842	0.143092
45	8	-0.030112	-0.208973	-2.417566
46	1	0.651956	-0.885628	-2.575971
47	1	-0.874669	-0.659075	-2.594371
48	7	3.066943	3.264712	-0.649489
49	7	-3.266283	3.065654	-0.649950
50	7	-3.075762	-3.259884	-0.626642
51	7	3.281395	-3.050033	-0.634461
52	1	3.305471	4.215576	-0.324631
53	1	-4.211640	3.312730	-0.315387
54	1	-3.312716	-4.210004	-0.296086
55	1	4.225649	-3.298057	-0.295202
56	6	2.913318	-3.995261	-1.680930
57	1	2.707362	-3.470896	-2.619550
58	6	4.018283	2.885575	-1.686108
59	1	3.498818	2.658433	-2.622530
60	6	-2.894995	4.004502	-1.700746
61	1	-1.999893	4.569270	-1.420245
62	6	-4.031928	-2.889600	-1.662295
63	1	-3.516385	-2.666657	-2.601927
64	1	4.583793	1.995997	-1.389964
65	1	2.014045	-4.553809	-1.401748
66	1	-4.598677	-1.999986	-1.368927
67	1	-2.680118	3.474104	-2.633889
68	6	-4.993431	-4.107882	-1.839806
69	8	-4.751911	-5.095112	-1.078635
70	8	-5.888222	-3.974599	-2.704064
71	6	4.123041	-4.968885	-1.850467
72	8	5.103761	-4.743069	-1.076243
73	8	3.990007	-5.856539	-2.722078
74	6	4.983784	4.098581	-1.875762

75	8	4.749908	5.092707	-1.121394
76	8	5.874902	3.955196	-2.742626
77	6	-4.106203	4.972878	-1.886931
78	8	-5.091565	4.752768	-1.117056
79	8	-3.970037	5.852593	-2.766467
80	6	1.589709	4.437219	1.487919
81	1	2.547089	4.270325	1.988180
82	1	1.717168	5.247793	0.765230
83	1	0.866579	4.785448	2.229450
84	6	-4.423845	1.609962	1.510400
85	1	-5.241997	1.731875	0.795334
86	1	-4.765601	0.895487	2.263268
87	1	-4.249415	2.572122	1.998799
88	6	-1.579547	-4.417706	1.504760
89	1	-2.534881	-4.253937	2.009865
90	1	-1.706917	-5.230640	0.784772
91	1	-0.850769	-4.760578	2.243093
92	6	4.429104	-1.579059	1.520986
93	1	4.257179	-2.538841	2.014882
94	1	5.248507	-1.702198	0.807619
95	1	4.767397	-0.859045	2.270018
96	64	-0.000114	0.013810	0.092362

E(RTPSSh) = -2547.1848096 Hartree
Zero-point correction = 0.782524 Hartree/particle
Sum of electronic and thermal Energies = -2546.349186 Hartree
Sum of electronic and thermal Enthalpies = -2546.348242 Hartree
Sum of electronic and thermal Free Energies = -2546.492923 Hartree

Table S40. [Tb(DOTMA-(gly)$_4$(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.462196	2.108049	1.683115
2	6	1.411078	1.718875	2.768353
3	6	2.561342	0.826565	2.312074
4	7	2.099226	-0.454109	1.692589
5	6	1.702667	-1.404751	2.773040
6	6	0.813972	-2.552931	2.305090
7	7	-0.465617	-2.086623	1.686460
8	6	-1.415447	-1.695844	2.770642
9	6	-2.565171	-0.804576	2.311376
10	7	-2.101191	0.473735	1.689290
11	6	-1.706798	1.427606	2.768122
12	6	-0.817888	2.575516	2.299170
13	6	1.000196	3.191383	0.790339
14	6	1.972841	2.572192	-0.214457
15	8	1.749655	1.396544	-0.641535
16	6	3.193056	-0.989462	0.811274
17	6	2.590933	-1.954829	-0.208442
18	8	1.423563	-1.720653	-0.659975
19	6	-1.001852	-3.175186	0.798708
20	6	-1.980867	-2.567569	-0.205485
21	8	-1.757888	-1.394400	-0.645279
22	6	-3.190917	1.008016	0.802304
23	6	-2.578531	1.967426	-0.218215
24	8	-1.410977	1.730141	-0.660756
25	1	0.827482	1.201882	3.533222
26	1	1.831679	2.605071	3.258858

27	1	3.184592	1.346429	1.582652
28	1	3.194958	0.631081	3.187370
29	1	1.179793	-0.823009	3.535385
30	1	2.584705	-1.826950	3.269602
31	1	1.336361	-3.169672	1.571703
32	1	0.616674	-3.194227	3.174263
33	1	-0.832345	-1.176992	3.534590
34	1	-1.835711	-2.581320	3.262643
35	1	-3.188117	-1.326007	1.582760
36	1	-3.199784	-0.606668	3.185422
37	1	-1.184825	0.847929	3.532693
38	1	-2.590163	1.850101	3.262065
39	1	-1.339658	3.190359	1.563782
40	1	-0.621867	3.218696	3.167292
41	1	0.142852	3.475651	0.168926
42	1	3.484165	-0.129844	0.196385
43	1	-0.145461	-3.456141	0.174270
44	1	-3.484212	0.146584	0.191148
45	8	-0.020612	-0.173089	-2.354613
46	1	0.660823	-0.850714	-2.511842
47	1	-0.864841	-0.620747	-2.538343
48	7	-3.300590	2.976455	-0.661828
49	7	-2.989083	-3.294225	-0.640287
50	7	3.315600	-2.962505	-0.646444
51	7	2.979783	3.296872	-0.659156
52	1	-4.255861	3.206317	-0.343447
53	1	-3.207831	-4.254721	-0.327569
54	1	4.270317	-3.192584	-0.324017
55	1	3.198519	4.258425	-0.352300
56	6	3.919343	2.929258	-1.710631
57	1	3.386424	2.671859	-2.631512
58	6	-2.940002	3.902831	-1.727384
59	1	-2.698115	3.358770	-2.645912
60	6	-3.932323	-2.934507	-1.691469
61	1	-4.531614	-2.066799	-1.397119
62	6	2.960224	-3.893070	-1.710395
63	1	2.724701	-3.352496	-2.632683
64	1	-2.064321	4.497551	-1.447475
65	1	4.517665	2.061657	-1.413739
66	1	2.082390	-4.485335	-1.432445
67	1	-3.402415	-2.681058	-2.615196
68	6	4.196034	-4.825112	-1.920740
69	8	5.179058	-4.591536	-1.151773
70	8	4.078608	-5.691830	-2.815287
71	6	4.843834	4.167057	-1.940278
72	8	4.598123	5.165276	-1.195153
73	8	5.718745	4.036623	-2.825472
74	6	-4.174185	4.833787	-1.950147
75	8	-5.162578	4.605493	-1.186565
76	8	-4.050995	5.695900	-2.848778
77	6	-4.853541	-4.176967	-1.910248
78	8	-4.602787	-5.168050	-1.157154
79	8	-5.729920	-4.056329	-2.794941
80	6	-4.439642	1.535409	1.522383
81	1	-4.279274	2.505741	1.999289
82	1	-5.259535	1.636931	0.806141
83	1	-4.771132	0.824823	2.283565
84	6	-1.509360	-4.433148	1.516917
85	1	-1.615571	-5.248404	0.796144
86	1	-0.785425	-4.765242	2.264965
87	1	-2.473533	-4.282738	2.009161
88	6	4.441800	-1.509317	1.536715

89	1	4.283448	-2.477819	2.017931
90	1	5.263805	-1.611179	0.822999
91	1	4.768818	-0.793957	2.295236
92	6	1.516821	4.448513	1.503628
93	1	2.482453	4.294965	1.992107
94	1	1.623876	5.261724	0.780618
95	1	0.797485	4.785621	2.254021
96	66	-0.000247	0.013594	0.132652

E(RTPSSh) = -2547.7748475 Hartree
Zero-point correction = 0.782674 Hartree/particle
Sum of electronic and thermal Energies = -2546.939179 Hartree
Sum of electronic and thermal Enthalpies = -2546.938235 Hartree
Sum of electronic and thermal Free Energies = -2547.082011 Hartree

Table S41. [Dy(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.462196	2.108049	1.683115
2	6	1.411078	1.718875	2.768353
3	6	2.561342	0.826565	2.312074
4	7	2.099226	-0.454109	1.692589
5	6	1.702667	-1.404751	2.773040
6	6	0.813972	-2.552931	2.305090
7	7	-0.465617	-2.086623	1.686460
8	6	-1.415447	-1.695844	2.770642
9	6	-2.565171	-0.804576	2.311376
10	7	-2.101191	0.473735	1.689290
11	6	-1.706798	1.427606	2.768122
12	6	-0.817888	2.575516	2.299170
13	6	1.000196	3.191383	0.790339
14	6	1.972841	2.572192	-0.214457
15	8	1.749655	1.396544	-0.641535
16	6	3.193056	-0.989462	0.811274
17	6	2.590933	-1.954829	-0.208442
18	8	1.423563	-1.720653	-0.659975
19	6	-1.001852	-3.175186	0.798708
20	6	-1.980867	-2.567569	-0.205485
21	8	-1.757888	-1.394400	-0.645279
22	6	-3.190917	1.008016	0.802304
23	6	-2.578531	1.967426	-0.218215
24	8	-1.410977	1.730141	-0.660756
25	1	0.827482	1.201882	3.533222
26	1	1.831679	2.605071	3.258858
27	1	3.184592	1.346429	1.582652
28	1	3.194958	0.631081	3.187370
29	1	1.179793	-0.823009	3.535385
30	1	2.584705	-1.826950	3.269602
31	1	1.336361	-3.169672	1.571703
32	1	0.616674	-3.194227	3.174263
33	1	-0.832345	-1.176992	3.534590
34	1	-1.835711	-2.581320	3.262643
35	1	-3.188117	-1.326007	1.582760
36	1	-3.199784	-0.606668	3.185422
37	1	-1.184825	0.847929	3.532693
38	1	-2.590163	1.850101	3.262065
39	1	-1.339658	3.190359	1.563782
40	1	-0.621867	3.218696	3.167292

41	1	0.142852	3.475651	0.168926
42	1	3.484165	-0.129844	0.196385
43	1	-0.145461	-3.456141	0.174270
44	1	-3.484212	0.146584	0.191148
45	8	-0.020612	-0.173089	-2.354613
46	1	0.660823	-0.850714	-2.511842
47	1	-0.864841	-0.620747	-2.538343
48	7	-3.300590	2.976455	-0.661828
49	7	-2.989083	-3.294225	-0.640287
50	7	3.315600	-2.962505	-0.646444
51	7	2.979783	3.296872	-0.659156
52	1	-4.255861	3.206317	-0.343447
53	1	-3.207831	-4.254721	-0.327569
54	1	4.270317	-3.192584	-0.324017
55	1	3.198519	4.258425	-0.352300
56	6	3.919343	2.929258	-1.710631
57	1	3.386424	2.671859	-2.631512
58	6	-2.940002	3.902831	-1.727384
59	1	-2.698115	3.358770	-2.645912
60	6	-3.932323	-2.934507	-1.691469
61	1	-4.531614	-2.066799	-1.397119
62	6	2.960224	-3.893070	-1.710395
63	1	2.724701	-3.352496	-2.632683
64	1	-2.064321	4.497551	-1.447475
65	1	4.517665	2.061657	-1.413739
66	1	2.082390	-4.485335	-1.432445
67	1	-3.402415	-2.681058	-2.615196
68	6	4.196034	-4.825112	-1.920740
69	8	5.179058	-4.591536	-1.151773
70	8	4.078608	-5.691830	-2.815287
71	6	4.843834	4.167057	-1.940278
72	8	4.598123	5.165276	-1.195153
73	8	5.718745	4.036623	-2.825472
74	6	-4.174185	4.833787	-1.950147
75	8	-5.162578	4.605493	-1.186565
76	8	-4.050995	5.695900	-2.848778
77	6	-4.853541	-4.176967	-1.910248
78	8	-4.602787	-5.168050	-1.157154
79	8	-5.729920	-4.056329	-2.794941
80	6	-4.439642	1.535409	1.522383
81	1	-4.279274	2.505741	1.999289
82	1	-5.259535	1.636931	0.806141
83	1	-4.771132	0.824823	2.283565
84	6	-1.509360	-4.433148	1.516917
85	1	-1.615571	-5.248404	0.796144
86	1	-0.785425	-4.765242	2.264965
87	1	-2.473533	-4.282738	2.009161
88	6	4.441800	-1.509317	1.536715
89	1	4.283448	-2.477819	2.017931
90	1	5.263805	-1.611179	0.822999
91	1	4.768818	-0.793957	2.295236
92	6	1.516821	4.448513	1.503628
93	1	2.482453	4.294965	1.992107
94	1	1.623876	5.261724	0.780618
95	1	0.797485	4.785621	2.254021
96	66	-0.000247	0.013594	0.132652

E(RTPSSh) = -2548.3611382 Hartree
Zero-point correction = 0.782441 Hartree/particle
Sum of electronic and thermal Energies = -2547.525645 Hartree
Sum of electronic and thermal Enthalpies = -2547.524701 Hartree
Sum of electronic and thermal Free Energies = -2547.668697 Hartree

Table S42. [Ho(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.421790	2.110654	1.692702
2	6	1.375666	1.741808	2.779863
3	6	2.542771	0.871355	2.324214
4	7	2.102852	-0.416340	1.703914
5	6	1.726988	-1.374594	2.784137
6	6	0.859137	-2.537764	2.314139
7	7	-0.427226	-2.091688	1.695188
8	6	-1.383384	-1.721248	2.780102
9	6	-2.549330	-0.851802	2.319968
10	7	-2.106891	0.433752	1.697875
11	6	-1.734504	1.394857	2.777210
12	6	-0.865826	2.557905	2.307636
13	6	0.942275	3.201017	0.798006
14	6	1.920840	2.594522	-0.208255
15	8	1.717957	1.411996	-0.626381
16	6	3.204236	-0.931576	0.820116
17	6	2.615701	-1.901533	-0.202606
18	8	1.441026	-1.687463	-0.645523
19	6	-0.944726	-3.187273	0.805052
20	6	-1.928088	-2.592505	-0.201951
21	8	-1.724206	-1.412479	-0.632940
22	6	-3.203355	0.948269	0.807592
23	6	-2.603098	1.913550	-0.214023
24	8	-1.428344	1.695909	-0.647587
25	1	0.800717	1.216138	3.545226
26	1	1.778884	2.636876	3.268840
27	1	3.155888	1.403222	1.594711
28	1	3.180401	0.686766	3.198946
29	1	1.196397	-0.803551	3.549071
30	1	2.617971	-1.781508	3.277415
31	1	1.393028	-3.143279	1.579541
32	1	0.672112	-3.184703	3.181402
33	1	-0.809914	-1.193671	3.545229
34	1	-1.786965	-2.615524	3.270139
35	1	-3.161088	-1.385081	1.590297
36	1	-3.189236	-0.665010	3.192587
37	1	-1.205672	0.825606	3.544727
38	1	-2.627373	1.801817	3.267019
39	1	-1.398138	3.161807	1.570599
40	1	-0.681042	3.206382	3.174269
41	1	0.079711	3.472544	0.178235
42	1	3.481854	-0.065506	0.208124
43	1	-0.082400	-3.455663	0.183370
44	1	-3.482221	0.080820	0.198282
45	8	-0.015933	-0.160315	-2.329951
46	1	0.673897	-0.829773	-2.485785
47	1	-0.854432	-0.619176	-2.512466
48	7	-3.340897	2.906284	-0.668258
49	7	-2.919042	-3.335174	-0.648904
50	7	3.357376	-2.891089	-0.652979
51	7	2.910637	3.335551	-0.664270
52	1	-4.302555	3.119840	-0.357702
53	1	-3.121923	-4.301654	-0.343795
54	1	4.318389	-3.105033	-0.338025

55	1	3.111898	4.303617	-0.365929
56	6	3.849447	2.979813	-1.720532
57	1	3.314793	2.704100	-2.635072
58	6	-2.990896	3.832927	-1.737152
59	1	-2.732328	3.288055	-2.650632
60	6	-3.860600	-2.987173	-1.705591
61	1	-4.478553	-2.132677	-1.411256
62	6	3.014271	-3.819553	-1.722858
63	1	2.763461	-3.275923	-2.639321
64	1	-2.128787	4.446203	-1.455186
65	1	4.468474	2.127419	-1.422059
66	1	2.149527	-4.430992	-1.445451
67	1	-3.328229	-2.718455	-2.623579
68	6	4.266658	-4.725720	-1.947642
69	8	5.249291	-4.479434	-1.182173
70	8	4.160826	-5.587107	-2.848735
71	6	4.745735	4.234957	-1.966970
72	8	4.485397	5.233261	-1.226976
73	8	5.615508	4.116250	-2.858838
74	6	-4.241103	4.738633	-1.973960
75	8	-5.230144	4.495971	-1.215696
76	8	-4.128129	5.597417	-2.877096
77	6	-4.756418	-4.245467	-1.938838
78	8	-4.492687	-5.236169	-1.189710
79	8	-5.628011	-4.135934	-2.829649
80	6	-4.461606	1.458621	1.523418
81	1	-4.318174	2.433685	1.996052
82	1	-5.281959	1.543110	0.805515
83	1	-4.782449	0.746085	2.287389
84	6	-1.437026	-4.453003	1.520251
85	1	-1.526717	-5.269615	0.798795
86	1	-0.711978	-4.774096	2.272056
87	1	-2.405996	-4.317670	2.007408
88	6	4.461458	-1.435327	1.542331
89	1	4.319200	-2.408948	2.018213
90	1	5.284798	-1.519653	0.827886
91	1	4.776939	-0.718818	2.304691
92	6	1.442797	4.466072	1.508831
93	1	2.412963	4.327811	1.992863
94	1	1.533634	5.280785	0.785325
95	1	0.721669	4.791993	2.262485
96	67	-0.000406	0.013639	0.148922

E(RTPSSh) = -2548.9451695 Hartree
Zero-point correction = 0.781951 Hartree/particle
Sum of electronic and thermal Energies = -2548.110041 Hartree
Sum of electronic and thermal Enthalpies = -2548.109097 Hartree
Sum of electronic and thermal Free Energies = -2548.253789 Hartree

Table S43. [Tm(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.383432	2.101856	1.701972
2	6	1.340905	1.758366	2.791895
3	6	2.521585	0.909481	2.335410
4	7	2.093047	-0.381066	1.715770
5	6	1.738860	-1.341074	2.799144
6	6	0.895140	-2.518772	2.326594

7	7	-0.393215	-2.085368	1.705931
8	6	-1.354455	-1.738754	2.791705
9	6	-2.533386	-0.892087	2.327406
10	7	-2.101958	0.395973	1.705938
11	6	-1.752984	1.359987	2.787933
12	6	-0.907260	2.537061	2.316262
13	6	0.889317	3.194254	0.799270
14	6	1.874607	2.595128	-0.204811
15	8	1.691347	1.403646	-0.607851
16	6	3.197989	-0.884142	0.827773
17	6	2.618297	-1.855947	-0.198035
18	8	1.435918	-1.657305	-0.630602
19	6	-0.895144	-3.184302	0.809298
20	6	-1.880084	-2.598389	-0.200955
21	8	-1.689832	-1.411494	-0.622055
22	6	-3.200664	0.897165	0.809013
23	6	-2.606903	1.865896	-0.213010
24	8	-1.424507	1.662982	-0.634702
25	1	0.777047	1.224263	3.560191
26	1	1.725628	2.663244	3.278333
27	1	3.123976	1.453012	1.605271
28	1	3.164399	0.731653	3.207929
29	1	1.196550	-0.779924	3.563699
30	1	2.637870	-1.728279	3.294357
31	1	1.442852	-3.112362	1.592026
32	1	0.714350	-3.171780	3.190810
33	1	-0.793438	-1.201523	3.559839
34	1	-1.740121	-2.642097	3.280176
35	1	-3.132260	-1.437908	1.595923
36	1	-3.180926	-0.712044	3.195988
37	1	-1.213818	0.801365	3.556564
38	1	-2.654684	1.747846	3.277704
39	1	-1.452000	3.128862	1.578104
40	1	-0.729788	3.191842	3.179840
41	1	0.021684	3.451572	0.180302
42	1	3.467584	-0.012906	0.219346
43	1	-0.026388	-3.442040	0.191847
44	1	-3.469109	0.024505	0.202254
45	8	-0.012831	-0.142773	-2.312619
46	1	0.675638	-0.812481	-2.473194
47	1	-0.852216	-0.596949	-2.502269
48	7	-3.354966	2.844888	-0.676678
49	7	-2.855822	-3.352069	-0.658834
50	7	3.372268	-2.829041	-0.659791
51	7	2.848846	3.348441	-0.670778
52	1	-4.327709	3.054311	-0.383617
53	1	-3.056617	-4.328751	-0.369290
54	1	4.344839	-3.038464	-0.362182
55	1	3.043462	4.327841	-0.390570
56	6	3.794091	3.006056	-1.726173
57	1	3.265924	2.711339	-2.638476
58	6	-3.017624	3.775546	-1.746668
59	1	-2.742683	3.234799	-2.657765
60	6	-3.797702	-3.019277	-1.720693
61	1	-4.435864	-2.178949	-1.428634
62	6	3.045375	-3.758025	-1.734845
63	1	2.779337	-3.215620	-2.647751
64	1	-2.171356	4.409363	-1.461998
65	1	4.435031	2.171734	-1.422659
66	1	2.197135	-4.392727	-1.458671
67	1	-3.266582	-2.738757	-2.635881
68	6	4.326822	-4.628790	-1.961464

69	8	5.296083	-4.349828	-1.188350
70	8	4.248899	-5.487983	-2.864338
71	6	4.655916	4.289046	-1.974956
72	8	4.362522	5.274779	-1.228699
73	8	5.525154	4.196593	-2.867448
74	6	-4.294395	4.649503	-1.984689
75	8	-5.272209	4.374025	-1.221304
76	8	-4.205641	5.509213	-2.886650
77	6	-4.662117	-4.303975	-1.953362
78	8	-4.369907	-5.279732	-1.193408
79	8	-5.530296	-4.221879	-2.847272
80	6	-4.465475	1.399995	1.518594
81	1	-4.330710	2.378775	1.986454
82	1	-5.283051	1.476348	0.796565
83	1	-4.784847	0.688994	2.284808
84	6	-1.380106	-4.456675	1.517894
85	1	-1.457973	-5.271196	0.792625
86	1	-0.658250	-4.774347	2.274399
87	1	-2.354544	-4.331556	1.997269
88	6	4.459360	-1.382604	1.546563
89	1	4.323679	-2.360479	2.016001
90	1	5.281923	-1.458581	0.830248
91	1	4.771802	-0.668898	2.312992
92	6	1.377861	4.468285	1.502374
93	1	2.351902	4.342556	1.982448
94	1	1.458338	5.279508	0.773663
95	1	0.656690	4.791374	2.257406
96	69	-0.000706	0.014471	0.141891

E(RTPSSh) = -2550.0860564 Hartree
Zero-point correction = 0.782014 Hartree/particle
Sum of electronic and thermal Energies = -2549.251264 Hartree
Sum of electronic and thermal Enthalpies = -2549.250319 Hartree
Sum of electronic and thermal Free Energies = -2549.393187 Hartree

Table S44. [Yb(DOTMA-(gly)$_4$(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.310352	2.109248	1.709231
2	6	1.278930	1.802774	2.800005
3	6	2.487343	0.994346	2.342759
4	7	2.100283	-0.310045	1.724941
5	6	1.782151	-1.281163	2.808957
6	6	0.979196	-2.486204	2.334464
7	7	-0.322258	-2.093791	1.713986
8	6	-1.295594	-1.784049	2.799612
9	6	-2.501781	-0.978059	2.333089
10	7	-2.111106	0.323563	1.713029
11	6	-1.799536	1.299188	2.795462
12	6	-0.994047	2.503548	2.322206
13	6	0.780058	3.215189	0.803083
14	6	1.780592	2.644112	-0.201808
15	8	1.637481	1.444052	-0.595416
16	6	3.218931	-0.776907	0.834217
17	6	2.667165	-1.762579	-0.193419
18	8	1.476052	-1.602611	-0.618134
19	6	-0.786822	-3.206248	0.813730
20	6	-1.785830	-2.648890	-0.198454

21	8	-1.635344	-1.453168	-0.610683
22	6	-3.222828	0.788118	0.812145
23	6	-2.656342	1.772331	-0.210107
24	8	-1.464971	1.608494	-0.623481
25	1	0.734936	1.250608	3.569737
26	1	1.631688	2.721885	3.283811
27	1	3.069122	1.557528	1.610582
28	1	3.138029	0.838894	3.213676
29	1	1.221050	-0.740160	3.574344
30	1	2.694905	-1.636764	3.302555
31	1	1.547178	-3.059015	1.598545
32	1	0.819894	-3.147313	3.196727
33	1	-0.755372	-1.228336	3.569382
34	1	-1.649642	-2.701471	3.285628
35	1	-3.079393	-1.543768	1.599391
36	1	-3.157941	-0.820000	3.199437
37	1	-1.242477	0.761015	3.565804
38	1	-2.715545	1.655552	3.282439
39	1	-1.558320	3.074514	1.582091
40	1	-0.838749	3.166463	3.183814
41	1	-0.096334	3.444842	0.185499
42	1	3.461703	0.103381	0.227436
43	1	0.091298	-3.436168	0.198396
44	1	-3.462627	-0.093537	0.206264
45	8	-0.008396	-0.131958	-2.294471
46	1	0.697302	-0.783595	-2.454574
47	1	-0.835457	-0.608268	-2.483860
48	7	-3.434083	2.723385	-0.683025
49	7	-2.731891	-3.433058	-0.667239
50	7	3.451127	-2.706644	-0.665317
51	7	2.725806	3.427304	-0.678204
52	1	-4.415389	2.901058	-0.397435
53	1	-2.899827	-4.418327	-0.385735
54	1	4.432249	-2.884359	-0.374949
55	1	2.887424	4.415129	-0.406590
56	6	3.679017	3.109547	-1.734236
57	1	3.158733	2.790228	-2.642756
58	6	-3.121201	3.661576	-1.753974
59	1	-2.822888	3.127752	-2.661766
60	6	-3.680222	-3.126090	-1.731246
61	1	-4.348448	-2.310462	-1.436404
62	6	3.149648	-3.641459	-1.742864
63	1	2.860031	-3.104643	-2.651878
64	1	-2.298386	4.324157	-1.466053
65	1	4.349033	2.300026	-1.426394
66	1	2.325291	-4.306053	-1.464621
67	1	-3.155528	-2.821400	-2.642403
68	6	4.458569	-4.467321	-1.980546
69	8	5.422177	-4.159029	-1.211521
70	8	4.404191	-5.324844	-2.886664
71	6	4.496080	4.419205	-1.994961
72	8	4.170300	5.400430	-1.256269
73	8	5.366614	4.348735	-2.888142
74	6	-4.424700	4.492213	-2.002188
75	8	-5.397434	4.186919	-1.243686
76	8	-4.358501	5.351751	-2.906192
77	6	-4.498886	-4.438332	-1.975690
78	8	-4.175801	-5.408482	-1.221095
79	8	-5.366156	-4.379839	-2.872268
80	6	-4.503928	1.254710	1.517226
81	1	-4.399587	2.239208	1.980910
82	1	-5.322019	1.302961	0.793398

83	1	-4.802713	0.537490	2.285932
84	6	-1.236672	-4.493558	1.518472
85	1	-1.285198	-5.309551	0.792339
86	1	-0.510123	-4.789351	2.279334
87	1	-2.217530	-4.398052	1.991589
88	6	4.495176	-1.240776	1.549898
89	1	4.388376	-2.224430	2.014777
90	1	5.319092	-1.289169	0.832780
91	1	4.786669	-0.521464	2.319334
92	6	1.233076	4.504248	1.502533
93	1	2.213353	4.408158	1.976773
94	1	1.284544	5.316967	0.772911
95	1	0.506713	4.805540	2.261580
96	70	-0.001338	0.014745	0.154291

E(RTPSSh) = -2550.6693165 Hartree
Zero-point correction = 0.782274 Hartree/particle
Sum of electronic and thermal Energies = -2549.834377 Hartree
Sum of electronic and thermal Enthalpies = -2549.833433 Hartree
Sum of electronic and thermal Free Energies = -2549.975698 Hartree

Table S45. [Lu(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.265920	2.111109	1.710223
2	6	1.241176	1.828352	2.801408
3	6	2.465826	1.045614	2.343312
4	7	2.103239	-0.266204	1.726348
5	6	1.808622	-1.243671	2.810976
6	6	1.031694	-2.464821	2.335030
7	7	-0.277067	-2.096775	1.714947
8	6	-1.256672	-1.811011	2.801296
9	6	-2.479325	-1.031184	2.334020
10	7	-2.113628	0.278055	1.714765
11	6	-1.825277	1.259956	2.797749
12	6	-1.045782	2.480623	2.323229
13	6	0.713315	3.224428	0.801581
14	6	1.723204	2.669767	-0.202829
15	8	1.604319	1.464964	-0.589882
16	6	3.228692	-0.711387	0.833043
17	6	2.692119	-1.705210	-0.194533
18	8	1.496437	-1.567057	-0.614260
19	6	-0.719483	-3.216141	0.812056
20	6	-1.726578	-2.674788	-0.200489
21	8	-1.598328	-1.474633	-0.607620
22	6	-3.233009	0.720600	0.812240
23	6	-2.682898	1.713823	-0.209835
24	8	-1.486783	1.573098	-0.617841
25	1	0.710515	1.265155	3.572420
26	1	1.573488	2.756011	3.283259
27	1	3.035004	1.620547	1.610196
28	1	3.120936	0.903217	3.213098
29	1	1.236022	-0.716145	3.577211
30	1	2.729776	-1.578832	3.303182
31	1	1.611748	-3.024466	1.598202
32	1	0.885400	-3.130820	3.195790
33	1	-0.729824	-1.243857	3.571944
34	1	-1.589837	-2.736965	3.285805

35	1	-3.043994	-1.608841	1.599344
36	1	-3.140052	-0.886601	3.199204
37	1	-1.256676	0.735113	3.568817
38	1	-2.749538	1.595850	3.283592
39	1	-1.622155	3.038893	1.582590
40	1	-0.903316	3.147917	3.183625
41	1	-0.167768	3.436537	0.184510
42	1	3.454697	0.173662	0.226835
43	1	0.163430	-3.428996	0.197544
44	1	-3.455892	-0.165738	0.206855
45	8	-0.006084	-0.117865	-2.283816
46	1	0.712302	-0.755095	-2.445189
47	1	-0.823863	-0.610999	-2.470543
48	7	-3.477443	2.648281	-0.687778
49	7	-2.656397	-3.474988	-0.674389
50	7	3.491615	-2.633710	-0.671087
51	7	2.651405	3.469631	-0.684847
52	1	-4.462813	2.808035	-0.405421
53	1	-2.807406	-4.463880	-0.395662
54	1	4.476582	-2.794716	-0.383726
55	1	2.794549	4.461454	-0.417304
56	6	3.612228	3.164978	-1.737866
57	1	3.099583	2.835580	-2.647163
58	6	-3.178258	3.593079	-1.756860
59	1	-2.870494	3.065687	-2.665284
60	6	-3.611141	-3.181413	-1.736481
61	1	-4.290919	-2.376085	-1.439809
62	6	3.202814	-3.574817	-1.746675
63	1	2.903658	-3.044198	-2.656255
64	1	-2.366495	4.268264	-1.466909
65	1	4.294310	2.366953	-1.426590
66	1	2.389281	-4.251613	-1.466031
67	1	-3.092867	-2.868467	-2.648540
68	6	4.523958	-4.380805	-1.985109
69	8	5.484612	-4.054601	-1.219790
70	8	4.480497	-5.242074	-2.888206
71	6	4.408799	4.487037	-1.999487
72	8	4.063192	5.465366	-1.265999
73	8	5.284574	4.427557	-2.888293
74	6	-4.494591	4.403152	-2.005423
75	8	-5.463747	4.079866	-1.249803
76	8	-4.440352	5.266065	-2.906962
77	6	-4.410491	-4.505633	-1.980410
78	8	-4.069717	-5.472021	-1.228776
79	8	-5.281672	-4.458652	-2.873805
80	6	-4.522937	1.165393	1.515090
81	1	-4.436736	2.152956	1.975960
82	1	-5.341106	1.196810	0.790435
83	1	-4.809130	0.444918	2.285530
84	6	-1.148034	-4.512271	1.513612
85	1	-1.179630	-5.327965	0.786216
86	1	-0.418644	-4.795816	2.276413
87	1	-2.131988	-4.434904	1.983575
88	6	4.513922	-1.154456	1.545583
89	1	4.424937	-2.141137	2.007805
90	1	5.337428	-1.186499	0.827079
91	1	4.793722	-0.432143	2.316545
92	6	1.144127	4.522528	1.498060
93	1	2.127548	4.445087	1.969161
94	1	1.178269	5.335056	0.767218
95	1	0.414610	4.811140	2.258995
96	71	-0.001153	0.015517	0.159928

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 E(RTPSSh) = -2551.223034 Hartree
 Zero-point correction = 0.782361 Hartree/particle
 Sum of electronic and thermal Energies = -2550.388051 Hartree
 Sum of electronic and thermal Enthalpies = -2550.387107 Hartree
 Sum of electronic and thermal Free Energies = -2550.529304 Hartree
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Table S46. [La(DOTMA-(gly)$_4$(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.103298	-1.857872	1.884380
2	6	-0.127138	-2.196801	2.956747
3	6	1.313925	-2.340860	2.473001
4	7	1.873925	-1.085634	1.886295
5	6	2.197301	-0.113317	2.967407
6	6	2.345072	1.330112	2.491723
7	7	1.096238	1.887350	1.889247
8	6	0.120365	2.228950	2.960362
9	6	-1.320790	2.368936	2.475952
10	7	-1.874430	1.112895	1.884955
11	6	-2.207476	0.142304	2.963357
12	6	-2.353231	-1.300742	2.486272
13	6	-1.431119	-3.044119	1.021245
14	6	-2.223798	-2.517906	-0.186976
15	8	-1.951239	-1.363779	-0.650500
16	6	3.073631	-1.402740	1.038395
17	6	2.572778	-2.167813	-0.197150
18	8	1.425283	-1.890148	-0.676016
19	6	1.422990	3.067994	1.019597
20	6	2.207222	2.531363	-0.190916
21	8	1.945952	1.368197	-0.633479
22	6	-3.061105	1.428513	1.019263
23	6	-2.535429	2.199062	-0.204362
24	8	-1.376593	1.931760	-0.656078
25	1	-0.188900	-1.409783	3.711264
26	1	-0.406257	-3.123092	3.473752
27	1	1.375516	-3.123082	1.711233
28	1	1.920137	-2.677204	3.324562
29	1	1.400661	-0.177714	3.711515
30	1	3.117134	-0.393767	3.495327
31	1	3.139169	1.397220	1.743289
32	1	2.666305	1.933915	3.350875
33	1	0.182617	1.444402	3.717587
34	1	0.398149	3.157635	3.474249
35	1	-1.384627	3.153471	1.717006
36	1	-1.929525	2.699998	3.327864
37	1	-1.416556	0.206533	3.713718
38	1	-3.131528	0.422989	3.484083
39	1	-3.146146	-1.367481	1.736396
40	1	-2.675228	-1.905921	3.344037
41	1	-0.476275	-3.390103	0.604896
42	1	3.429408	-0.441092	0.647715
43	1	0.466692	3.415572	0.608208
44	1	-3.409561	0.467254	0.620717
45	8	-0.031286	-0.379519	-2.463817
46	1	0.610195	-1.110188	-2.533969
47	1	-0.897496	-0.787741	-2.636402
48	7	-3.313610	3.095973	-0.770500

49	7	3.110524	3.304824	-0.753967
50	7	3.356049	-3.067033	-0.749836
51	7	-3.141667	-3.287146	-0.729795
52	1	-4.254849	3.381480	-0.441414
53	1	3.391432	4.249121	-0.430390
54	1	4.287613	-3.363815	-0.401419
55	1	-3.430604	-4.225141	-0.392758
56	6	-3.945134	-2.989089	-1.908794
57	1	-3.307009	-2.832734	-2.784707
58	6	-3.019453	3.877707	-1.964879
59	1	-2.853563	3.222814	-2.826419
60	6	3.907729	2.998618	-1.934973
61	1	3.264247	2.824174	-2.803395
62	6	3.081215	-3.844003	-1.952089
63	1	2.947820	-3.186660	-2.817559
64	1	-4.537303	-2.081214	-1.755390
65	1	-2.117257	4.480981	-1.821124
66	1	4.508928	2.097878	-1.774203
67	1	2.167485	-4.434696	-1.831581
68	6	-4.263514	4.796952	-2.202835
69	8	-5.195296	4.654209	-1.350377
70	8	-4.198957	5.558088	-3.191371
71	6	4.830562	4.239811	-2.173786
72	8	4.677947	5.180170	-1.332608
73	8	5.603916	4.165063	-3.152156
74	6	4.319508	-4.779316	-2.158319
75	8	4.269735	-5.540797	-3.147034
76	8	5.231274	-4.645819	-1.283048
77	6	-4.879675	-4.226420	-2.123132
78	8	-4.727352	-5.155391	-1.269269
79	8	-5.659474	-4.159471	-3.096513
80	6	2.110703	4.243645	1.728637
81	1	3.125364	3.994048	2.049763
82	1	2.159987	5.103861	1.055568
83	1	1.539070	4.566354	2.601366
84	6	-4.233437	2.121886	1.728372
85	1	-3.985613	3.142231	2.032356
86	1	-5.099476	2.157711	1.061928
87	1	-4.546203	1.561930	2.612227
88	6	-2.109494	-4.219891	1.738711
89	1	-3.120858	-3.971155	2.070626
90	1	-2.164903	-5.080197	1.066230
91	1	-1.528352	-4.542075	2.605168
92	6	4.233456	-2.100149	1.764311
93	1	3.980068	-3.121587	2.059906
94	1	5.110097	-2.133751	1.111762
95	1	4.532628	-1.544473	2.655428
96	57	-0.000071	0.014789	0.146876

E(RTPSSh) = -2542.844013 Hartree
Zero-point correction = 0.780582 Hartree/particle
Sum of electronic and thermal Energies = -2542.009901 Hartree
Sum of electronic and thermal Enthalpies = -2542.008957 Hartree
Sum of electronic and thermal Free Energies = -2542.155485 Hartree

Table S47. [Ce(DOTMA-(gly)$_4$(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z

1	7	-1.080883	-1.864952	1.891583
2	6	-0.104898	-2.200902	2.964673
3	6	1.337429	-2.328916	2.481620
4	7	1.879676	-1.066191	1.894653
5	6	2.200043	-0.094470	2.976700
6	6	2.331640	1.350432	2.501990
7	7	1.075563	1.890244	1.898597
8	6	0.099650	2.228045	2.970567
9	6	-1.342801	2.352454	2.487065
10	7	-1.879281	1.089685	1.894674
11	6	-2.208247	0.117886	2.972835
12	6	-2.338140	-1.325893	2.494501
13	6	-1.392810	-3.047303	1.017645
14	6	-2.187292	-2.517325	-0.187179
15	8	-1.928254	-1.353055	-0.633936
16	6	3.075194	-1.367252	1.035648
17	6	2.569084	-2.134502	-0.195883
18	8	1.410275	-1.872178	-0.656645
19	6	1.387171	3.067498	1.019105
20	6	2.174167	2.527120	-0.187552
21	8	1.928744	1.353695	-0.612324
22	6	-3.063261	1.390943	1.020619
23	6	-2.534467	2.163657	-0.199722
24	8	-1.366383	1.910451	-0.635736
25	1	-0.175057	-1.417492	3.722275
26	1	-0.377368	-3.131893	3.476839
27	1	1.408204	-3.110709	1.720296
28	1	1.948307	-2.657238	3.332950
29	1	1.407104	-0.167716	3.724029
30	1	3.124792	-0.368160	3.499658
31	1	3.125765	1.427385	1.754622
32	1	2.644087	1.958549	3.361304
33	1	0.170330	1.446530	3.730266
34	1	0.370697	3.161041	3.480280
35	1	-1.416056	3.137272	1.729319
36	1	-1.955870	2.674685	3.339216
37	1	-1.420391	0.189770	3.725788
38	1	-3.136739	0.390972	3.489754
39	1	-3.130678	-1.400993	1.745075
40	1	-2.652179	-1.936509	3.351342
41	1	-0.433094	-3.378337	0.600357
42	1	3.415741	-0.400758	0.643355
43	1	0.426268	3.400746	0.606637
44	1	-3.398627	0.425402	0.621341
45	8	-0.032388	-0.327117	-2.420345
46	1	0.613882	-1.051284	-2.509553
47	1	-0.897283	-0.740809	-2.587601
48	7	-3.317952	3.047272	-0.779026
49	7	3.062916	3.306261	-0.765312
50	7	3.357649	-3.019298	-0.763708
51	7	-3.091926	-3.291556	-0.744225
52	1	-4.266381	3.321934	-0.461276
53	1	3.331947	4.258470	-0.454645
54	1	4.297525	-3.304950	-0.428165
55	1	-3.370783	-4.237077	-0.419247
56	6	-3.896756	-2.989145	-1.921302
57	1	-3.259090	-2.817240	-2.794650
58	6	-3.020050	3.830136	-1.971789
59	1	-2.838062	3.175631	-2.830385
60	6	3.861486	2.995957	-1.944400
61	1	3.218489	2.803051	-2.809280
62	6	3.076979	-3.798037	-1.963534

63	1	2.927477	-3.141317	-2.826881
64	1	-4.498344	-2.089073	-1.759085
65	1	-2.126153	4.443623	-1.819808
66	1	4.474397	2.104937	-1.774125
67	1	2.170569	-4.397790	-1.833274
68	6	-4.271879	4.734954	-2.224243
69	8	-5.211073	4.582498	-1.381648
70	8	-4.205438	5.495584	-3.212999
71	6	4.767831	4.246050	-2.199888
72	8	4.603669	5.194753	-1.370327
73	8	5.541117	4.169179	-3.178082
74	6	4.322506	-4.720357	-2.184668
75	8	4.268607	-5.482836	-3.172320
76	8	5.243612	-4.576424	-1.320894
77	6	-4.817972	-4.234053	-2.149831
78	8	-4.655517	-5.170823	-1.306398
79	8	-5.598204	-4.164669	-3.122585
80	6	2.064063	4.256092	1.716428
81	1	3.082626	4.020678	2.035884
82	1	2.101324	5.111521	1.036518
83	1	1.491685	4.578970	2.588619
84	6	-4.247321	2.074283	1.719527
85	1	-4.012686	3.098398	2.021275
86	1	-5.109356	2.098347	1.047378
87	1	-4.559583	1.514111	2.603445
88	6	-2.060395	-4.236672	1.722261
89	1	-3.075808	-4.002624	2.052498
90	1	-2.103490	-5.091745	1.042241
91	1	-1.478651	-4.559448	2.588097
92	6	4.249212	-2.053720	1.748615
93	1	4.010417	-3.079034	2.042977
94	1	5.120358	-2.075040	1.088229
95	1	4.549667	-1.497455	2.638951
96	58	0.000838	0.015141	0.176751

E(RTPSSh) = -2543.4959883 Hartree
Zero-point correction = 0.780687 Hartree/particle
Sum of electronic and thermal Energies = -2542.661835 Hartree
Sum of electronic and thermal Enthalpies = -2542.660891 Hartree
Sum of electronic and thermal Free Energies = -2542.807008 Hartree

Table S48. [Pr(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.071933	-1.870342	1.902771
2	6	-0.092654	-2.204791	2.973695
3	6	1.350998	-2.323848	2.490341
4	7	1.890567	-1.060348	1.900694
5	6	2.214077	-0.085861	2.979413
6	6	2.335355	1.359952	2.503058
7	7	1.075975	1.898238	1.903195
8	6	0.100425	2.235711	2.976043
9	6	-1.344617	2.351441	2.496288
10	7	-1.881520	1.087111	1.905679
11	6	-2.208416	0.113673	2.983373
12	6	-2.330113	-1.331839	2.506155
13	6	-1.382560	-3.047935	1.024777
14	6	-2.168911	-2.509045	-0.180434

15	8	-1.908713	-1.341588	-0.616611
16	6	3.076902	-1.360677	1.031302
17	6	2.552837	-2.124542	-0.193874
18	8	1.393014	-1.852549	-0.644124
19	6	1.383648	3.070447	1.018060
20	6	2.157727	2.520343	-0.191399
21	8	1.901280	1.348065	-0.611294
22	6	-3.061743	1.386914	1.028475
23	6	-2.524316	2.150515	-0.192683
24	8	-1.357299	1.888439	-0.624486
25	1	-0.167038	-1.423442	3.732556
26	1	-0.359202	-3.139879	3.481231
27	1	1.426146	-3.107526	1.731589
28	1	1.962679	-2.647986	3.342563
29	1	1.425910	-0.163734	3.730906
30	1	3.144897	-0.353970	3.494167
31	1	3.127026	1.439858	1.753474
32	1	2.649062	1.969349	3.360818
33	1	0.177556	1.456890	3.737393
34	1	0.367628	3.173072	3.479402
35	1	-1.423586	3.136124	1.739120
36	1	-1.955800	2.672231	3.350157
37	1	-1.422078	0.191185	3.736886
38	1	-3.140651	0.381843	3.495794
39	1	-3.123113	-1.410720	1.757793
40	1	-2.642177	-1.942112	3.363809
41	1	-0.422255	-3.379119	0.609579
42	1	3.413896	-0.394595	0.635462
43	1	0.421005	3.404310	0.610863
44	1	-3.398130	0.420841	0.631981
45	8	-0.049982	-0.272696	-2.373640
46	1	0.605229	-0.984914	-2.487603
47	1	-0.912067	-0.702321	-2.515445
48	59	0.001450	0.016342	0.217832
49	7	-3.297856	3.038795	-0.780736
50	7	3.050029	3.288887	-0.780194
51	7	3.324828	-3.020721	-0.769204
52	7	-3.071372	-3.277153	-0.751682
53	1	-4.238610	3.316932	-0.457958
54	1	3.325201	4.232623	-0.463784
55	1	4.259198	-3.307719	-0.434048
56	1	-3.348035	-4.217644	-0.425042
57	6	-3.867310	-2.951958	-1.928477
58	1	-3.222763	-2.778927	-2.796408
59	6	-2.977099	3.811202	-1.974104
60	1	-2.800206	3.148885	-2.827756
61	6	3.835449	2.955899	-1.961638
62	1	3.182784	2.771212	-2.821042
63	6	3.010691	-3.796925	-1.962063
64	1	2.858946	-3.138275	-2.823419
65	1	-4.455215	-2.044376	-1.758178
66	1	-2.072152	4.406931	-1.816951
67	1	4.428829	2.052224	-1.788852
68	1	2.093966	-4.376709	-1.814641
69	6	-4.200240	4.741478	-2.249720
70	8	-5.156365	4.629278	-1.421697
71	8	-4.102088	5.489275	-3.248438
72	6	4.769877	4.175660	-2.238700
73	8	4.650321	5.139231	-1.420422
74	8	5.527942	4.067843	-3.228650
75	6	4.224387	-4.747430	-2.210879
76	8	4.128512	-5.505576	-3.201637

77	8	5.171218	-4.637169	-1.372077
78	6	-4.808373	-4.171714	-2.183798
79	8	-4.681431	-5.127725	-1.357823
80	8	-5.576907	-4.070833	-3.165994
81	6	2.067979	4.261176	1.704232
82	1	3.090425	4.028355	2.012816
83	1	2.094959	5.115856	1.022927
84	1	1.504786	4.583016	2.582599
85	6	-4.247066	2.075686	1.719634
86	1	-4.014000	3.102107	2.014445
87	1	-5.108727	2.092933	1.046826
88	1	-4.558243	1.521501	2.607544
89	6	-2.055213	-4.239278	1.721169
90	1	-3.073838	-4.007956	2.043165
91	1	-2.089559	-5.094457	1.040841
92	1	-1.480040	-4.559865	2.592063
93	6	4.258545	-2.047448	1.730687
94	1	4.024156	-3.072983	2.027602
95	1	5.123495	-2.066505	1.062155
96	1	4.565517	-1.490781	2.618400

E(RTPSSh) = -2544.1537075 Hartree
Zero-point correction = 0.781716 Hartree/particle
Sum of electronic and thermal Energies = -2543.318716 Hartree
Sum of electronic and thermal Enthalpies = -2543.317772 Hartree
Sum of electronic and thermal Free Energies = -2543.462370 Hartree

Table S49. [Nd(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.057838	-1.873705	1.910346
2	6	-0.079345	-2.208310	2.981428
3	6	1.364594	-2.317502	2.497996
4	7	1.892581	-1.049094	1.908921
5	6	2.214572	-0.076204	2.988941
6	6	2.326660	1.370285	2.513624
7	7	1.063041	1.897868	1.913355
8	6	0.088333	2.234531	2.986771
9	6	-1.357029	2.340641	2.507045
10	7	-1.882393	1.072083	1.915466
11	6	-2.207535	0.098659	2.993161
12	6	-2.320196	-1.346855	2.514903
13	6	-1.359438	-3.048023	1.025248
14	6	-2.143887	-2.504875	-0.178615
15	8	-1.890883	-1.330865	-0.602013
16	6	3.076337	-1.339137	1.033123
17	6	2.549026	-2.099870	-0.191972
18	8	1.381842	-1.836681	-0.629073
19	6	1.362159	3.067830	1.022565
20	6	2.134407	2.514451	-0.185985
21	8	1.886670	1.335670	-0.593206
22	6	-3.060602	1.362974	1.032924
23	6	-2.520460	2.124962	-0.187415
24	8	-1.347178	1.871540	-0.607657
25	1	-0.158659	-1.430185	3.743126
26	1	-0.342236	-3.146784	3.484619
27	1	1.445217	-3.100449	1.739070
28	1	1.979563	-2.637192	3.349528

29	1	1.428205	-0.159624	3.741776
30	1	3.147998	-0.340933	3.500770
31	1	3.118354	1.456175	1.764734
32	1	2.635101	1.982251	3.371428
33	1	0.170644	1.458426	3.750387
34	1	0.351714	3.174969	3.486383
35	1	-1.441539	3.125339	1.750491
36	1	-1.971490	2.655989	3.360567
37	1	-1.422930	0.180592	3.748049
38	1	-3.142263	0.362775	3.503159
39	1	-3.113327	-1.430569	1.767202
40	1	-2.627083	-1.960822	3.371780
41	1	-0.396237	-3.371963	0.611292
42	1	3.406015	-0.369709	0.639582
43	1	0.396762	3.395439	0.617093
44	1	-3.390342	0.394142	0.637914
45	8	-0.045372	-0.244835	-2.339299
46	1	0.608370	-0.958199	-2.455045
47	1	-0.908094	-0.672775	-2.482604
48	7	-3.297125	3.002899	-0.786555
49	7	3.016235	3.285645	-0.786677
50	7	3.324559	-2.983955	-0.780702
51	7	-3.037324	-3.274544	-0.761413
52	1	-4.243535	3.272996	-0.473369
53	1	3.283485	4.235184	-0.480696
54	1	4.264449	-3.264329	-0.455136
55	1	-3.307830	-4.220290	-0.444519
56	6	-3.830963	-2.945814	-1.938796
57	1	-3.184395	-2.760103	-2.802564
58	6	-2.975277	3.771103	-1.982341
59	1	-2.783043	3.105304	-2.829916
60	6	3.797692	2.950670	-1.970205
61	1	3.141627	2.750969	-2.823622
62	6	3.008579	-3.755357	-1.976197
63	1	2.845386	-3.092708	-2.832395
64	1	-4.427290	-2.044782	-1.763182
65	1	-2.079103	4.378978	-1.821485
66	1	4.402477	2.055430	-1.793184
67	1	2.098005	-4.343996	-1.826045
68	6	-4.207246	4.684613	-2.274497
69	8	-5.167985	4.567858	-1.452444
70	8	-4.110649	5.425236	-3.278671
71	6	4.716420	4.178471	-2.264062
72	8	4.591721	5.147403	-1.452893
73	8	5.468915	4.070733	-3.258229
74	6	4.228975	-4.693479	-2.239332
75	8	4.132921	-5.446490	-3.233939
76	8	5.180704	-4.579503	-1.406558
77	6	-4.760570	-4.171433	-2.208134
78	8	-4.629105	-5.132827	-1.389118
79	8	-5.525715	-4.069150	-3.192777
80	6	2.043899	4.264457	1.700897
81	1	3.069358	4.038279	2.004298
82	1	2.062290	5.117000	1.016633
83	1	1.483894	4.586017	2.581412
84	6	-4.252105	2.048372	1.716647
85	1	-4.026205	3.077768	2.006541
86	1	-5.111733	2.056781	1.041069
87	1	-4.562620	1.497047	2.606586
88	6	-2.029098	-4.246424	1.712222
89	1	-3.050985	-4.022721	2.029182
90	1	-2.054304	-5.098676	1.027826

91	1	-1.457087	-4.567401	2.585059
92	6	4.264967	-2.022977	1.723335
93	1	4.038565	-3.052112	2.013846
94	1	5.127583	-2.031562	1.051578
95	1	4.571272	-1.470371	2.613827
96	60	0.002159	0.016846	0.240085

E(RTPSSh) = -2544.7764488 Hartree
Zero-point correction = 0.781819 Hartree/particle
Sum of electronic and thermal Energies = -2543.941424 Hartree
Sum of electronic and thermal Enthalpies = -2543.940480 Hartree
Sum of electronic and thermal Free Energies = -2544.084912 Hartree

Table S50. [Sm(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.030977	-1.881051	1.921819
2	6	-0.054085	-2.217618	2.992962
3	6	1.389959	-2.308247	2.509087
4	7	1.896781	-1.030603	1.922224
5	6	2.216497	-0.061711	3.005631
6	6	2.311705	1.386150	2.533334
7	7	1.040565	1.895003	1.932940
8	6	0.067513	2.229650	3.007834
9	6	-1.378053	2.318417	2.527926
10	7	-1.882236	1.042805	1.933575
11	6	-2.204310	0.068664	3.010737
12	6	-2.300625	-1.376005	2.529013
13	6	-1.317806	-3.049325	1.024627
14	6	-2.097779	-2.497565	-0.176877
15	8	-1.855327	-1.312943	-0.577537
16	6	3.076525	-1.302986	1.036549
17	6	2.543558	-2.055551	-0.189900
18	8	1.363965	-1.807069	-0.603213
19	6	1.325404	3.062658	1.034893
20	6	2.092642	2.505256	-0.173795
21	8	1.859945	1.315932	-0.559355
22	6	-3.057965	1.319618	1.043894
23	6	-2.513602	2.078644	-0.175075
24	8	-1.330183	1.839363	-0.575274
25	1	-0.141969	-1.446889	3.761270
26	1	-0.310428	-3.162974	3.486577
27	1	1.480564	-3.088724	1.748737
28	1	2.011175	-2.620756	3.358710
29	1	1.434056	-0.155789	3.761423
30	1	3.154917	-0.320773	3.511278
31	1	3.102844	1.483478	1.785250
32	1	2.610991	2.002141	3.391415
33	1	0.158920	1.458576	3.775589
34	1	0.323983	3.175451	3.500843
35	1	-1.472678	3.103355	1.772774
36	1	-1.998742	2.623321	3.380688
37	1	-1.423223	0.157589	3.768557
38	1	-3.143569	0.325188	3.516301
39	1	-3.093594	-1.467818	1.782027
40	1	-2.598289	-1.997755	3.383506
41	1	-0.349965	-3.362252	0.613795
42	1	3.396073	-0.327881	0.649609

43	1	0.355386	3.381778	0.634482
44	1	-3.378139	0.346716	0.651914
45	8	-0.033095	-0.192930	-2.276261
46	1	0.617131	-0.909175	-2.394634
47	1	-0.897256	-0.616047	-2.425202
48	7	-3.294596	2.939190	-0.793010
49	7	2.954769	3.281260	-0.795974
50	7	3.324470	-2.917065	-0.803714
51	7	-2.976739	-3.267674	-0.779974
52	1	-4.250903	3.194274	-0.497261
53	1	3.206899	4.240978	-0.508938
54	1	4.273707	-3.186342	-0.495502
55	1	-3.237897	-4.221999	-0.480575
56	6	-3.765912	-2.930977	-1.958134
57	1	-3.115762	-2.724508	-2.814431
58	6	-2.971226	3.698550	-1.994022
59	1	-2.748768	3.026183	-2.828816
60	6	3.726296	2.944180	-1.985428
61	1	3.062625	2.715451	-2.825505
62	6	3.005204	-3.677907	-2.005127
63	1	2.822269	-3.007151	-2.850991
64	1	-4.374908	-2.040350	-1.773399
65	1	-2.093060	4.330604	-1.827106
66	1	4.354263	2.066192	-1.802911
67	1	2.105531	-4.282155	-1.851752
68	6	-4.220012	4.577714	-2.318679
69	8	-5.189366	4.451737	-1.508113
70	8	-4.126237	5.304250	-3.333315
71	6	4.612131	4.187688	-2.312631
72	8	4.477187	5.166429	-1.514923
73	8	5.351956	4.081137	-3.316342
74	6	4.236824	-4.593575	-2.294585
75	8	4.140656	-5.335489	-3.297394
76	8	5.196405	-4.475055	-1.471427
77	6	-4.677695	-4.164514	-2.252022
78	8	-4.538861	-5.136096	-1.446269
79	8	-5.437509	-4.057776	-3.240218
80	6	2.005142	4.266939	1.701496
81	1	3.035742	4.050063	1.993859
82	1	2.009007	5.117206	1.014180
83	1	1.452355	4.586881	2.587188
84	6	-4.258239	2.000509	1.716545
85	1	-4.042975	3.034549	1.997688
86	1	-5.115131	1.994932	1.037476
87	1	-4.567153	1.454387	2.610303
88	6	-1.984371	-4.258722	1.694963
89	1	-3.011493	-4.046516	2.002657
90	1	-1.994849	-5.105688	1.003652
91	1	-1.418715	-4.581351	2.571352
92	6	4.274998	-1.985351	1.710982
93	1	4.060219	-3.020883	1.987042
94	1	5.134490	-1.975406	1.035263
95	1	4.579658	-1.442791	2.608232
96	62	0.004513	0.017584	0.277744

E(RTPSSh) = -2545.9989206 Hartree
Zero-point correction = 0.781851 Hartree/particle
Sum of electronic and thermal Energies = -2545.163921 Hartree
Sum of electronic and thermal Enthalpies = -2545.162977 Hartree
Sum of electronic and thermal Free Energies = -2545.307690 Hartree

Table S51. [Eu(DOTMA-(gly)$_4$(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.007411	-1.895922	1.929433
2	6	-0.029610	-2.233553	2.999084
3	6	1.414539	-2.307948	2.514347
4	7	1.906367	-1.021965	1.933855
5	6	2.220517	-0.057483	3.022379
6	6	2.302706	1.392879	2.556392
7	7	1.026297	1.890902	1.958318
8	6	0.052733	2.217222	3.035132
9	6	-1.393207	2.292977	2.555322
10	7	-1.882095	1.014240	1.955385
11	6	-2.198580	0.034320	3.028607
12	6	-2.282120	-1.408216	2.539421
13	6	-1.282926	-3.059736	1.023342
14	6	-2.059417	-2.503656	-0.177791
15	8	-1.828358	-1.311012	-0.561676
16	6	3.085926	-1.277947	1.043705
17	6	2.554545	-2.020130	-0.189231
18	8	1.367425	-1.784250	-0.588753
19	6	1.300054	3.062432	1.062291
20	6	2.060713	2.511938	-0.153185
21	8	1.845973	1.315649	-0.527626
22	6	-3.058404	1.282988	1.064452
23	6	-2.516117	2.042806	-0.154335
24	8	-1.325136	1.820888	-0.541958
25	1	-0.124305	-1.469800	3.773511
26	1	-0.278938	-3.185011	3.484493
27	1	1.512628	-3.083446	1.749870
28	1	2.040100	-2.618189	3.361585
29	1	1.439508	-0.162044	3.778352
30	1	3.161768	-0.312077	3.525095
31	1	3.092567	1.500736	1.808440
32	1	2.596025	2.008780	3.416543
33	1	0.151790	1.447026	3.802865
34	1	0.301769	3.165349	3.527482
35	1	-1.495825	3.079878	1.803252
36	1	-2.018081	2.587531	3.408665
37	1	-1.419130	0.126209	3.787788
38	1	-3.140634	0.281864	3.533476
39	1	-3.074230	-1.503665	1.791949
40	1	-2.573507	-2.038129	3.390094
41	1	-0.311458	-3.364897	0.615682
42	1	3.399392	-0.297151	0.666824
43	1	0.326064	3.379321	0.670183
44	1	-3.373283	0.308034	0.673732
45	8	-0.026260	-0.159074	-2.240518
46	1	0.626408	-0.871873	-2.365935
47	1	-0.889184	-0.585230	-2.388590
48	7	-3.306019	2.885371	-0.785464
49	7	2.897794	3.300625	-0.793233
50	7	3.344256	-2.859035	-0.822606
51	7	-2.923275	-3.277361	-0.797545
52	1	-4.271574	3.121088	-0.503819
53	1	3.128676	4.270090	-0.521237
54	1	4.301162	-3.115576	-0.526950
55	1	-3.174257	-4.238762	-0.511762
56	6	-3.706858	-2.938554	-1.978877

57	1	-3.052483	-2.715181	-2.827696
58	6	-2.988766	3.634464	-1.994525
59	1	-2.732734	2.955512	-2.814121
60	6	3.648282	2.975678	-1.999404
61	1	2.970446	2.710200	-2.816935
62	6	3.031174	-3.603821	-2.035682
63	1	2.826924	-2.921613	-2.867349
64	1	-4.329188	-2.058065	-1.790300
65	1	-2.133703	4.297130	-1.826138
66	1	4.314254	2.124713	-1.823509
67	1	2.146400	-4.230730	-1.886802
68	6	-4.259476	4.468416	-2.351772
69	8	-5.234063	4.330207	-1.549501
70	8	-4.175516	5.175898	-3.380642
71	6	4.477466	4.246474	-2.367557
72	8	4.333877	5.228046	-1.574821
73	8	5.186800	4.155749	-3.394588
74	6	4.280171	-4.486454	-2.352385
75	8	4.192450	-5.209909	-3.369314
76	8	5.243207	-4.363332	-1.533907
77	6	-4.599516	-4.181214	-2.292972
78	8	-4.453437	-5.159356	-1.496455
79	8	-5.354292	-4.073653	-3.284871
80	6	1.980752	4.267146	1.727564
81	1	3.016085	4.054674	2.005858
82	1	1.971365	5.121392	1.045294
83	1	1.436822	4.578627	2.621752
84	6	-4.263031	1.959043	1.734449
85	1	-4.054842	2.996069	2.009639
86	1	-5.120451	1.943845	1.056223
87	1	-4.567255	1.415422	2.631382
88	6	-1.946950	-4.277208	1.681683
89	1	-2.978386	-4.073899	1.980651
90	1	-1.944780	-5.120910	0.986328
91	1	-1.387040	-4.599701	2.561802
92	6	4.289405	-1.961200	1.708507
93	1	4.082517	-3.002455	1.968423
94	1	5.149302	-1.934276	1.033798
95	1	4.588929	-1.429921	2.614217
96	63	0.009644	0.016577	0.304796

E(RTPSSh) = -2546.5959563 Hartree
Zero-point correction = 0.781668 Hartree/particle
Sum of electronic and thermal Energies = -2545.761098 Hartree
Sum of electronic and thermal Enthalpies = -2545.760154 Hartree
Sum of electronic and thermal Free Energies = -2545.905674 Hartree

Table S52. [Gd(DOTMA-$(gly)_4(H_2O)]^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	1.004271	-1.898560	-1.936043
2	6	0.028477	-2.236341	-3.007393
3	6	-1.416892	-2.303715	-2.525066
4	7	-1.903672	-1.015602	-1.943989
5	6	-2.216976	-0.050783	-3.032452
6	6	-2.293629	1.399742	-2.565699
7	7	-1.015827	1.892029	-1.964858
8	6	-0.041163	2.219586	-3.040526

9	6	1.405041	2.288364	-2.560096
10	7	1.888231	1.007269	-1.959633
11	6	2.206050	0.027492	-3.032681
12	6	2.282332	-1.415708	-2.543757
13	6	1.274439	-3.060102	-1.026343
14	6	2.043079	-2.499470	0.177190
15	8	1.817125	-1.301430	0.547263
16	6	-3.081486	-1.268617	-1.051655
17	6	-2.545624	-2.006156	0.181697
18	8	-1.352581	-1.777694	0.567871
19	6	-1.287234	3.060069	-1.064460
20	6	-2.045180	2.502188	0.148902
21	8	-1.836361	1.300476	0.508615
22	6	3.060631	1.272373	-1.063512
23	6	2.510109	2.027789	0.153813
24	8	1.313828	1.809769	0.526486
25	1	0.127788	-1.475071	-3.783776
26	1	0.275484	-3.190347	-3.489017
27	1	-1.519933	-3.079349	-1.761408
28	1	-2.042939	-2.610129	-3.373295
29	1	-1.437023	-0.157815	-3.789248
30	1	-3.159913	-0.302559	-3.533448
31	1	-3.084362	1.510473	-1.819133
32	1	-2.582630	2.017715	-3.425795
33	1	-0.142744	1.452428	-3.811045
34	1	-0.286995	3.170564	-3.528978
35	1	1.511006	3.074947	-1.808201
36	1	2.031747	2.579872	-3.413116
37	1	1.429536	0.122663	-3.794560
38	1	3.151092	0.272182	-3.533355
39	1	3.072622	-1.514629	-1.794835
40	1	2.572556	-2.046883	-3.393846
41	1	0.300749	-3.364694	-0.623968
42	1	-3.394799	-0.286894	-0.677527
43	1	-0.312592	3.375554	-0.673284
44	1	3.374002	0.296272	-0.674877
45	8	0.024108	-0.141880	2.207017
46	1	-0.625595	-0.857842	2.330175
47	1	0.888840	-0.566743	2.349089
48	7	3.297595	2.862701	0.798083
49	7	-2.874197	3.289373	0.801221
50	7	-3.336883	-2.833206	0.828529
51	7	2.894669	-3.273974	0.812609
52	1	4.267300	3.094602	0.527749
53	1	-3.099246	4.263344	0.540690
54	1	-4.299107	-3.082371	0.543750
55	1	3.141748	-4.239487	0.537110
56	6	3.669877	-2.931333	1.998432
57	1	3.009551	-2.700028	2.840556
58	6	2.971772	3.607790	2.007357
59	1	2.704226	2.926386	2.821233
60	6	-3.621985	2.956999	2.007061
61	1	-2.942682	2.678470	2.819025
62	6	-3.021304	-3.570442	2.045603
63	1	-2.801722	-2.883188	2.869145
64	1	4.298074	-2.055065	1.809893
65	1	2.121777	4.275554	1.833566
66	1	-4.294657	2.112583	1.825245
67	1	-2.145475	-4.209337	1.894821
68	6	4.243028	4.433638	2.381288
69	8	5.225028	4.293917	1.588361
70	8	4.152226	5.136883	3.412488

71	6	-4.440690	4.229710	2.391848
72	8	-4.292581	5.218812	1.609365
73	8	-5.147325	4.132869	3.420184
74	6	-4.277805	-4.435106	2.381857
75	8	-4.190560	-5.147580	3.406521
76	8	-5.245390	-4.310613	1.568959
77	6	4.553404	-4.176580	2.327934
78	8	4.406673	-5.160021	1.538069
79	8	5.300827	-4.066305	3.325047
80	6	-1.970043	4.266827	-1.723716
81	1	-3.006168	4.055265	-1.999705
82	1	-1.958758	5.118912	-1.038788
83	1	-1.428464	4.580909	-2.618479
84	6	4.267763	1.951297	-1.725933
85	1	4.060645	2.989399	-1.997798
86	1	5.122577	1.933452	-1.044494
87	1	4.575148	1.411025	-2.623862
88	6	1.943454	-4.278538	-1.677802
89	1	2.977496	-4.076086	-1.968108
90	1	1.934910	-5.121674	-0.981820
91	1	1.390650	-4.601174	-2.562388
92	6	-4.285500	-1.955245	-1.712007
93	1	-4.078967	-2.997839	-1.966697
94	1	-5.145043	-1.924796	-1.037018
95	1	-4.585220	-1.428308	-2.620243
96	64	-0.008244	0.015899	-0.333447

E(RTPSSh) = -2547.1861739 Hartree
Zero-point correction = 0.781926 Hartree/particle
Sum of electronic and thermal Energies = -2546.351198 Hartree
Sum of electronic and thermal Enthalpies = -2546.350253 Hartree
Sum of electronic and thermal Free Energies = -2546.495205 Hartree

Table S53. [Tb(DOTMA-(gly)$_4$(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.987420	-1.901001	-1.939571
2	6	0.012399	-2.239763	-3.010797
3	6	-1.431718	-2.298918	-2.527162
4	7	-1.905882	-1.006087	-1.947920
5	6	-2.217982	-0.044537	-3.039015
6	6	-2.287099	1.406032	-2.574414
7	7	-1.005318	1.888103	-1.975249
8	6	-0.032439	2.213112	-3.052775
9	6	1.412972	2.274178	-2.572015
10	7	1.883785	0.990112	-1.969454
11	6	2.199574	0.009616	-3.041906
12	6	2.268794	-1.431593	-2.549313
13	6	1.250305	-3.061185	-1.025547
14	6	2.019733	-2.499264	0.176125
15	8	1.798851	-1.298113	0.539573
16	6	-3.083388	-1.249567	-1.052173
17	6	-2.547538	-1.984485	0.182073
18	8	-1.352049	-1.759405	0.563387
19	6	-1.268785	3.058060	-1.074467
20	6	-2.024071	2.503650	0.141489
21	8	-1.822459	1.299291	0.496992
22	6	3.057430	1.248755	-1.072453

23	6	2.509410	2.005055	0.144723
24	8	1.310515	1.794087	0.513663
25	1	0.114861	-1.482318	-3.790483
26	1	0.256758	-3.196581	-3.488104
27	1	-1.538555	-3.072536	-1.761958
28	1	-2.062010	-2.602225	-3.373320
29	1	-1.440191	-0.156920	-3.797285
30	1	-3.162960	-0.294537	-3.537028
31	1	-3.076376	1.521984	-1.827083
32	1	-2.572714	2.025949	-3.434184
33	1	-0.138419	1.447856	-3.824619
34	1	-0.275318	3.165893	-3.539139
35	1	1.523338	3.060869	-1.820825
36	1	2.043526	2.559887	-3.424109
37	1	1.424779	0.106989	-3.805258
38	1	3.146266	0.250806	-3.541111
39	1	3.058820	-1.532880	-1.800407
40	1	2.554050	-2.068449	-3.396801
41	1	0.274858	-3.359888	-0.623503
42	1	-3.391352	-0.265189	-0.681122
43	1	-0.291735	3.370411	-0.687273
44	1	3.366571	0.271177	-0.684665
45	8	0.021147	-0.122149	2.188347
46	1	-0.625862	-0.838791	2.320046
47	1	0.887015	-0.540964	2.339572
48	7	3.299859	2.833597	0.793159
49	7	-2.843572	3.294765	0.800675
50	7	-3.339603	-2.805924	0.834671
51	7	2.867148	-3.274043	0.816433
52	1	4.272857	3.057877	0.528121
53	1	-3.061268	4.271925	0.545785
54	1	-4.304585	-3.050703	0.555204
55	1	3.109125	-4.242574	0.546836
56	6	3.641609	-2.929801	2.002258
57	1	2.980653	-2.690162	2.841533
58	6	2.976408	3.574175	2.005846
59	1	2.695965	2.890274	2.813197
60	6	-3.582632	2.965744	2.012813
61	1	-2.898141	2.674014	2.815662
62	6	-3.024359	-3.537394	2.055285
63	1	-2.794897	-2.846329	2.872869
64	1	4.275554	-2.058382	1.810671
65	1	2.135584	4.253492	1.832155
66	1	-4.268202	2.131090	1.833999
67	1	-2.154801	-4.184859	1.904806
68	6	4.255596	4.381813	2.392287
69	8	5.239333	4.237275	1.602349
70	8	4.168540	5.077390	3.428987
71	6	-4.380546	4.246937	2.412785
72	8	-4.230273	5.237101	1.632019
73	8	-5.074910	4.154973	3.449892
74	6	-4.286701	-4.388776	2.403442
75	8	-4.201462	-5.093515	3.433583
76	8	-5.256319	-4.262756	1.593167
77	6	4.516471	-4.178889	2.340355
78	8	4.364130	-5.166036	1.556162
79	8	5.263217	-4.067756	3.337852
80	6	-1.950177	4.267169	-1.731070
81	1	-2.988659	4.059819	-2.001243
82	1	-1.931654	5.119510	-1.046628
83	1	-1.412200	4.578656	-2.628911
84	6	4.268124	1.923519	-1.732727

85	1	4.066337	2.963308	-2.002042
86	1	5.122277	1.899794	-1.050644
87	1	4.573716	1.384057	-2.631766
88	6	1.914973	-4.285219	-1.671000
89	1	2.950624	-4.088951	-1.959736
90	1	1.900779	-5.125459	-0.971621
91	1	1.362687	-4.608892	-2.555525
92	6	-4.292291	-1.933465	-1.706445
93	1	-4.091606	-2.978236	-1.956799
94	1	-5.149838	-1.895629	-1.029293
95	1	-4.591918	-1.409340	-2.616341
96	65	-0.010325	0.016241	-0.336397

E(RTPSSh) = -2547.7756483 Hartree
Zero-point correction = 0.781873 Hartree/particle
Sum of electronic and thermal Energies = -2546.940698 Hartree
Sum of electronic and thermal Enthalpies = -2546.939754 Hartree
Sum of electronic and thermal Free Energies = -2547.084962 Hartree

Table S54. [Dy(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.963223	-1.918700	-1.948641
2	6	-0.011133	-2.248593	-3.023373
3	6	-1.457079	-2.282927	-2.543986
4	7	-1.909270	-0.980935	-1.966850
5	6	-2.205140	-0.016081	-3.060096
6	6	-2.256673	1.434394	-2.593633
7	7	-0.969152	1.896273	-1.990466
8	6	0.008556	2.212677	-3.066366
9	6	1.453378	2.248699	-2.582140
10	7	1.898963	0.955716	-1.978745
11	6	2.203596	-0.028542	-3.051140
12	6	2.253263	-1.469345	-2.555519
13	6	1.206970	-3.079286	-1.030183
14	6	1.973370	-2.521204	0.174405
15	8	1.768579	-1.313510	0.525546
16	6	-3.092249	-1.205130	-1.073586
17	6	-2.567206	-1.936073	0.167249
18	8	-1.364548	-1.734617	0.539416
19	6	-1.218208	3.068081	-1.088310
20	6	-1.973774	2.518928	0.129404
21	8	-1.798854	1.306000	0.469208
22	6	3.073420	1.195453	-1.077505
23	6	2.528770	1.953190	0.139551
24	8	1.322668	1.761919	0.495269
25	1	0.105892	-1.496411	-3.805603
26	1	0.221442	-3.211207	-3.494936
27	1	-1.578469	-3.053179	-1.777659
28	1	-2.091630	-2.575650	-3.390485
29	1	-1.426242	-0.138549	-3.815855
30	1	-3.151646	-0.254738	-3.560833
31	1	-3.045748	1.559043	-1.847641
32	1	-2.530905	2.061050	-3.452040
33	1	-0.108018	1.454058	-3.843079
34	1	-0.220134	3.171938	-3.546897
35	1	1.574821	3.032352	-1.829582
36	1	2.092509	2.523429	-3.431274

37	1	1.431117	0.078061	-3.815531
38	1	3.153919	0.200832	-3.549114
39	1	3.039291	-1.579015	-1.803696
40	1	2.531594	-2.113106	-3.399881
41	1	0.225636	-3.367300	-0.634970
42	1	-3.392284	-0.215168	-0.711480
43	1	-0.237115	3.372855	-0.705827
44	1	3.370146	0.212977	-0.692767
45	8	0.012785	-0.106473	2.157081
46	1	-0.640222	-0.817656	2.287971
47	1	0.874732	-0.532149	2.310740
48	66	-0.009020	0.012938	-0.362202
49	7	3.327739	2.763426	0.800420
50	7	-2.767250	3.322497	0.805137
51	7	-3.375324	-2.729287	0.834753
52	7	2.801609	-3.304236	0.829416
53	1	4.308703	2.967834	0.548865
54	1	-2.960389	4.308490	0.565043
55	1	-4.349931	-2.948209	0.567406
56	1	3.024239	-4.281761	0.575671
57	6	3.563212	-2.965697	2.025199
58	1	2.894172	-2.691472	2.847147
59	6	3.007333	3.500939	2.015846
60	1	2.700851	2.817029	2.813636
61	6	-3.501125	2.999203	2.021973
62	1	-2.816379	2.675760	2.812302
63	6	-3.074968	-3.445573	2.068069
64	1	-2.810276	-2.746319	2.867710
65	1	4.228922	-2.118306	1.833077
66	1	2.185378	4.202070	1.838416
67	1	-4.214078	2.188331	1.840426
68	1	-2.232694	-4.129523	1.923367
69	6	4.301256	4.273667	2.424096
70	8	5.290564	4.110426	1.644774
71	8	4.219057	4.963110	3.465229
72	6	-4.255113	4.298406	2.448359
73	8	-4.081331	5.294842	1.680490
74	8	-4.942432	4.212662	3.490637
75	6	-4.365032	-4.240664	2.447223
76	8	-4.298010	-4.920299	3.495403
77	8	-5.335232	-4.100965	1.639852
78	6	4.391085	-4.236358	2.399219
79	8	4.224417	-5.229137	1.624962
80	8	5.119356	-4.134337	3.411175
81	6	-1.893601	4.282761	-1.741708
82	1	-2.935914	4.084566	-2.003681
83	1	-1.862333	5.135856	-1.058708
84	1	-1.359386	4.588248	-2.643947
85	6	4.293836	1.859627	-1.730838
86	1	4.104367	2.902494	-1.997093
87	1	5.145013	1.824377	-1.045537
88	1	4.597226	1.319945	-2.630541
89	6	1.864935	-4.311275	-1.667561
90	1	2.904662	-4.125593	-1.948493
91	1	1.837337	-5.149589	-0.966265
92	1	1.316375	-4.631848	-2.555588
93	6	-4.305519	-1.883013	-1.726702
94	1	-4.115011	-2.932502	-1.964813
95	1	-5.165899	-1.828852	-1.054300
96	1	-4.595201	-1.365223	-2.643507

E(RTPSSh) = -2548.3617554 Hartree

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Zero-point correction = 0.781867 Hartree/particle
Sum of electronic and thermal Energies = -2547.526848 Hartree
Sum of electronic and thermal Enthalpies = -2547.525903 Hartree
Sum of electronic and thermal Free Energies = -2547.670990 Hartree
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Table S55. [Ho(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.982834	-1.896760	-1.945957
2	6	0.012559	-2.240717	-3.019174
3	6	-1.431155	-2.296584	-2.536809
4	7	-1.898352	-1.001796	-1.956728
5	6	-2.215245	-0.043441	-3.048646
6	6	-2.281281	1.406167	-2.583343
7	7	-0.997465	1.880931	-1.982983
8	6	-0.027722	2.210730	-3.061343
9	6	1.416758	2.268607	-2.580080
10	7	1.880032	0.982674	-1.976034
11	6	2.201576	0.005068	-3.048828
12	6	2.267059	-1.435062	-2.555116
13	6	1.242045	-3.052010	-1.025256
14	6	2.004615	-2.479047	0.174383
15	8	1.784089	-1.272362	0.520060
16	6	-3.071367	-1.243627	-1.055438
17	6	-2.524414	-1.971957	0.176795
18	8	-1.323174	-1.747843	0.541156
19	6	-1.258871	3.046356	-1.076141
20	6	-2.008952	2.480586	0.136804
21	8	-1.808430	1.271006	0.474908
22	6	3.048501	1.239653	-1.072374
23	6	2.488090	1.990450	0.141493
24	8	1.283761	1.779706	0.492607
25	1	0.116910	-1.486329	-3.801625
26	1	0.258676	-3.199637	-3.491379
27	1	-1.540534	-3.070543	-1.772346
28	1	-2.063462	-2.596465	-3.382593
29	1	-1.440164	-0.156693	-3.809625
30	1	-3.162317	-0.294437	-3.542214
31	1	-3.070701	1.523488	-1.836429
32	1	-2.563788	2.029064	-3.441894
33	1	-0.134621	1.448217	-3.835833
34	1	-0.271695	3.165509	-3.543230
35	1	1.528694	3.055664	-1.829555
36	1	2.050231	2.550873	-3.431068
37	1	1.430238	0.103111	-3.815643
38	1	3.150822	0.247157	-3.542745
39	1	3.056214	-1.537585	-1.805509
40	1	2.550324	-2.075172	-3.400734
41	1	0.265580	-3.349650	-0.625606
42	1	-3.380046	-0.259054	-0.686286
43	1	-0.281497	3.358206	-0.690163
44	1	3.357434	0.261886	-0.685687
45	8	0.020443	-0.097553	2.150583
46	1	-0.617726	-0.821390	2.285346
47	1	0.891173	-0.506419	2.300914
48	7	3.270636	2.814924	0.804193
49	7	-2.823986	3.264446	0.809713
50	7	-3.310210	-2.787127	0.844178

51	7	2.846346	-3.246895	0.829865
52	1	4.247711	3.038248	0.553470
53	1	-3.040368	4.245383	0.568395
54	1	-4.279634	-3.030152	0.578337
55	1	3.087501	-4.219551	0.574173
56	6	3.615599	-2.890042	2.015456
57	1	2.951108	-2.639775	2.848794
58	6	2.932657	3.551279	2.015564
59	1	2.640433	2.864729	2.816441
60	6	-3.559621	2.921687	2.020186
61	1	-2.873031	2.618960	2.817120
62	6	-2.983793	-3.511264	2.066284
63	1	-2.741104	-2.815468	2.875958
64	1	4.252197	-2.022175	1.816901
65	1	2.095562	4.232994	1.833606
66	1	-4.247296	2.090566	1.833325
67	1	-2.119610	-4.164540	1.910207
68	6	4.208483	4.354647	2.421789
69	8	5.202366	4.210163	1.644599
70	8	4.109234	5.046886	3.459601
71	6	-4.353765	4.199433	2.438496
72	8	-4.202807	5.199291	1.670286
73	8	-5.045880	4.095620	3.475963
74	6	-4.246608	-4.353162	2.435749
75	8	-4.153703	-5.049211	3.471057
76	8	-5.224274	-4.228998	1.634869
77	6	4.486019	-4.137127	2.372375
78	8	4.333826	-5.133494	1.599855
79	8	5.228998	-4.015608	3.371402
80	6	-1.945138	4.257587	-1.723513
81	1	-2.984089	4.049933	-1.991592
82	1	-1.926132	5.105917	-1.034113
83	1	-1.410422	4.575165	-2.621207
84	6	4.261484	1.919846	-1.722652
85	1	4.059146	2.960112	-1.989656
86	1	5.111341	1.895747	-1.035230
87	1	4.573744	1.383965	-2.621574
88	6	1.912251	-4.278561	-1.659964
89	1	2.948999	-4.082372	-1.944739
90	1	1.896013	-5.114608	-0.955617
91	1	1.364678	-4.608276	-2.545215
92	6	-4.281574	-1.933883	-1.700501
93	1	-4.079649	-2.979317	-1.947022
94	1	-5.135898	-1.895059	-1.019346
95	1	-4.586673	-1.414460	-2.611311
96	67	-0.009968	0.016223	-0.358846

E(RTPSSh) = -2548.9457093 Hartree
Zero-point correction = 0.782121 Hartree/particle
Sum of electronic and thermal Energies = -2548.110619 Hartree
Sum of electronic and thermal Enthalpies = -2548.109675 Hartree
Sum of electronic and thermal Free Energies = -2548.254636 Hartree

Table S56. [Tm(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.921450	-1.915423	-1.961735
2	6	-0.055751	-2.236628	-3.035756

3	6	-1.499120	-2.246174	-2.552251
4	7	-1.919367	-0.937130	-1.968567
5	6	-2.216536	0.029729	-3.058761
6	6	-2.236805	1.478219	-2.588859
7	7	-0.937056	1.906884	-1.988752
8	6	0.037964	2.217055	-3.068497
9	6	1.483118	2.227689	-2.588527
10	7	1.902277	0.925477	-1.987579
11	6	2.200120	-0.056489	-3.063456
12	6	2.220395	-1.497437	-2.570866
13	6	1.146615	-3.075486	-1.038023
14	6	1.915280	-2.515942	0.162982
15	8	1.734906	-1.297393	0.489722
16	6	-3.094539	-1.145727	-1.061396
17	6	-2.556417	-1.884074	0.167733
18	8	-1.342613	-1.699013	0.512141
19	6	-1.162274	3.074637	-1.074776
20	6	-1.917904	2.518129	0.138055
21	8	-1.756620	1.296959	0.454052
22	6	3.075819	1.145385	-1.080638
23	6	2.528460	1.899913	0.135782
24	8	1.313062	1.727485	0.468591
25	1	0.071428	-1.489482	-3.821935
26	1	0.163004	-3.204823	-3.502384
27	1	-1.632812	-3.017139	-1.788586
28	1	-2.143839	-2.522501	-3.396561
29	1	-1.450549	-0.105007	-3.825607
30	1	-3.174031	-0.193979	-3.545123
31	1	-3.020187	1.616967	-1.839278
32	1	-2.500352	2.114697	-3.443333
33	1	-0.093346	1.464290	-3.848901
34	1	-0.179708	3.182514	-3.541450
35	1	1.620240	3.008279	-1.835434
36	1	2.126558	2.490820	-3.438001
37	1	1.435121	0.066299	-3.833275
38	1	3.158263	0.159880	-3.551981
39	1	3.005040	-1.624446	-1.820270
40	1	2.482699	-2.147883	-3.415186
41	1	0.161340	-3.350023	-0.644288
42	1	-3.377515	-0.153085	-0.694071
43	1	-0.175169	3.361227	-0.694907
44	1	3.360954	0.157583	-0.702105
45	8	0.019206	-0.078550	2.111548
46	1	-0.626046	-0.795822	2.246731
47	1	0.885129	-0.496845	2.262322
48	7	3.330033	2.689096	0.818310
49	7	-2.698535	3.318492	0.831904
50	7	-3.358617	-2.668741	0.851692
51	7	2.721988	-3.303817	0.838315
52	1	4.316977	2.882395	0.582102
53	1	-2.884853	4.309394	0.607237
54	1	-4.340106	-2.879246	0.602859
55	1	2.927941	-4.289179	0.601466
56	6	3.487159	-2.958136	2.029864
57	1	2.820717	-2.665554	2.847668
58	6	3.000635	3.423091	2.033500
59	1	2.677558	2.737653	2.823387
60	6	-3.435359	2.980366	2.042982
61	1	-2.752305	2.646274	2.830342
62	6	-3.038106	-3.396482	2.073279
63	1	-2.758157	-2.704759	2.874348
64	1	4.162519	-2.121161	1.826948

65	1	2.187347	4.132419	1.849163
66	1	-4.148495	2.172511	1.849666
67	1	-2.200126	-4.080829	1.907895
68	6	4.295735	4.181838	2.463648
69	8	5.294765	4.013015	1.698109
70	8	4.204861	4.866998	3.506967
71	6	-4.188350	4.274696	2.485557
72	8	-4.011966	5.281373	1.731869
73	8	-4.877113	4.175639	3.525767
74	6	-4.323689	-4.190923	2.468599
75	8	-4.237986	-4.884730	3.506024
76	8	-5.309900	-4.035528	1.683882
77	6	4.299146	-4.233477	2.422574
78	8	4.116750	-5.236893	1.665941
79	8	5.031541	-4.124256	3.430816
80	6	-1.825061	4.305216	-1.710204
81	1	-2.871292	4.124222	-1.968802
82	1	-1.778759	5.149341	-1.016942
83	1	-1.291557	4.614129	-2.611695
84	6	4.305976	1.802799	-1.722229
85	1	4.129330	2.850317	-1.978569
86	1	5.153542	1.751169	-1.033493
87	1	4.607085	1.268834	-2.626168
88	6	1.793747	-4.318367	-1.665184
89	1	2.837704	-4.147181	-1.939165
90	1	1.750491	-5.152873	-0.960155
91	1	1.247633	-4.636056	-2.555784
92	6	-4.324557	-1.809187	-1.697117
93	1	-4.149428	-2.860202	-1.939984
94	1	-5.173728	-1.746815	-1.011277
95	1	-4.622308	-1.286319	-2.608463
96	69	-0.008761	0.018839	-0.389646

E(RTPSSh) = -2550.108215 Hartree
Zero-point correction = 0.782127 Hartree/particle
Sum of electronic and thermal Energies = -2549.273182 Hartree
Sum of electronic and thermal Enthalpies = -2549.272238 Hartree
Sum of electronic and thermal Free Energies = -2549.416688 Hartree

Table S57. [Yb(DOTMA-(gly)$_4$(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.743136	-1.973706	-1.955866
2	6	-0.256741	-2.209884	-3.029996
3	6	-1.693306	-2.098844	-2.540867
4	7	-1.990397	-0.758514	-1.953083
5	6	-2.212566	0.228227	-3.042882
6	6	-2.106508	1.671291	-2.570595
7	7	-0.770019	1.976001	-1.976899
8	6	0.218945	2.203040	-3.063320
9	6	1.660384	2.091849	-2.587878
10	7	1.960328	0.757394	-1.987987
11	6	2.175175	-0.243466	-3.065060
12	6	2.073573	-1.678548	-2.567593
13	6	0.865148	-3.147164	-1.027967
14	6	1.685936	-2.653190	0.167154
15	8	1.611927	-1.421191	0.488699
16	6	-3.173868	-0.865461	-1.036460

17	6	-2.691861	-1.658068	0.181820
18	8	-1.463354	-1.575258	0.517661
19	6	-0.885125	3.155311	-1.054986
20	6	-1.690093	2.660985	0.153312
21	8	-1.641935	1.426123	0.455595
22	6	3.148713	0.873744	-1.079044
23	6	2.666650	1.675207	0.134882
24	8	1.438768	1.609095	0.462547
25	1	-0.072511	-1.470809	-3.812947
26	1	-0.117709	-3.190279	-3.502322
27	1	-1.888003	-2.857207	-1.777468
28	1	-2.365554	-2.313651	-3.381398
29	1	-1.463588	0.034497	-3.814245
30	1	-3.187285	0.083437	-3.525271
31	1	-2.870759	1.874510	-1.815969
32	1	-2.314202	2.333180	-3.420982
33	1	0.025015	1.460461	-3.840925
34	1	0.077478	3.181464	-3.539111
35	1	1.864092	2.857344	-1.834204
36	1	2.324951	2.295609	-3.437177
37	1	1.421585	-0.062124	-3.835244
38	1	3.147125	-0.106455	-3.555476
39	1	2.844722	-1.868527	-1.816062
40	1	2.274212	-2.355219	-3.407970
41	1	-0.140085	-3.329935	-0.630453
42	1	-3.363015	0.146787	-0.661255
43	1	0.124689	3.346587	-0.674029
44	1	3.346003	-0.135516	-0.700391
45	8	0.014148	-0.061437	2.115863
46	1	-0.681808	-0.727866	2.258660
47	1	0.847292	-0.538832	2.274178
48	7	3.531206	2.393961	0.815918
49	7	-2.395712	3.523051	0.851213
50	7	-3.549439	-2.384074	0.860752
51	7	2.425900	-3.504297	0.839730
52	1	4.537120	2.508192	0.589926
53	1	-2.501345	4.532929	0.639769
54	1	-4.551123	-2.523495	0.623463
55	1	2.557735	-4.509106	0.612110
56	6	3.232148	-3.226090	2.022290
57	1	2.603438	-2.883939	2.850765
58	6	3.270240	3.164598	2.025477
59	1	2.895144	2.517166	2.824463
60	6	-3.173439	3.250031	2.053526
61	1	-2.531512	2.863267	2.851393
62	6	-3.284420	-3.164442	2.063480
63	1	-2.958646	-2.516809	2.884011
64	1	3.972088	-2.447124	1.813442
65	1	2.519531	3.939157	1.838306
66	1	-3.949636	2.504443	1.853169
67	1	-2.498067	-3.903576	1.881397
68	6	4.635257	3.811382	2.434468
69	8	5.604512	3.531466	1.661707
70	8	4.615470	4.521696	3.461947
71	6	-3.817545	4.612234	2.476153
72	8	-3.527539	5.590733	1.718979
73	8	-4.535909	4.581546	3.497803
74	6	-4.633499	-3.870728	2.427949
75	8	-4.607658	-4.608561	3.435001
76	8	-5.596745	-3.600616	1.644221
77	6	3.938555	-4.573531	2.392306
78	8	3.650842	-5.545806	1.626180

79	8	4.694162	-4.537748	3.385833
80	6	-1.432970	4.445367	-1.681748
81	1	-2.489758	4.360168	-1.947246
82	1	-1.318245	5.274056	-0.977740
83	1	-0.869223	4.718064	-2.576804
84	6	4.432174	1.422332	-1.718785
85	1	4.346064	2.480446	-1.978539
86	1	5.268733	1.302969	-1.024890
87	1	4.693328	0.862427	-2.619755
88	6	1.396344	-4.445276	-1.652075
89	1	2.448801	-4.367308	-1.936521
90	1	1.289652	-5.266800	-0.938445
91	1	0.816442	-4.723630	-2.534780
92	6	-4.462904	-1.413661	-1.664694
93	1	-4.381346	-2.472928	-1.921481
94	1	-5.294342	-1.289161	-0.965583
95	1	-4.728637	-0.856387	-2.565744
96	70	-0.010768	0.023529	-0.370722

E(RTPSSh) = -2550.6691996 Hartree
Zero-point correction = 0.781801 Hartree/particle
Sum of electronic and thermal Energies = -2549.834625 Hartree
Sum of electronic and thermal Enthalpies = -2549.833681 Hartree
Sum of electronic and thermal Free Energies = -2549.977647 Hartree

Table S58. [Lu(DOTMA-(gly)$_4$(H$_2$O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-0.722692	1.977754	-1.959643
2	6	0.278238	2.205533	-3.034352
3	6	1.713074	2.079627	-2.544781
4	7	1.994788	0.736274	-1.956402
5	6	2.209767	-0.252298	-3.045790
6	6	2.087932	-1.693336	-2.572107
7	7	0.747865	-1.981801	-1.978349
8	6	-0.242143	-2.201917	-3.065045
9	6	-1.681934	-2.075181	-2.589544
10	7	-1.966202	-0.737002	-1.990438
11	6	-2.173681	0.264420	-3.068142
12	6	-2.056699	1.697964	-2.570629
13	6	-0.832494	3.151028	-1.030332
14	6	-1.654630	2.661260	0.165127
15	8	-1.593228	1.426771	0.480463
16	6	3.177468	0.832338	-1.037842
17	6	2.698337	1.627756	0.179332
18	8	1.467066	1.557591	0.508492
19	6	0.851080	-3.159747	-1.053402
20	6	1.659556	-2.667918	0.153183
21	8	1.623614	-1.430843	0.448347
22	6	-3.153654	-0.840849	-1.079016
23	6	-2.674608	-1.645002	0.133876
24	8	-1.444239	-1.591271	0.454635
25	1	0.086910	1.469038	-3.818010
26	1	0.148541	3.187868	-3.505332
27	1	1.915013	2.835903	-1.781221
28	1	2.388533	2.286843	-3.384582
29	1	1.464481	-0.051758	-3.818949
30	1	3.187101	-0.117381	-3.525764

31	1	2.849548	-1.903735	-1.816829
32	1	2.288263	-2.359000	-3.421255
33	1	-0.041007	-1.462796	-3.844061
34	1	-0.109859	-3.182810	-3.538399
35	1	-1.893359	-2.837824	-1.835153
36	1	-2.349448	-2.271980	-3.438122
37	1	-1.423413	0.075618	-3.839720
38	1	-3.148038	0.136570	-3.556265
39	1	-2.825114	1.895618	-1.818298
40	1	-2.250403	2.377483	-3.410288
41	1	0.174930	3.324950	-0.634630
42	1	3.357797	-0.181587	-0.663087
43	1	-0.160550	-3.340653	-0.672430
44	1	-3.340840	0.170542	-0.701107
45	8	-0.014895	0.055072	2.104839
46	1	0.685728	0.716741	2.246610
47	1	-0.844819	0.539416	2.258946
48	7	-3.543076	-2.353405	0.820569
49	7	2.355706	-3.533252	0.856342
50	7	3.560005	2.343381	0.863868
51	7	-2.382878	3.517225	0.844014
52	1	-4.551191	-2.457488	0.599345
53	1	2.451568	-4.545138	0.649688
54	1	4.564128	2.473055	0.631167
55	1	-2.504795	4.524337	0.620708
56	6	-3.188218	3.243355	2.028213
57	1	-2.560371	2.892434	2.853714
58	6	-3.284972	-3.124677	2.030339
59	1	-2.899565	-2.479933	2.826580
60	6	3.135177	-3.262602	2.058064
61	1	2.496570	-2.865915	2.853717
62	6	3.297873	3.124594	2.066686
63	1	2.963088	2.478908	2.885165
64	1	-3.936728	2.472645	1.819359
65	1	-2.543496	-3.907634	1.841347
66	1	3.919080	-2.525841	1.855099
67	1	2.519293	3.871455	1.882827
68	6	-4.655101	-3.755948	2.446500
69	8	-5.624509	-3.467661	1.677017
70	8	-4.638552	-4.463864	3.475678
71	6	3.765102	-4.629368	2.487354
72	8	3.466404	-5.608136	1.733916
73	8	4.482326	-4.601423	3.509870
74	6	4.652372	3.817252	2.437289
75	8	4.629789	4.553753	3.445365
76	8	5.615877	3.538943	1.656773
77	6	-3.879488	4.596857	2.404794
78	8	-3.583976	5.568572	1.640946
79	8	-4.632314	4.565541	3.400544
80	6	1.387921	-4.456507	-1.675722
81	1	2.445631	-4.381795	-1.940762
82	1	1.265295	-5.281850	-0.969103
83	1	0.822242	-4.726973	-2.570256
84	6	-4.443584	-1.378895	-1.714623
85	1	-4.367715	-2.438053	-1.973364
86	1	-5.277122	-1.251286	-1.018552
87	1	-4.702184	-0.817654	-2.615513
88	6	-1.354591	4.454390	-1.651191
89	1	-2.408548	4.385939	-1.932552
90	1	-1.238705	5.274045	-0.936839
91	1	-0.774887	4.728816	-2.535281
92	6	4.472152	1.370983	-1.662735

93	1	4.400089	2.431329	-1.917913
94	1	5.301169	1.238360	-0.962237
95	1	4.734883	0.812902	-2.564192
96	71	0.010271	-0.024211	-0.377916

E(RTPSSh) = -2551.2227829 Hartree
Zero-point correction = 0.782063 Hartree/particle
Sum of electronic and thermal Energies = -2550.388021 Hartree
Sum of electronic and thermal Enthalpies = -2550.387077 Hartree
Sum of electronic and thermal Free Energies = -2550.530756 Hartree

Table S59. [La(DOTA-(gly)$_4$(H$_2$O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.557447	2.097087	1.714084
2	6	1.472982	1.641656	2.800937
3	6	2.585585	0.704094	2.337715
4	7	2.070047	-0.555499	1.718586
5	6	1.623713	-1.487480	2.795804
6	6	0.678853	-2.588645	2.321499
7	7	-0.578973	-2.056635	1.714203
8	6	-1.499209	-1.602491	2.798708
9	6	-2.610664	-0.664496	2.332747
10	7	-2.089768	0.590973	1.711676
11	6	-1.648421	1.526237	2.787930
12	6	-0.702523	2.627838	2.316343
13	6	1.188505	3.166099	0.899726
14	6	2.172016	2.604023	-0.118297
15	8	1.938497	1.475728	-0.652390
16	6	3.151858	-1.164918	0.902877
17	6	2.617099	-2.144335	-0.132417
18	8	1.497122	-1.910640	-0.690553
19	6	-1.206188	-3.128372	0.898761
20	6	-2.199209	-2.571613	-0.112593
21	8	-1.971929	-1.443566	-0.650573
22	6	-3.159887	1.201541	0.882159
23	6	-2.602827	2.169143	-0.153927
24	8	-1.477535	1.926585	-0.693419
25	1	0.863879	1.145224	3.559854
26	1	1.934559	2.509621	3.293325
27	1	3.220500	1.202542	1.604154
28	1	3.222360	0.474178	3.203337
29	1	1.137294	-0.889552	3.569792
30	1	2.495307	-1.958913	3.271632
31	1	1.170470	-3.214562	1.575271
32	1	0.448986	-3.237425	3.178247
33	1	-0.892429	-1.105989	3.559378
34	1	-1.960525	-2.471801	3.288618
35	1	-3.246583	-1.163139	1.600299
36	1	-3.246979	-0.431715	3.197995
37	1	-1.165097	0.930577	3.565666
38	1	-2.522720	1.997927	3.258764
39	1	-1.192542	3.253166	1.568521
40	1	-0.477311	3.277107	3.174174
41	1	0.396539	3.648031	0.317289
42	1	3.624850	-0.360110	0.330951
43	1	-0.412806	-3.602059	0.311326
44	1	-3.634407	0.396036	0.312613

45	8	0.136162	-0.459643	-2.608038
46	1	0.695234	-1.254987	-2.660444
47	1	-0.676888	-0.669165	-3.093229
48	7	-3.350605	3.198861	-0.488794
49	7	-3.233422	-3.323381	-0.425828
50	7	3.376383	-3.169368	-0.450813
51	7	3.206383	3.351962	-0.440731
52	1	-4.270260	3.436500	-0.068628
53	1	-3.454068	-4.247452	-0.005966
54	1	4.293034	-3.398562	-0.016805
55	1	3.432737	4.275672	-0.023911
56	6	4.204120	3.081802	-1.467760
57	1	3.730426	2.959162	-2.447065
58	6	-3.083449	4.179702	-1.532877
59	1	-2.970973	3.690426	-2.505694
60	6	-4.239318	-3.059550	-1.446572
61	1	-4.794738	-2.143730	-1.220130
62	6	3.134062	-4.160817	-1.491244
63	1	3.039249	-3.681297	-2.470894
64	1	-2.161034	4.730703	-1.323141
65	1	4.757318	2.164234	-1.242826
66	1	2.211148	-4.715365	-1.294269
67	1	-3.773084	-2.937788	-2.429596
68	6	4.367899	-5.125922	-1.467826
69	8	5.248950	-4.831505	-0.601192
70	8	4.341322	-6.061261	-2.294528
71	6	5.171651	4.313242	-1.474160
72	8	4.880179	5.217241	-0.630744
73	8	6.107796	4.263090	-2.299611
74	6	-4.310049	5.153187	-1.543830
75	8	-5.208107	4.879230	-0.688188
76	8	-4.262253	6.076059	-2.384074
77	6	-5.201486	-4.295432	-1.442573
78	8	-4.898919	-5.195905	-0.599237
79	8	-6.144294	-4.251628	-2.260590
80	57	-0.007600	0.011505	-0.037303
81	1	-1.667111	-3.898649	1.531088
82	1	-3.929057	1.680124	1.502610
83	1	1.659423	3.931186	1.531212
84	1	3.919458	-1.631230	1.534379

E(RTPSSh) = -2385.5681949 Hartree
Zero-point correction = 0.667502 Hartree/particle
Sum of electronic and thermal Energies = -2384.852552 Hartree
Sum of electronic and thermal Enthalpies = -2384.851608 Hartree
Sum of electronic and thermal Free Energies = -2384.988373 Hartree

Table S60. [Ce(DOTA-(gly)$_4$(H$_2$O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.620739	2.076249	1.749507
2	6	1.527594	1.601488	2.835605
3	6	2.610713	0.630549	2.371346
4	7	2.055289	-0.613175	1.754246
5	6	1.585754	-1.532909	2.832084
6	6	0.610976	-2.607057	2.356680
7	7	-0.631244	-2.038009	1.750189
8	6	-1.541212	-1.564125	2.835039

9	6	-2.623731	-0.593089	2.369114
10	7	-2.064341	0.646923	1.749709
11	6	-1.598390	1.570127	2.826158
12	6	-0.622855	2.644549	2.352329
13	6	1.278582	3.121480	0.925303
14	6	2.230644	2.524087	-0.102344
15	8	1.961122	1.395619	-0.619198
16	6	3.113132	-1.253909	0.931188
17	6	2.541864	-2.199108	-0.116139
18	8	1.422942	-1.925730	-0.658315
19	6	-1.286107	-3.086793	0.926686
20	6	-2.248896	-2.496069	-0.094214
21	8	-1.984619	-1.369112	-0.618367
22	6	-3.112933	1.287748	0.915487
23	6	-2.523951	2.223192	-0.132218
24	8	-1.399362	1.945751	-0.656204
25	1	0.908406	1.126661	3.600097
26	1	2.015754	2.458829	3.320702
27	1	3.258129	1.109415	1.635506
28	1	3.242679	0.382414	3.235432
29	1	1.115912	-0.922676	3.606770
30	1	2.444873	-2.027976	3.306562
31	1	1.085265	-3.244852	1.609134
32	1	0.362923	-3.251003	3.211948
33	1	-0.923362	-1.088797	3.600242
34	1	-2.028531	-2.422550	3.318756
35	1	-3.272533	-1.072217	1.634738
36	1	-3.254668	-0.342368	3.233228
37	1	-1.130826	0.962191	3.604092
38	1	-2.459675	2.065601	3.296441
39	1	-1.096111	3.281633	1.603513
40	1	-0.378283	3.289099	3.208329
41	1	0.497117	3.628417	0.350152
42	1	3.616368	-0.461322	0.368299
43	1	-0.503487	-3.585708	0.345932
44	1	-3.617481	0.494514	0.354783
45	8	0.065348	-0.395281	-2.536165
46	1	0.631607	-1.182648	-2.621996
47	1	-0.784694	-0.635903	-2.937576
48	7	-3.246020	3.261460	-0.495400
49	7	-3.295564	-3.218272	-0.433325
50	7	3.269957	-3.236934	-0.464392
51	7	3.276053	3.243084	-0.453029
52	1	-4.166523	3.524314	-0.092718
53	1	-3.544148	-4.142956	-0.030971
54	1	4.186869	-3.496951	-0.049700
55	1	3.529653	4.167933	-0.055144
56	6	4.240618	2.941198	-1.502762
57	1	3.736924	2.803466	-2.464817
58	6	-2.953058	4.207490	-1.564488
59	1	-2.826900	3.687058	-2.519189
60	6	-4.268589	-2.923072	-1.477191
61	1	-4.819776	-2.005096	-1.248818
62	6	2.991733	-4.191545	-1.530013
63	1	2.882383	-3.679363	-2.491324
64	1	-2.030471	4.758525	-1.355290
65	1	4.790783	2.022475	-1.274701
66	1	2.065249	-4.739290	-1.330307
67	1	-3.772358	-2.787991	-2.443548
68	6	4.210385	-5.174669	-1.564742
69	8	5.107822	-4.931615	-0.699076
70	8	4.157100	-6.073324	-2.430151

71	6	5.220850	4.160888	-1.562268
72	8	4.973271	5.078597	-0.719581
73	8	6.123806	4.089242	-2.422476
74	6	-4.171220	5.189339	-1.627015
75	8	-5.079996	4.957900	-0.770062
76	8	-4.107240	6.077506	-2.502943
77	6	-5.245163	-4.146421	-1.524558
78	8	-4.986504	-5.060507	-0.681250
79	8	-6.155636	-4.080495	-2.377043
80	58	-0.004181	0.012694	0.028128
81	1	-1.778942	-3.842442	1.552441
82	1	-3.863419	1.800045	1.531777
83	1	1.782129	3.871849	1.549118
84	1	3.862702	-1.756488	1.556374

E(RTPSSh) = -2386.2199658 Hartree
Zero-point correction = 0.667579 Hartree/particle
Sum of electronic and thermal Energies = -2385.504316 Hartree
Sum of electronic and thermal Enthalpies = -2385.503372 Hartree
Sum of electronic and thermal Free Energies = -2385.639620 Hartree

Table S61. [Pr(DOTA-(gly)$_4$(H$_2$O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.629835	2.070053	1.761120
2	6	1.538916	1.599392	2.847257
3	6	2.616965	0.623450	2.382739
4	7	2.052927	-0.617441	1.768109
5	6	1.581418	-1.534742	2.847107
6	6	0.604823	-2.606683	2.370752
7	7	-0.634334	-2.032568	1.762449
8	6	-1.546488	-1.561789	2.846863
9	6	-2.623792	-0.586364	2.379286
10	7	-2.055858	0.650807	1.762199
11	6	-1.588515	1.572196	2.839784
12	6	-0.610625	2.644126	2.365214
13	6	1.288200	3.107485	0.927328
14	6	2.226398	2.499081	-0.106533
15	8	1.947401	1.367094	-0.611688
16	6	3.103483	-1.264846	0.941171
17	6	2.519334	-2.197365	-0.109883
18	8	1.397779	-1.912625	-0.640680
19	6	-1.289581	-3.075405	0.931670
20	6	-2.239431	-2.476915	-0.096247
21	8	-1.963143	-1.349055	-0.614829
22	6	-3.097879	1.297557	0.924341
23	6	-2.497096	2.223342	-0.124653
24	8	-1.369356	1.937088	-0.636839
25	1	0.921038	1.129929	3.616124
26	1	2.030657	2.458115	3.326049
27	1	3.264654	1.098468	1.644500
28	1	3.249985	0.373179	3.245375
29	1	1.111886	-0.922745	3.620620
30	1	2.439231	-2.031010	3.322569
31	1	1.078516	-3.245970	1.624086
32	1	0.353082	-3.249856	3.225403
33	1	-0.930116	-1.090954	3.616024
34	1	-2.037485	-2.421034	3.325190

35	1	-3.272058	-1.062223	1.642121
36	1	-3.256784	-0.333260	3.241114
37	1	-1.121804	0.962785	3.617094
38	1	-2.448844	2.069054	3.310185
39	1	-1.082700	3.282811	1.617045
40	1	-0.362432	3.287811	3.220676
41	1	0.505677	3.617795	0.356807
42	1	3.614932	-0.475190	0.381789
43	1	-0.505908	-3.577982	0.355606
44	1	-3.608878	0.507267	0.365483
45	8	-0.017993	-0.372852	-2.488074
46	1	0.608338	-1.108108	-2.609939
47	1	-0.885725	-0.729273	-2.741739
48	7	-3.211446	3.262304	-0.500478
49	7	-3.285854	-3.191471	-0.449393
50	7	3.238190	-3.236281	-0.473280
51	7	3.269419	3.211834	-0.475272
52	1	-4.134037	3.531771	-0.106676
53	1	-3.544941	-4.116459	-0.053237
54	1	4.156212	-3.506454	-0.067544
55	1	3.532022	4.138045	-0.085857
56	6	4.221479	2.899917	-1.533568
57	1	3.707090	2.759292	-2.489500
58	6	-2.907189	4.200284	-1.573577
59	1	-2.775929	3.673308	-2.523973
60	6	-4.245975	-2.887986	-1.503010
61	1	-4.795646	-1.968553	-1.277020
62	6	2.945866	-4.179383	-1.545399
63	1	2.832207	-3.657666	-2.501054
64	1	-1.984439	4.749817	-1.361193
65	1	4.770291	1.980246	-1.306204
66	1	2.017554	-4.723254	-1.343478
67	1	-3.738571	-2.751756	-2.463366
68	6	4.158245	-5.169454	-1.598045
69	8	5.062113	-4.941406	-0.734923
70	8	4.094792	-6.058369	-2.472797
71	6	5.205797	4.115549	-1.609363
72	8	4.971191	5.037641	-0.767654
73	8	6.098826	4.037100	-2.479200
74	6	-4.121849	5.185609	-1.650767
75	8	-5.036791	4.962815	-0.798021
76	8	-4.049771	6.067753	-2.532099
77	6	-5.226834	-4.107428	-1.564544
78	8	-4.982086	-5.023524	-0.719141
79	8	-6.126336	-4.036851	-2.428021
80	59	-0.000816	0.014038	0.065037
81	1	-1.792440	-3.829101	1.551602
82	1	-3.844666	1.818645	1.537631
83	1	1.802899	3.856319	1.543677
84	1	3.848076	-1.778946	1.562846

E(RTPSSh) = -2386.855262 Hartree
Zero-point correction = 0.668001 Hartree/particle
Sum of electronic and thermal Energies = -2386.139418 Hartree
Sum of electronic and thermal Enthalpies = -2386.138473 Hartree
Sum of electronic and thermal Free Energies = -2386.273313 Hartree

Table S62. [Nd(DOTA-$(gly)_4(H_2O)]^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center	Atomic	Coordinates (Angstroms)

Number	Number	X	Y	Z
1	7	0.636359	2.061983	1.770181
2	6	1.545300	1.592297	2.856914
3	6	2.618900	0.612162	2.391926
4	7	2.046946	-0.625000	1.777473
5	6	1.575387	-1.541768	2.856778
6	6	0.595346	-2.609626	2.379251
7	7	-0.639835	-2.028075	1.770243
8	6	-1.552379	-1.558604	2.854862
9	6	-2.625073	-0.578944	2.386692
10	7	-2.049344	0.654958	1.770562
11	6	-1.582273	1.575311	2.849136
12	6	-0.600670	2.643300	2.374265
13	6	1.298199	3.092814	0.930959
14	6	2.227037	2.473688	-0.104583
15	8	1.939425	1.339467	-0.600051
16	6	3.090539	-1.276690	0.945374
17	6	2.494921	-2.199200	-0.107622
18	8	1.370593	-1.905472	-0.627829
19	6	-1.298040	-3.064142	0.933578
20	6	-2.238072	-2.454993	-0.096526
21	8	-1.952157	-1.324965	-0.605910
22	6	-3.084803	1.306178	0.928189
23	6	-2.473154	2.222637	-0.122221
24	8	-1.343315	1.927560	-0.624747
25	1	0.927079	1.126449	3.627721
26	1	2.039984	2.451014	3.332529
27	1	3.267992	1.084405	1.653083
28	1	3.251725	0.358315	3.253601
29	1	1.108384	-0.929270	3.631453
30	1	2.432697	-2.040685	3.330278
31	1	1.067074	-3.250383	1.632567
32	1	0.339459	-3.252506	3.232825
33	1	-0.936093	-1.091580	3.626430
34	1	-2.046604	-2.417939	3.329594
35	1	-3.274392	-1.051800	1.648456
36	1	-3.258298	-0.322708	3.247364
37	1	-1.118457	0.965001	3.627498
38	1	-2.442232	2.074670	3.317438
39	1	-1.070281	3.283426	1.625745
40	1	-0.348917	3.286604	3.228883
41	1	0.517014	3.606912	0.362184
42	1	3.606716	-0.488842	0.387973
43	1	-0.515306	-3.570178	0.359428
44	1	-3.600556	0.517961	0.370968
45	8	-0.030494	-0.336361	-2.449764
46	1	0.596791	-1.068661	-2.583855
47	1	-0.902148	-0.700396	-2.679350
48	7	-3.179886	3.262684	-0.508872
49	7	-3.285761	-3.161443	-0.461013
50	7	3.206057	-3.238847	-0.483345
51	7	3.271072	3.178877	-0.484295
52	1	-4.103540	3.539086	-0.121955
53	1	-3.552324	-4.087599	-0.071981
54	1	4.125793	-3.515914	-0.085835
55	1	3.540002	4.106658	-0.102514
56	6	4.215205	2.857615	-1.546970
57	1	3.694245	2.712692	-2.498669
58	6	-2.865687	4.193589	-1.585317
59	1	-2.732041	3.661193	-2.532344
60	6	-4.237975	-2.847961	-1.518967

61	1	-4.785638	-1.927789	-1.291150
62	6	2.903088	-4.173575	-1.559893
63	1	2.784952	-3.645079	-2.511254
64	1	-1.941759	4.740421	-1.371117
65	1	4.763308	1.938017	-1.317595
66	1	1.974386	-4.715819	-1.355424
67	1	-3.724186	-2.708551	-2.475432
68	6	4.111928	-5.167282	-1.626746
69	8	5.021387	-4.948387	-0.767045
70	8	4.040913	-6.049800	-2.507330
71	6	5.202210	4.070329	-1.635962
72	8	4.975855	4.997199	-0.797150
73	8	6.089078	3.985480	-2.511416
74	6	-4.075905	5.183683	-1.673334
75	8	-4.995592	4.969121	-0.823484
76	8	-3.996545	6.061018	-2.558769
77	6	-5.223066	-4.063478	-1.592824
78	8	-4.987501	-4.984154	-0.749693
79	8	-6.116377	-3.985980	-2.462016
80	60	0.000210	0.014265	0.091733
81	1	-1.809332	-3.816153	1.548580
82	1	-3.828879	1.835482	1.537664
83	1	1.820991	3.839571	1.542912
84	1	3.832571	-1.799702	1.562624

E(RTPSSh) = -2387.4782215 Hartree
Zero-point correction = 0.668374 Hartree/particle
Sum of electronic and thermal Energies = -2386.762193 Hartree
Sum of electronic and thermal Enthalpies = -2386.761249 Hartree
Sum of electronic and thermal Free Energies = -2386.894987 Hartree

Table S63. $[Sm(DOTA\text{-}(gly)_4(H_2O)]^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.641598	2.048986	1.784322
2	6	1.552417	1.584347	2.871796
3	6	2.620941	0.600150	2.405710
4	7	2.037644	-0.631387	1.791313
5	6	1.570597	-1.549009	2.871735
6	6	0.587116	-2.612222	2.392909
7	7	-0.642879	-2.020182	1.784014
8	6	-1.555929	-1.555993	2.870447
9	6	-2.624059	-0.572566	2.402074
10	7	-2.038258	0.656538	1.786529
11	6	-1.574384	1.576983	2.866344
12	6	-0.589742	2.640563	2.389715
13	6	1.306346	3.070021	0.935389
14	6	2.220720	2.434470	-0.102305
15	8	1.923899	1.293812	-0.577159
16	6	3.070345	-1.287600	0.949495
17	6	2.456731	-2.194156	-0.106111
18	8	1.326222	-1.888545	-0.606365
19	6	-1.305080	-3.046230	0.938491
20	6	-2.228944	-2.421014	-0.095602
21	8	-1.931847	-1.284738	-0.585290
22	6	-3.064857	1.312511	0.937191
23	6	-2.437488	2.213496	-0.116300
24	8	-1.303144	1.906721	-0.601166

25	1	0.935558	1.122691	3.646233
26	1	2.049424	2.444799	3.341579
27	1	3.271378	1.069252	1.666017
28	1	3.253844	0.341187	3.265652
29	1	1.107166	-0.937423	3.649305
30	1	2.429282	-2.049975	3.340372
31	1	1.056331	-3.254043	1.645499
32	1	0.326322	-3.255429	3.244604
33	1	-0.940299	-1.093172	3.645090
34	1	-2.052030	-2.417067	3.339841
35	1	-3.274441	-1.042371	1.662716
36	1	-3.257618	-0.312149	3.261102
37	1	-1.113238	0.967079	3.646649
38	1	-2.435078	2.078142	3.331153
39	1	-1.057424	3.282026	1.641077
40	1	-0.332168	3.283847	3.242432
41	1	0.526125	3.588446	0.369576
42	1	3.592361	-0.501684	0.395210
43	1	-0.523817	-3.558612	0.368331
44	1	-3.587824	0.526658	0.383768
45	8	-0.041686	-0.264728	-2.379109
46	1	0.579874	-0.997153	-2.536353
47	1	-0.919516	-0.624907	-2.591833
48	7	-3.134239	3.252670	-0.522630
49	7	-3.273879	-3.118364	-0.483431
50	7	3.157116	-3.232606	-0.503719
51	7	3.261921	3.131271	-0.504197
52	1	-4.060589	3.537658	-0.148187
53	1	-3.549613	-4.047943	-0.108351
54	1	4.080175	-3.519407	-0.120273
55	1	3.538053	4.062867	-0.136613
56	6	4.194371	2.795139	-1.572622
57	1	3.663818	2.641933	-2.517689
58	6	-2.805404	4.171502	-1.605089
59	1	-2.665623	3.629573	-2.545790
60	6	-4.212711	-2.789739	-1.548833
61	1	-4.759325	-1.869735	-1.317984
62	6	2.837090	-4.155169	-1.585885
63	1	2.711663	-3.617109	-2.530896
64	1	-1.881228	4.716680	-1.387794
65	1	4.742320	1.876548	-1.338957
66	1	1.908050	-4.694964	-1.376631
67	1	-3.687884	-2.643425	-2.498232
68	6	4.040513	-5.154119	-1.674406
69	8	4.959570	-4.947272	-0.821849
70	8	3.956934	-6.028417	-2.561961
71	6	5.183897	4.004254	-1.683722
72	8	4.967714	4.940350	-0.852444
73	8	6.062468	3.908415	-2.566331
74	6	-4.010737	5.165892	-1.713361
75	8	-4.938954	4.962703	-0.869952
76	8	-3.920227	6.035034	-2.605750
77	6	-5.201735	-4.000791	-1.644823
78	8	-4.978896	-4.929789	-0.807283
79	8	-6.085286	-3.912450	-2.522793
80	62	0.001496	0.014439	0.137504
81	1	-1.830123	-3.794692	1.546031
82	1	-3.804365	1.854749	1.540648
83	1	1.841086	3.814471	1.539570
84	1	3.809240	-1.824161	1.558675

E(RTPSSh) = -2388.7008851 Hartree

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 Zero-point correction = 0.668914 Hartree/particle
 Sum of electronic and thermal Energies = -2387.984561 Hartree
 Sum of electronic and thermal Enthalpies = -2387.983617 Hartree
 Sum of electronic and thermal Free Energies = -2388.116192 Hartree
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Table S64. [Eu(DOTA-(gly)$_4$(H$_2$O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.604309	2.055480	1.793150
2	6	1.523889	1.610713	2.881564
3	6	2.608894	0.645531	2.415128
4	7	2.044446	-0.594774	1.801448
5	6	1.595639	-1.520447	2.882629
6	6	0.631860	-2.600529	2.402546
7	7	-0.606835	-2.027925	1.793193
8	6	-1.528854	-1.582795	2.879988
9	6	-2.613382	-0.618713	2.410385
10	7	-2.046453	0.619354	1.795707
11	6	-1.601127	1.547965	2.876287
12	6	-0.635802	2.628162	2.398491
13	6	1.251538	3.084312	0.940310
14	6	2.168848	2.458583	-0.100441
15	8	1.889599	1.308688	-0.563618
16	6	3.085310	-1.234435	0.957386
17	6	2.481456	-2.143690	-0.101070
18	8	1.341959	-1.854471	-0.590679
19	6	-1.251112	-3.062602	0.944758
20	6	-2.176917	-2.448875	-0.093998
21	8	-1.897014	-1.303305	-0.572675
22	6	-3.081276	1.258688	0.944007
23	6	-2.463396	2.162917	-0.111830
24	8	-1.320658	1.872032	-0.586630
25	1	0.916171	1.139238	3.657335
26	1	2.005044	2.481403	3.348893
27	1	3.250253	1.125582	1.674524
28	1	3.247132	0.396981	3.274114
29	1	1.121195	-0.917654	3.660449
30	1	2.463785	-2.005425	3.350584
31	1	1.112770	-3.233205	1.654766
32	1	0.381044	-3.249070	3.253093
33	1	-0.922577	-1.109586	3.655756
34	1	-2.009275	-2.453659	3.347566
35	1	-3.254011	-1.099642	1.669647
36	1	-3.252953	-0.368765	3.267974
37	1	-1.129357	0.946894	3.657106
38	1	-2.471378	2.033307	3.339994
39	1	-1.114766	3.260610	1.649335
40	1	-0.388158	3.276697	3.250058
41	1	0.462380	3.592075	0.377532
42	1	3.597923	-0.440409	0.406175
43	1	-0.460741	-3.565082	0.378658
44	1	-3.594447	0.464661	0.393369
45	8	-0.041188	-0.240392	-2.345816
46	1	0.598376	-0.955682	-2.509562
47	1	-0.911472	-0.626299	-2.545247
48	7	-3.174987	3.186977	-0.530399
49	7	-3.203883	-3.163390	-0.497525
50	7	3.197344	-3.166252	-0.511763

51	7	3.192974	3.171673	-0.517063
52	1	-4.108221	3.459209	-0.163479
53	1	-3.466608	-4.100089	-0.130529
54	1	4.127702	-3.439829	-0.136236
55	1	3.455690	4.110495	-0.157983
56	6	4.123673	2.843901	-1.589623
57	1	3.589599	2.677859	-2.530565
58	6	-2.854129	4.104746	-1.616180
59	1	-2.702008	3.560005	-2.553361
60	6	-4.139621	-2.844570	-1.568659
61	1	-4.701559	-1.933789	-1.338466
62	6	2.885924	-4.087187	-1.597859
63	1	2.747955	-3.545642	-2.539150
64	1	-1.939109	4.664486	-1.397432
65	1	4.686796	1.934926	-1.354755
66	1	1.966245	-4.642234	-1.387474
67	1	-3.610074	-2.686968	-2.513672
68	6	4.103827	-5.067297	-1.698277
69	8	5.025377	-4.849618	-0.851155
70	8	4.027482	-5.939157	-2.588856
71	6	5.094210	4.067075	-1.713528
72	8	4.867227	5.005799	-0.888129
73	8	5.970418	3.978025	-2.599176
74	6	-4.073102	5.081032	-1.735719
75	8	-5.003630	4.867375	-0.897474
76	8	-3.989372	5.948073	-2.630789
77	6	-5.109533	-4.070001	-1.676778
78	8	-4.876259	-5.000676	-0.843956
79	8	-5.989985	-3.989660	-2.558564
80	63	0.000840	0.014857	0.159795
81	1	-1.769854	-3.817705	1.549423
82	1	-3.827635	1.794106	1.545046
83	1	1.779561	3.836229	1.541062
84	1	3.830682	-1.764636	1.564184

E(RTPSSh) = -2389.2980601 Hartree
Zero-point correction = 0.669019 Hartree/particle
Sum of electronic and thermal Energies = -2388.581684 Hartree
Sum of electronic and thermal Enthalpies = -2388.580739 Hartree
Sum of electronic and thermal Free Energies = -2388.713252 Hartree

Table S65. [Gd(DOTA-$(gly)_4(H_2O)]^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.565235	2.062205	1.800227
2	6	1.493808	1.637795	2.889083
3	6	2.595647	0.692523	2.422344
4	7	2.051251	-0.556514	1.808808
5	6	1.621722	-1.490400	2.890654
6	6	0.678418	-2.587713	2.409842
7	7	-0.569145	-2.035723	1.800051
8	6	-1.500016	-1.610688	2.887267
9	6	-2.601337	-0.666541	2.417212
10	7	-2.054481	0.580513	1.803052
11	6	-1.628625	1.517096	2.884470
12	6	-0.683581	2.614508	2.406181
13	6	1.193553	3.099421	0.943454
14	6	2.116448	2.484702	-0.098649

15	8	1.856093	1.326742	-0.552651
16	6	3.100589	-1.178268	0.962093
17	6	2.507708	-2.092717	-0.097722
18	8	1.359915	-1.821883	-0.578654
19	6	-1.194442	-3.079045	0.948250
20	6	-2.125359	-2.477036	-0.092433
21	8	-1.864418	-1.322991	-0.561757
22	6	-3.097710	1.202125	0.948649
23	6	-2.490380	2.111761	-0.108336
24	8	-1.339769	1.838866	-0.574652
25	1	0.895706	1.156277	3.666175
26	1	1.958551	2.518482	3.354219
27	1	3.228041	1.183929	1.681510
28	1	3.238723	0.454571	3.280644
29	1	1.136368	-0.896762	3.668799
30	1	2.499393	-1.958669	3.357790
31	1	1.171134	-3.211142	1.662017
32	1	0.438294	-3.241245	3.259568
33	1	-0.903312	-1.127326	3.664212
34	1	-1.964076	-2.491372	3.352912
35	1	-3.232633	-1.158720	1.675883
36	1	-3.246077	-0.427478	3.273954
37	1	-1.146089	0.925017	3.665596
38	1	-2.508435	1.985676	3.347307
39	1	-1.174233	3.238164	1.657258
40	1	-0.446296	3.267630	3.257094
41	1	0.395046	3.593903	0.382217
42	1	3.601192	-0.375624	0.412481
43	1	-0.394838	-3.568668	0.384007
44	1	-3.598885	0.399521	0.399572
45	8	-0.037165	-0.222221	-2.318847
46	1	0.615808	-0.925303	-2.482925
47	1	-0.900405	-0.626491	-2.513144
48	7	-3.217281	3.120989	-0.536482
49	7	-3.135832	-3.208138	-0.507341
50	7	3.239476	-3.099747	-0.518434
51	7	3.124934	3.213647	-0.525755
52	1	-4.157524	3.378246	-0.176735
53	1	-3.382728	-4.152378	-0.148504
54	1	4.177147	-3.358119	-0.150148
55	1	3.371939	4.159746	-0.174700
56	6	4.056290	2.895319	-1.600624
57	1	3.520914	2.713790	-2.537969
58	6	-2.905549	4.039582	-1.624264
59	1	-2.738695	3.493709	-2.558279
60	6	-4.071586	-2.899731	-1.581572
61	1	-4.651322	-2.000871	-1.348843
62	6	2.937949	-4.021406	-1.606765
63	1	2.785673	-3.478538	-2.545086
64	1	-2.001542	4.615761	-1.402516
65	1	4.636488	1.998015	-1.362572
66	1	2.029223	-4.593190	-1.393765
67	1	-3.540115	-2.726789	-2.522816
68	6	4.171968	-4.979967	-1.718159
69	8	5.095143	-4.748869	-0.876427
70	8	4.104563	-5.850184	-2.611009
71	6	5.004569	4.134454	-1.737265
72	8	4.763357	5.075640	-0.918799
73	8	5.879494	4.053524	-2.624941
74	6	-4.140016	4.994647	-1.755309
75	8	-5.072708	4.767732	-0.923045
76	8	-4.064399	5.860140	-2.652586

77	6	-5.018309	-4.141995	-1.702490
78	8	-4.770724	-5.074476	-0.875892
79	8	-5.896834	-4.070963	-2.586935
80	64	0.000273	0.015155	0.177742
81	1	-1.703979	-3.842710	1.549902
82	1	-3.852259	1.728667	1.547224
83	1	1.712206	3.860353	1.540917
84	1	3.854082	-1.699614	1.566448

E(RTPSSh) = -2389.8881476 Hartree
Zero-point correction = 0.669060 Hartree/particle
Sum of electronic and thermal Energies = -2389.171748 Hartree
Sum of electronic and thermal Enthalpies = -2389.170804 Hartree
Sum of electronic and thermal Free Energies = -2389.303648 Hartree

Table S66. [Tb(DOTA-(gly)$_4$(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-0.532165	-2.065134	1.808101
2	6	-1.466916	-1.657363	2.898053
3	6	-2.582797	-0.729119	2.431570
4	7	-2.054962	0.526274	1.816978
5	6	-1.642130	1.467340	2.899029
6	6	-0.715693	2.578053	2.417209
7	7	0.538173	2.041834	1.806758
8	6	1.475832	1.633708	2.894588
9	6	2.590904	0.706517	2.424264
10	7	2.060262	-0.547236	1.809998
11	6	1.651918	-1.490440	2.892350
12	6	0.723370	-2.601434	2.414170
13	6	-1.146109	-3.108735	0.948669
14	6	-2.073141	-2.501625	-0.093725
15	8	-1.827932	-1.336814	-0.538363
16	6	-3.110858	1.132653	0.967408
17	6	-2.526110	2.049364	-0.094432
18	8	-1.371631	1.792189	-0.567083
19	6	1.148744	3.091141	0.951772
20	6	2.082369	2.496369	-0.090114
21	8	1.834943	1.335599	-0.550134
22	6	3.109452	-1.153984	0.952304
23	6	2.509116	-2.067144	-0.105106
24	8	1.351974	-1.807945	-0.562967
25	1	-0.876070	-1.167122	3.675312
26	1	-1.917224	-2.545910	3.362334
27	1	-3.208647	-1.230174	1.691652
28	1	-3.228840	-0.499237	3.289763
29	1	-1.148277	0.881762	3.677993
30	1	-2.527644	1.921851	3.364848
31	1	-1.217965	3.194162	1.669665
32	1	-0.483611	3.235518	3.266068
33	1	0.886945	1.141848	3.672226
34	1	1.925797	2.522332	3.358898
35	1	3.214999	1.208118	1.683154
36	1	3.239404	0.475969	3.280436
37	1	1.161481	-0.906005	3.674367
38	1	2.539879	-1.945058	3.353430
39	1	1.223085	-3.218151	1.665491
40	1	0.494599	-3.258054	3.264638

41	1	-0.340947	-3.593552	0.388662
42	1	-3.602149	0.322757	0.420174
43	1	0.342329	3.571549	0.389463
44	1	3.600660	-0.344500	0.404475
45	8	0.030348	0.200195	-2.287880
46	1	-0.624489	0.901214	-2.453054
47	1	0.891735	0.604671	-2.489076
48	7	3.246219	-3.065193	-0.541833
49	7	3.080966	3.237917	-0.514944
50	7	-3.269237	3.043726	-0.524960
51	7	-3.069523	-3.241661	-0.530042
52	1	4.192915	-3.310104	-0.190597
53	1	3.315684	4.188599	-0.165067
54	1	-4.213249	3.289813	-0.164499
55	1	-3.302940	-4.194533	-0.188303
56	6	-4.000101	-2.928922	-1.607248
57	1	-3.462664	-2.729934	-2.539831
58	6	2.939622	-3.982711	-1.632012
59	1	2.756259	-3.434533	-2.561534
60	6	4.014906	2.934916	-1.592323
61	1	4.609850	2.047056	-1.355766
62	6	-2.974860	3.962906	-1.617373
63	1	-2.807286	3.416434	-2.550961
64	1	2.046613	-4.574151	-1.405833
65	1	-4.596374	-2.043511	-1.364356
66	1	-2.077030	4.550775	-1.401946
67	1	3.480533	2.745902	-2.528794
68	6	-4.223258	4.900719	-1.743677
69	8	-5.148784	4.659804	-0.907322
70	8	-4.163159	5.766512	-2.641330
71	6	-4.927120	-4.182043	-1.760472
72	8	-4.673954	-5.127268	-0.950357
73	8	-5.799318	-4.106613	-2.651335
74	6	4.187436	-4.917890	-1.778675
75	8	5.123817	-4.680781	-0.953459
76	8	4.117144	-5.780043	-2.679609
77	6	4.941763	4.190401	-1.729368
78	8	4.684124	5.126434	-0.909909
79	8	5.816693	4.124824	-2.617788
80	65	0.000115	-0.014821	0.196065
81	1	1.652377	3.860971	1.550491
82	1	3.870894	-1.673927	1.547866
83	1	-1.657861	-3.876155	1.543730
84	1	-3.870531	1.648157	1.568979

E(RTPSSh) = -2390.4784054 Hartree
Zero-point correction = 0.669136 Hartree/particle
Sum of electronic and thermal Energies = -2389.761947 Hartree
Sum of electronic and thermal Enthalpies = -2389.761003 Hartree
Sum of electronic and thermal Free Energies = -2389.894084 Hartree

Table S67. [Dy(DOTA-$(gly)_4(H_2O)]^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.507766	2.066079	1.815528
2	6	1.447456	1.671507	2.906039
3	6	2.573361	0.756094	2.439263
4	7	2.057368	-0.503587	1.823861

5	6	1.657649	-1.450087	2.905940
6	6	0.744118	-2.570591	2.423055
7	7	-0.514218	-2.045949	1.812213
8	6	-1.457104	-1.651884	2.900593
9	6	-2.582085	-0.737263	2.430287
10	7	-2.063286	0.521478	1.816599
11	6	-1.668259	1.469003	2.900001
12	6	-0.752502	2.590261	2.422021
13	6	1.110269	3.113818	0.953092
14	6	2.039632	2.511460	-0.089631
15	8	1.805487	1.341185	-0.525725
16	6	3.117091	-1.098222	0.970985
17	6	2.536824	-2.015928	-0.092057
18	8	1.376648	-1.768833	-0.556307
19	6	-1.113124	-3.098644	0.953332
20	6	-2.047975	-2.507986	-0.089375
21	8	-1.810128	-1.341746	-0.540804
22	6	-3.116328	1.117369	0.956199
23	6	-2.520223	2.032805	-0.101234
24	8	-1.357642	1.783926	-0.550921
25	1	0.862599	1.175126	3.683981
26	1	1.887033	2.566064	3.368982
27	1	3.194065	1.264335	1.699919
28	1	3.221817	0.531890	3.297089
29	1	1.157533	-0.870919	3.685749
30	1	2.549105	-1.894153	3.370429
31	1	1.253540	-3.180985	1.675670
32	1	0.517943	-3.231124	3.271075
33	1	-0.874522	-1.154465	3.679489
34	1	-1.896503	-2.546837	3.362802
35	1	-3.200569	-1.245469	1.688962
36	1	-3.233501	-0.513341	3.285951
37	1	-1.171705	0.890176	3.682375
38	1	-2.562239	1.912695	3.360022
39	1	-1.259129	3.201912	1.673842
40	1	-0.529866	3.249157	3.272290
41	1	0.299826	3.590726	0.394007
42	1	3.600883	-0.282849	0.425292
43	1	-0.301311	-3.571140	0.392195
44	1	-3.600019	0.302982	0.409046
45	8	-0.026347	-0.179761	-2.262552
46	1	0.632938	-0.876156	-2.429689
47	1	-0.885215	-0.589914	-2.463224
48	7	-3.264620	3.021737	-0.546065
49	7	-3.037196	-3.256533	-0.523375
50	7	3.287665	-3.000245	-0.531856
51	7	3.026317	3.259241	-0.534537
52	1	-4.216709	3.256866	-0.202742
53	1	-3.263629	-4.211932	-0.180722
54	1	4.236826	-3.237248	-0.178702
55	1	3.249735	4.217334	-0.200781
56	6	3.955615	2.949929	-1.613882
57	1	3.416264	2.737494	-2.542378
58	6	-2.961248	3.937848	-1.638374
59	1	-2.764045	3.387613	-2.563823
60	6	-3.969682	-2.956367	-1.602858
61	1	-4.574517	-2.075961	-1.363606
62	6	2.997244	-3.917161	-1.627280
63	1	2.817591	-3.367930	-2.556994
64	1	-2.077485	4.541594	-1.408400
65	1	4.563858	2.073703	-1.367381
66	1	2.107820	-4.516805	-1.409487

67	1	-3.433455	-2.756619	-2.536033
68	6	4.255882	-4.839465	-1.766208
69	8	5.183986	-4.591368	-0.934846
70	8	4.200186	-5.701642	-2.667577
71	6	4.866460	4.213130	-1.780931
72	8	4.604900	5.161675	-0.977399
73	8	5.735871	4.141268	-2.674800
74	6	-4.219533	4.856566	-1.798943
75	8	-5.159361	4.611102	-0.980128
76	8	-4.152974	5.715575	-2.703148
77	6	-4.883164	-4.220380	-1.751187
78	8	-4.618253	-5.159161	-0.937210
79	8	-5.755883	-4.157890	-2.641960
80	66	-0.000018	0.014903	0.211512
81	1	-1.612636	-3.873893	1.548477
82	1	-3.882964	1.632640	1.549129
83	1	1.617210	3.886533	1.545378
84	1	3.881896	-1.609808	1.569368

E(RTPSSh) = -2391.0648857 Hartree
Zero-point correction = 0.669280 Hartree/particle
Sum of electronic and thermal Energies = -2390.348330 Hartree
Sum of electronic and thermal Enthalpies = -2390.347386 Hartree
Sum of electronic and thermal Free Energies = -2390.480645 Hartree

Table S68. [Ho(DOTA-$(gly)_4(H_2O)]^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.486760	2.065896	1.822604
2	6	1.430503	1.682961	2.913589
3	6	2.564852	0.778565	2.446583
4	7	2.058680	-0.484480	1.830376
5	6	1.670946	-1.435856	2.912263
6	6	0.768246	-2.564347	2.428390
7	7	-0.493584	-2.049027	1.817268
8	6	-1.440959	-1.667599	2.906030
9	6	-2.574211	-0.763688	2.435760
10	7	-2.065202	0.499108	1.822670
11	6	-1.682400	1.450427	2.906902
12	6	-0.777357	2.580159	2.429230
13	6	1.079668	3.116709	0.957261
14	6	2.010549	2.517679	-0.085636
15	8	1.785854	1.342504	-0.513305
16	6	3.121106	-1.068756	0.973883
17	6	2.543904	-1.986961	-0.089966
18	8	1.378546	-1.748627	-0.545925
19	6	-1.082491	-3.103951	0.954355
20	6	-2.017881	-2.515865	-0.088866
21	8	-1.788033	-1.344745	-0.531898
22	6	-3.120947	1.085498	0.959171
23	6	-2.527661	2.002549	-0.098006
24	8	-1.360150	1.762526	-0.539592
25	1	0.850943	1.181268	3.692107
26	1	1.860746	2.582596	3.375419
27	1	3.181173	1.292864	1.707778
28	1	3.215279	0.559066	3.304078
29	1	1.165848	-0.862630	3.693265
30	1	2.567605	-1.871090	3.375068

31	1	1.283574	-3.169833	1.681072
32	1	0.546941	-3.227505	3.275595
33	1	-0.864089	-1.165574	3.686241
34	1	-1.871253	-2.568016	3.366171
35	1	-3.187842	-1.277379	1.694175
36	1	-3.228107	-0.545403	3.290934
37	1	-1.181021	0.876757	3.690010
38	1	-2.581668	1.884809	3.365466
39	1	-1.289587	3.187435	1.681313
40	1	-0.559964	3.240993	3.279284
41	1	0.264785	3.586865	0.398987
42	1	3.597869	-0.248508	0.429376
43	1	-0.266006	-3.569363	0.394156
44	1	-3.597682	0.266782	0.412439
45	8	-0.022329	-0.160842	-2.239053
46	1	0.639067	-0.855014	-2.407203
47	1	-0.879815	-0.573997	-2.439604
48	7	-3.277754	2.983443	-0.550836
49	7	-2.999038	-3.269638	-0.531723
50	7	3.300876	-2.962348	-0.538806
51	7	2.988587	3.271573	-0.539106
52	1	-4.234488	3.210536	-0.215064
53	1	-3.219140	-4.228978	-0.195831
54	1	4.254621	-3.191731	-0.192870
55	1	3.203983	4.234027	-0.212719
56	6	3.916377	2.964029	-1.620317
57	1	3.375258	2.739929	-2.545021
58	6	-2.975905	3.898084	-1.644863
59	1	-2.766541	3.346032	-2.566533
60	6	-3.929973	-2.970406	-1.612872
61	1	-4.542268	-2.095918	-1.370911
62	6	3.012689	-3.876865	-1.636880
63	1	2.821861	-3.325043	-2.562821
64	1	-2.099970	4.511762	-1.411210
65	1	4.534324	2.095561	-1.370467
66	1	2.130690	-4.486460	-1.416520
67	1	-3.392017	-2.761504	-2.543041
68	6	4.279665	-4.785788	-1.788077
69	8	5.210581	-4.531869	-0.961621
70	8	4.226814	-5.644528	-2.692863
71	6	4.813866	4.235045	-1.799838
72	8	4.545515	5.186729	-1.002287
73	8	5.680803	4.165403	-2.696277
74	6	-4.242312	4.803263	-1.818225
75	8	-5.185449	4.551494	-1.005157
76	8	-4.178008	5.659269	-2.725428
77	6	-4.833240	-4.240559	-1.771542
78	8	-4.562866	-5.182456	-0.962969
79	8	-5.704247	-4.179215	-2.664032
80	67	0.000161	0.014988	0.226151
81	1	-1.578689	-3.884025	1.545857
82	1	-3.892379	1.596777	1.549232
83	1	1.582740	3.893885	1.546895
84	1	3.890697	-1.576970	1.568902

E(RTPSSh) = -2391.6490573 Hartree
Zero-point correction = 0.669478 Hartree/particle
Sum of electronic and thermal Energies = -2390.932377 Hartree
Sum of electronic and thermal Enthalpies = -2390.931433 Hartree
Sum of electronic and thermal Free Energies = -2391.064576 Hartree

Table S69. [Er(DOTA-(gly)$_4$(H$_2$O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.474576	2.064306	1.828349
2	6	1.420770	1.689301	2.919769
3	6	2.559605	0.791030	2.452649
4	7	2.057847	-0.473369	1.836117
5	6	1.678117	-1.427476	2.918158
6	6	0.781517	-2.560233	2.433721
7	7	-0.481691	-2.049123	1.822402
8	6	-1.431572	-1.675875	2.911580
9	6	-2.569292	-0.778109	2.441214
10	7	-2.064844	0.486346	1.828297
11	6	-1.690024	1.440017	2.912993
12	6	-0.791044	2.574177	2.435122
13	6	1.062206	3.115577	0.959853
14	6	1.992147	2.515859	-0.083085
15	8	1.771938	1.336878	-0.502447
16	6	3.120543	-1.052116	0.976257
17	6	2.542633	-1.968755	-0.088132
18	8	1.373128	-1.734798	-0.535913
19	6	-1.065245	-3.104238	0.956122
20	6	-1.998869	-2.515386	-0.087798
21	8	-1.772734	-1.340448	-0.522799
22	6	-3.121077	1.067432	0.961824
23	6	-2.527140	1.983389	-0.095500
24	8	-1.355821	1.747609	-0.529173
25	1	0.844624	1.185029	3.699184
26	1	1.845612	2.592245	3.380112
27	1	3.173589	1.308487	1.714101
28	1	3.211172	0.573767	3.309788
29	1	1.170524	-0.857926	3.700255
30	1	2.578092	-1.857540	3.379341
31	1	1.300019	-3.163116	1.686490
32	1	0.562453	-3.224859	3.280300
33	1	-0.858242	-1.171402	3.692852
34	1	-1.856498	-2.579677	3.370061
35	1	-3.180297	-1.294826	1.699544
36	1	-3.224612	-0.562465	3.295908
37	1	-1.186141	0.869751	3.697004
38	1	-2.592582	1.869082	3.370080
39	1	-1.306343	3.178989	1.687314
40	1	-0.575972	3.236275	3.284714
41	1	0.244863	3.582218	0.402314
42	1	3.593912	-0.229277	0.432773
43	1	-0.246224	-3.566474	0.397082
44	1	-3.594483	0.246292	0.415907
45	8	-0.020265	-0.144625	-2.217689
46	1	0.640889	-0.838827	-2.386688
47	1	-0.877875	-0.557867	-2.417597
48	7	-3.279055	2.959129	-0.556221
49	7	-2.974867	-3.270675	-0.539072
50	7	3.301616	-2.938494	-0.545471
51	7	2.965075	3.271427	-0.544536
52	1	-4.239031	3.182462	-0.227246
53	1	-3.192277	-4.232879	-0.209527
54	1	4.258720	-3.164276	-0.206344
55	1	3.177033	4.236885	-0.224843
56	6	3.890958	2.961403	-1.626727

57	1	3.348007	2.729423	-2.548399
58	6	-2.975167	3.871377	-1.651731
59	1	-2.757074	3.317066	-2.570000
60	6	-3.903389	-2.969078	-1.621699
61	1	-4.519422	-2.097892	-1.377294
62	6	3.011590	-3.850316	-1.645364
63	1	2.812579	-3.296012	-2.568089
64	1	-2.103939	4.490531	-1.414874
65	1	4.513352	2.097052	-1.373588
66	1	2.134025	-4.465284	-1.422209
67	1	-3.363228	-2.753706	-2.549110
68	6	4.282410	-4.752020	-1.807425
69	8	5.216973	-4.495781	-0.985797
70	8	4.228633	-5.607955	-2.714802
71	6	4.782443	4.235185	-1.816666
72	8	4.512272	5.190631	-1.024244
73	8	5.647080	4.163830	-2.715188
74	6	-4.245263	4.769184	-1.836398
75	8	-5.192353	4.515026	-1.028689
76	8	-4.179624	5.622592	-2.745950
77	6	-4.801655	-4.241651	-1.789677
78	8	-4.530068	-5.186841	-0.985361
79	8	-5.670345	-4.178810	-2.684294
80	68	0.000170	0.015217	0.238934
81	1	-1.561423	-3.886596	1.544528
82	1	-3.894918	1.578140	1.549128
83	1	1.564641	3.895201	1.546692
84	1	3.892624	-1.560077	1.568175

E(RTPSSh) = -2392.2322841 Hartree
Zero-point correction = 0.669681 Hartree/particle
Sum of electronic and thermal Energies = -2391.515490 Hartree
Sum of electronic and thermal Enthalpies = -2391.514546 Hartree
Sum of electronic and thermal Free Energies = -2391.647366 Hartree

Table S70. [Tm(DOTA-$(gly)_4(H_2O)]^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.462736	2.062804	1.833815
2	6	1.411567	1.695193	2.925467
3	6	2.554695	0.803116	2.457747
4	7	2.057592	-0.462800	1.840872
5	6	1.685686	-1.419776	2.923043
6	6	0.795155	-2.556638	2.437821
7	7	-0.469661	-2.049960	1.826456
8	6	-1.422312	-1.685239	2.916071
9	6	-2.564205	-0.793220	2.445697
10	7	-2.064418	0.473187	1.833312
11	6	-1.697432	1.428955	2.918788
12	6	-0.804615	2.567643	2.440922
13	6	1.044744	3.115166	0.962883
14	6	1.973808	2.515747	-0.080665
15	8	1.758332	1.333448	-0.492824
16	6	3.120822	-1.035804	0.977945
17	6	2.542703	-1.951004	-0.087208
18	8	1.369547	-1.721238	-0.527706
19	6	-1.047406	-3.105246	0.956617
20	6	-1.979965	-2.515830	-0.087542

21	8	-1.758178	-1.337198	-0.514727
22	6	-3.121268	1.049064	0.964216
23	6	-2.527184	1.964369	-0.093228
24	8	-1.352438	1.733191	-0.519927
25	1	0.838749	1.188298	3.705643
26	1	1.831161	2.601248	3.384414
27	1	3.165871	1.323896	1.719204
28	1	3.207801	0.588200	3.314262
29	1	1.175718	-0.853797	3.706197
30	1	2.588954	-1.844711	3.382465
31	1	1.316897	-3.156521	1.690419
32	1	0.578578	-3.223084	3.283567
33	1	-0.852485	-1.178994	3.698775
34	1	-1.842129	-2.592601	3.372153
35	1	-3.172374	-1.312761	1.703653
36	1	-3.221144	-0.580505	3.299839
37	1	-1.191102	0.861703	3.703424
38	1	-2.603255	1.852519	3.374467
39	1	-1.323145	3.169858	1.693239
40	1	-0.592120	3.231012	3.290118
41	1	0.224793	3.578616	0.406588
42	1	3.590868	-0.210270	0.435748
43	1	-0.225674	-3.563616	0.398446
44	1	-3.591363	0.225563	0.419050
45	8	-0.017889	-0.131025	-2.199442
46	1	0.641170	-0.827018	-2.369188
47	1	-0.876292	-0.541777	-2.400849
48	7	-3.281259	2.934934	-0.561178
49	7	-2.950655	-3.272953	-0.546900
50	7	3.303793	-2.915123	-0.552580
51	7	2.941158	3.273728	-0.549743
52	1	-4.243899	3.154906	-0.237796
53	1	-3.165492	-4.237657	-0.222993
54	1	4.263526	-3.138097	-0.218972
55	1	3.149699	4.241668	-0.235440
56	6	3.864926	2.962494	-1.633412
57	1	3.320017	2.723736	-2.552187
58	6	-2.975717	3.845295	-1.657816
59	1	-2.750811	3.289226	-2.573373
60	6	-3.877492	-2.968851	-1.630291
61	1	-4.496481	-2.100477	-1.383370
62	6	3.012073	-3.824575	-1.654013
63	1	2.807129	-3.268148	-2.574169
64	1	-2.108246	4.468771	-1.418517
65	1	4.491913	2.102091	-1.378163
66	1	2.137699	-4.443372	-1.428951
67	1	-3.335786	-2.747832	-2.555485
68	6	4.285601	-4.721022	-1.824456
69	8	5.223092	-4.463143	-1.006702
70	8	4.230886	-5.574911	-2.733667
71	6	4.750160	4.239259	-1.832404
72	8	4.478234	5.197349	-1.043783
73	8	5.612243	4.167401	-2.733345
74	6	-4.248731	4.737068	-1.851371
75	8	-5.198805	4.480764	-1.047853
76	8	-4.182258	5.588327	-2.762873
77	6	-4.771571	-4.243309	-1.806221
78	8	-4.498201	-5.191684	-1.006266
79	8	-5.638991	-4.178725	-2.701933
80	69	0.000523	0.015269	0.250596
81	1	-1.543039	-3.890406	1.541767
82	1	-3.897500	1.559045	1.549013

83	1	1.546554	3.896940	1.547412
84	1	3.895318	-1.543495	1.566958

E(RTPSSh) = -2392.8121598 Hartree
Zero-point correction = 0.669890 Hartree/particle
Sum of electronic and thermal Energies = -2392.095245 Hartree
Sum of electronic and thermal Enthalpies = -2392.094301 Hartree
Sum of electronic and thermal Free Energies = -2392.226685 Hartree

Table S71. [Yb(DOTA-(gly)$_4$(H$_2$O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.450005	2.061915	1.839164
2	6	1.401813	1.702374	2.930705
3	6	2.549706	0.817261	2.462175
4	7	2.057872	-0.450552	1.845420
5	6	1.694239	-1.410116	2.927856
6	6	0.810815	-2.551914	2.442019
7	7	-0.455933	-2.050717	1.830583
8	6	-1.411388	-1.694971	2.920504
9	6	-2.558142	-0.809623	2.450141
10	7	-2.063903	0.459097	1.838542
11	6	-1.704813	1.416711	2.924768
12	6	-0.819226	2.560718	2.446865
13	6	1.025490	3.115771	0.965812
14	6	1.953960	2.517317	-0.078445
15	8	1.743831	1.331583	-0.483229
16	6	3.121841	-1.017627	0.979633
17	6	2.543778	-1.932016	-0.085827
18	8	1.366910	-1.706925	-0.518964
19	6	-1.027176	-3.106795	0.957559
20	6	-1.959472	-2.517755	-0.086595
21	8	-1.743051	-1.335238	-0.505897
22	6	-3.122001	1.029549	0.967497
23	6	-2.528474	1.944840	-0.089866
24	8	-1.350121	1.718901	-0.509291
25	1	0.833055	1.192413	3.711855
26	1	1.815700	2.611740	3.388243
27	1	3.157497	1.341596	1.723359
28	1	3.204677	0.605287	3.317951
29	1	1.181100	-0.848025	3.711746
30	1	2.600900	-1.829070	3.386060
31	1	1.336221	-3.148527	1.694563
32	1	0.597248	-3.220126	3.287086
33	1	-0.845536	-1.186268	3.704504
34	1	-1.825396	-2.606009	3.374537
35	1	-3.163087	-1.332273	1.707641
36	1	-3.216764	-0.600457	3.303816
37	1	-1.195088	0.852773	3.709608
38	1	-2.613857	1.834005	3.379799
39	1	-1.341547	3.160114	1.699559
40	1	-0.609636	3.225270	3.295808
41	1	0.202569	3.575367	0.410802
42	1	3.588122	-0.189440	0.438325
43	1	-0.202520	-3.560562	0.400013
44	1	-3.588545	0.203701	0.422920
45	8	-0.016920	-0.118787	-2.181834
46	1	0.642123	-0.814751	-2.351944

47	1	-0.875336	-0.530328	-2.381749
48	7	-3.285195	2.909748	-0.564992
49	7	-2.924174	-3.277542	-0.553822
50	7	3.307119	-2.890491	-0.558841
51	7	2.915146	3.278244	-0.555234
52	1	-4.250826	3.125395	-0.247514
53	1	-3.134827	-4.245276	-0.235992
54	1	4.269689	-3.110080	-0.230925
55	1	3.119309	4.249000	-0.246684
56	6	3.836818	2.966843	-1.640690
57	1	3.290022	2.719957	-2.556171
58	6	-2.978915	3.818533	-1.662793
59	1	-2.745844	3.260970	-2.575378
60	6	-3.849616	-2.972603	-1.638233
61	1	-4.473851	-2.108783	-1.388577
62	6	3.014188	-3.798084	-1.661554
63	1	2.802736	-3.239966	-2.579211
64	1	-2.116378	4.447774	-1.420730
65	1	4.470012	2.111643	-1.383238
66	1	2.143518	-4.421292	-1.434301
67	1	-3.306595	-2.744124	-2.560841
68	6	4.290933	-4.688399	-1.840367
69	8	5.231342	-4.427910	-1.026760
70	8	4.235673	-5.540552	-2.751139
71	6	4.713613	4.247877	-1.849532
72	8	4.439149	5.208162	-1.064439
73	8	5.572470	4.176829	-2.753605
74	6	-4.256032	4.702333	-1.865902
75	8	-5.208793	4.443090	-1.066489
76	8	-4.189793	5.551033	-2.779791
77	6	-4.736394	-4.250954	-1.823098
78	8	-4.459679	-5.202045	-1.027499
79	8	-5.602055	-4.186402	-2.720479
80	70	0.000806	0.015604	0.261071
81	1	-1.521184	-3.895126	1.539728
82	1	-3.900659	1.537998	1.550316
83	1	1.526013	3.900071	1.547972
84	1	3.899029	-1.524281	1.565902

E(RTPSSh) = -2393.3956217 Hartree
Zero-point correction = 0.670108 Hartree/particle
Sum of electronic and thermal Energies = -2392.678578 Hartree
Sum of electronic and thermal Enthalpies = -2392.677634 Hartree
Sum of electronic and thermal Free Energies = -2392.809527 Hartree

Table S72. [Lu(DOTA-(gly)$_4$(H_2O)]$^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	0.425905	2.064065	1.842674
2	6	1.382555	1.718367	2.934361
3	6	2.539977	0.846342	2.465576
4	7	2.060512	-0.426120	1.849041
5	6	1.710322	-1.389867	2.932022
6	6	0.840412	-2.541548	2.446012
7	7	-0.431134	-2.053282	1.834459
8	6	-1.390796	-1.711112	2.924876
9	6	-2.547352	-0.839098	2.454554
10	7	-2.066013	0.434364	1.842922

11	6	-1.719870	1.396228	2.929462
12	6	-0.847863	2.550045	2.450994
13	6	0.988880	3.122491	0.966703
14	6	1.920799	2.530443	-0.077837
15	8	1.722943	1.340405	-0.476120
16	6	3.128673	-0.981653	0.981057
17	6	2.556325	-1.899358	-0.084344
18	8	1.374967	-1.685906	-0.511340
19	6	-0.990310	-3.113997	0.959380
20	6	-1.925463	-2.531590	-0.085610
21	8	-1.720715	-1.344794	-0.498974
22	6	-3.129057	0.992970	0.970350
23	6	-2.542092	1.911888	-0.087205
24	8	-1.359395	1.698072	-0.500671
25	1	0.820547	1.202802	3.716711
26	1	1.785572	2.633493	3.390081
27	1	3.141880	1.377246	1.726627
28	1	3.197629	0.641026	3.320874
29	1	1.191087	-0.834200	3.716471
30	1	2.622583	-1.797646	3.389176
31	1	1.372637	-3.132303	1.698724
32	1	0.633673	-3.212298	3.290708
33	1	-0.831312	-1.196508	3.709589
34	1	-1.793665	-2.627745	3.377646
35	1	-3.146546	-1.368433	1.712113
36	1	-3.208418	-0.636700	3.307917
37	1	-1.203779	0.838712	3.714724
38	1	-2.634292	1.802420	3.383747
39	1	-1.377241	3.143702	1.704080
40	1	-0.644597	3.217048	3.299493
41	1	0.160469	3.573126	0.412611
42	1	3.586756	-0.148522	0.440431
43	1	-0.160425	-3.559656	0.403132
44	1	-3.587459	0.162008	0.426719
45	8	-0.015881	-0.108629	-2.167530
46	1	0.650542	-0.797342	-2.338231
47	1	-0.870246	-0.530047	-2.364311
48	7	-3.307462	2.866938	-0.568211
49	7	-2.880218	-3.299782	-0.559170
50	7	3.327510	-2.848509	-0.563167
51	7	2.871928	3.299819	-0.561019
52	1	-4.276794	3.072371	-0.255295
53	1	-3.081097	-4.271148	-0.245948
54	1	4.293933	-3.058428	-0.240162
55	1	3.066012	4.274132	-0.257234
56	6	3.793930	2.994129	-1.647817
57	1	3.247328	2.737512	-2.560740
58	6	-3.007157	3.775989	-1.667445
59	1	-2.764493	3.218449	-2.577550
60	6	-3.805646	-3.000968	-1.645317
61	1	-4.440262	-2.145130	-1.394369
62	6	3.039561	-3.756716	-1.666698
63	1	2.818548	-3.198873	-2.582277
64	1	-2.152618	4.415541	-1.424053
65	1	4.437310	2.147060	-1.388742
66	1	2.176441	-4.389580	-1.437339
67	1	-3.262431	-2.763056	-2.565429
68	6	4.324989	-4.633015	-1.852829
69	8	5.265997	-4.364154	-1.042656
70	8	4.274861	-5.483736	-2.765202
71	6	4.655837	4.283949	-1.864379
72	8	4.373040	5.244232	-1.082268

73	8	5.512790	4.218654	-2.770679
74	6	-4.293308	4.644823	-1.877963
75	8	-5.246116	4.377270	-1.081371
76	8	-4.233022	5.491419	-2.794207
77	6	-4.677484	-4.288493	-1.837758
78	8	-4.392546	-5.239375	-1.044826
79	8	-5.541001	-4.230139	-2.737583
80	71	0.000846	0.016094	0.269455
81	1	-1.478713	-3.907315	1.539456
82	1	-3.912811	1.495382	1.551532
83	1	1.483312	3.912380	1.546450
84	1	3.911121	-1.482597	1.565199

E(RTPSSh) = -2393.9493149 Hartree
Zero-point correction = 0.670236 Hartree/particle
Sum of electronic and thermal Energies = -2393.232197 Hartree
Sum of electronic and thermal Enthalpies = -2393.231253 Hartree
Sum of electronic and thermal Free Energies = -2393.362962 Hartree

Table S73. [La(DOTA-(gly)$_4$(H$_2$O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.202542	-1.752912	2.012427
2	6	-0.274710	-2.202297	3.087460
3	6	1.151324	-2.422170	2.591993
4	7	1.769325	-1.191199	2.014448
5	6	2.198971	-0.269923	3.103447
6	6	2.426797	1.159493	2.621833
7	7	1.205245	1.779370	2.027188
8	6	0.277201	2.225589	3.102563
9	6	-1.148831	2.444437	2.606894
10	7	-1.763143	1.215830	2.020249
11	6	-2.200383	0.289952	3.101002
12	6	-2.425678	-1.136340	2.609150
13	6	-1.613344	-2.907770	1.176996
14	6	-2.347741	-2.411375	-0.062626
15	8	-2.010605	-1.294022	-0.571563
16	6	2.937757	-1.587554	1.191438
17	6	2.467737	-2.298168	-0.071734
18	8	1.354152	-1.961598	-0.590933
19	6	1.615253	2.931338	1.188666
20	6	2.340894	2.426692	-0.054412
21	8	2.017180	1.298268	-0.543097
22	6	-2.920899	1.617044	1.185330
23	6	-2.426879	2.330080	-0.069059
24	8	-1.302500	2.001347	-0.564790
25	1	-0.287383	-1.446523	3.875685
26	1	-0.636408	-3.133531	3.545360
27	1	1.162633	-3.193242	1.817215
28	1	1.760697	-2.801751	3.423900
29	1	1.428636	-0.286840	3.877314
30	1	3.121537	-0.635000	3.576129
31	1	3.211884	1.176518	1.861695
32	1	2.790902	1.763287	3.464595
33	1	0.290199	1.468027	3.889292
34	1	0.637142	3.156375	3.563006
35	1	-1.160831	3.220368	1.837142
36	1	-1.760673	2.816978	3.440197

37	1	-1.435143	0.302450	3.880127
38	1	-3.126296	0.651501	3.569925
39	1	-3.208919	-1.148515	1.846867
40	1	-2.791283	-1.746766	3.446366
41	1	-0.716390	-3.434709	0.835736
42	1	3.469199	-0.684444	0.875127
43	1	0.718342	3.461297	0.852234
44	1	-3.453062	0.717017	0.861346
45	8	-0.049268	-0.379968	-2.372991
46	1	0.567688	-1.129140	-2.460461
47	1	-0.930567	-0.755469	-2.542653
48	7	-3.208776	3.242980	-0.600204
49	7	3.260449	3.203578	-0.582025
50	7	3.254313	-3.212673	-0.590283
51	7	-3.284215	-3.180518	-0.569308
52	1	-4.121166	3.554406	-0.211693
53	1	3.569911	4.117545	-0.196186
54	1	4.155916	-3.535018	-0.183930
55	1	-3.607144	-4.083351	-0.166332
56	6	-4.064951	-2.948004	-1.777694
57	1	-3.415154	-2.888220	-2.656756
58	6	-2.987569	3.990914	-1.831162
59	1	-2.893059	3.314035	-2.686249
60	6	4.025678	2.968583	-1.799679
61	1	3.361595	2.874111	-2.664780
62	6	3.048458	-3.962042	-1.823218
63	1	2.989169	-3.287954	-2.683830
64	1	-4.624560	-2.010209	-1.703923
65	1	-2.070042	4.584384	-1.765418
66	1	4.610475	2.046710	-1.718827
67	1	2.118513	-4.537480	-1.776807
68	6	-4.233225	4.924958	-2.001460
69	8	-5.098187	4.822570	-1.076232
70	8	-4.232099	5.655405	-3.014515
71	6	4.972305	4.205515	-1.963825
72	8	4.860457	5.079230	-1.048014
73	8	5.720609	4.189977	-2.963758
74	6	4.281383	-4.919010	-1.958832
75	8	4.289101	-5.657894	-2.965336
76	8	5.127835	-4.821962	-1.016109
77	6	-5.043878	-4.164852	-1.901199
78	8	-4.931918	-5.025640	-0.973194
79	8	-5.812138	-4.148009	-2.885426
80	57	0.004254	0.016772	0.230246
81	1	-2.226097	-3.620530	1.745192
82	1	3.640766	-2.210572	1.760684
83	1	2.235304	3.642632	1.750992
84	1	-3.628392	2.242254	1.746663

E(RTPSSh) = -2385.5707722 Hartree
Zero-point correction = 0.667737 Hartree/particle
Sum of electronic and thermal Energies = -2384.855077 Hartree
Sum of electronic and thermal Enthalpies = -2384.854133 Hartree
Sum of electronic and thermal Free Energies = -2384.990321 Hartree

Table S74. [Ce(DOTA-(gly)$_4$(H$_2$O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z

1	7	-1.187570	-1.759659	2.020346
2	6	-0.260260	-2.206846	3.096473
3	6	1.168148	-2.412725	2.602879
4	7	1.771833	-1.174581	2.025529
5	6	2.198482	-0.253042	3.115144
6	6	2.412692	1.178219	2.633758
7	7	1.184489	1.783001	2.037079
8	6	0.255397	2.227794	3.111845
9	6	-1.172211	2.432507	2.615755
10	7	-1.771064	1.196729	2.028151
11	6	-2.207081	0.270319	3.108637
12	6	-2.418242	-1.157774	2.616600
13	6	-1.584842	-2.911793	1.174826
14	6	-2.318277	-2.409189	-0.062566
15	8	-1.993194	-1.279634	-0.553065
16	6	2.938166	-1.557905	1.193620
17	6	2.462124	-2.268506	-0.066988
18	8	1.336701	-1.944689	-0.569121
19	6	1.582570	2.931404	1.187879
20	6	2.309840	2.418579	-0.050755
21	8	1.999413	1.278044	-0.520156
22	6	-2.925432	1.585119	1.182497
23	6	-2.423240	2.298854	-0.067913
24	8	-1.287548	1.981600	-0.545391
25	1	-0.280835	-1.454015	3.887453
26	1	-0.616303	-3.142622	3.549436
27	1	1.188026	-3.183833	1.828418
28	1	1.781344	-2.785289	3.435064
29	1	1.430538	-0.277123	3.891315
30	1	3.125375	-0.611966	3.583970
31	1	3.198655	1.202991	1.874864
32	1	2.768674	1.786992	3.476298
33	1	0.275038	1.473553	3.901731
34	1	0.609109	3.163332	3.567346
35	1	-1.191640	3.208588	1.846411
36	1	-1.789265	2.797768	3.448340
37	1	-1.445444	0.289777	3.891247
38	1	-3.137959	0.625792	3.572246
39	1	-3.200853	-1.177426	1.853939
40	1	-2.777647	-1.772917	3.452960
41	1	-0.682133	-3.427861	0.832332
42	1	3.458913	-0.649322	0.875432
43	1	0.680566	3.450267	0.848014
44	1	-3.446324	0.679664	0.855612
45	8	-0.051255	-0.336615	-2.328280
46	1	0.567129	-1.082929	-2.428302
47	1	-0.933155	-0.714696	-2.490155
48	7	-3.207581	3.200571	-0.613997
49	7	3.217111	3.198777	-0.594049
50	7	3.253675	-3.169637	-0.600713
51	7	-3.240813	-3.183243	-0.586336
52	1	-4.128283	3.504136	-0.238922
53	1	3.517495	4.121782	-0.222585
54	1	4.165072	-3.481727	-0.207949
55	1	-3.555453	-4.095430	-0.197455
56	6	-4.018515	-2.944067	-1.795478
57	1	-3.365497	-2.866128	-2.670731
58	6	-2.976404	3.948630	-1.843107
59	1	-2.865186	3.271446	-2.695941
60	6	3.982941	2.953975	-1.809474
61	1	3.318653	2.840757	-2.672167
62	6	3.042371	-3.916154	-1.834527

63	1	2.962275	-3.239137	-2.691123
64	1	-4.589048	-2.013681	-1.712509
65	1	-2.064761	4.549763	-1.766224
66	1	4.576862	2.039135	-1.716378
67	1	2.121551	-4.505281	-1.778786
68	6	-4.227529	4.871969	-2.030852
69	8	-5.103886	4.762698	-1.117180
70	8	-4.219116	5.601583	-3.044438
71	6	4.916945	4.197852	-1.992446
72	8	4.795925	5.084163	-1.089971
73	8	5.665622	4.174687	-2.991904
74	6	4.287172	-4.854645	-1.990280
75	8	4.293164	-5.588292	-3.000530
76	8	5.143518	-4.750128	-1.057301
77	6	-4.982874	-4.170502	-1.938493
78	8	-4.864317	-5.041533	-1.020861
79	8	-5.747492	-4.150092	-2.925376
80	58	0.003111	0.016163	0.262136
81	1	-2.194158	-3.634140	1.734413
82	1	3.650566	-2.176968	1.755348
83	1	2.198267	3.652737	1.742004
84	1	-3.643108	2.206068	1.735432

E(RTPSSh) = -2386.2222416 Hartree
Zero-point correction = 0.667943 Hartree/particle
Sum of electronic and thermal Energies = -2385.506442 Hartree
Sum of electronic and thermal Enthalpies = -2385.505498 Hartree
Sum of electronic and thermal Free Energies = -2385.641236 Hartree

Table S75. [Pr(DOTA-(gly)$_4$(H$_2$O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.153351	-1.774704	2.030664
2	6	-0.220761	-2.209662	3.106945
3	6	1.210782	-2.388991	2.613273
4	7	1.787932	-1.139078	2.034168
5	6	2.203900	-0.212330	3.123155
6	6	2.390847	1.222111	2.641006
7	7	1.150263	1.800371	2.043477
8	6	0.216991	2.234872	3.118588
9	6	-1.214335	2.412292	2.623800
10	7	-1.787697	1.163990	2.037157
11	6	-2.211112	0.233379	3.118760
12	6	-2.395775	-1.198383	2.627762
13	6	-1.527889	-2.928373	1.177317
14	6	-2.266387	-2.427956	-0.057589
15	8	-1.964283	-1.284707	-0.531657
16	6	2.954372	-1.499478	1.192649
17	6	2.477745	-2.216249	-0.063847
18	8	1.338733	-1.914519	-0.549634
19	6	1.525116	2.948540	1.183637
20	6	2.258860	2.434408	-0.050400
21	8	1.971067	1.280338	-0.501413
22	6	-2.943988	1.528735	1.183933
23	6	-2.443751	2.246988	-0.064243
24	8	-1.296447	1.950256	-0.527389
25	1	-0.255083	-1.459035	3.899634
26	1	-0.561687	-3.152513	3.556799

27	1	1.244922	-3.160448	1.839763
28	1	1.832411	-2.748229	3.444988
29	1	1.439342	-0.250277	3.902210
30	1	3.138688	-0.555978	3.587684
31	1	3.176391	1.261264	1.882430
32	1	2.734104	1.839372	3.482550
33	1	0.251320	1.484163	3.911417
34	1	0.556129	3.178237	3.568904
35	1	-1.248962	3.187423	1.854138
36	1	-1.838600	2.765582	3.456092
37	1	-1.451588	0.267505	3.903002
38	1	-3.149065	0.574058	3.579125
39	1	-3.178459	-1.233083	1.865825
40	1	-2.742133	-1.820921	3.464080
41	1	-0.615534	-3.425915	0.833123
42	1	3.455168	-0.581139	0.870636
43	1	0.613119	3.447209	0.840331
44	1	-3.446107	0.613210	0.855885
45	8	-0.048724	-0.297575	-2.286995
46	1	0.576768	-1.036679	-2.395706
47	1	-0.927461	-0.684290	-2.446425
48	7	-3.239328	3.130481	-0.623363
49	7	3.147822	3.224806	-0.608588
50	7	3.280203	-3.099258	-0.610986
51	7	-3.168699	-3.214964	-0.596482
52	1	-4.169534	3.418218	-0.259374
53	1	3.430908	4.158935	-0.251522
54	1	4.202165	-3.394915	-0.230215
55	1	-3.464799	-4.139192	-0.221647
56	6	-3.944483	-2.977360	-1.807234
57	1	-3.288139	-2.871952	-2.677104
58	6	-3.009109	3.877693	-1.853193
59	1	-2.875103	3.199239	-2.701707
60	6	3.915505	2.977207	-1.822314
61	1	3.251609	2.838783	-2.681617
62	6	3.067619	-3.847730	-1.843424
63	1	2.965225	-3.171146	-2.697981
64	1	-4.537333	-2.061921	-1.715134
65	1	-2.110388	4.497016	-1.768875
66	1	4.527234	2.075479	-1.717519
67	1	2.158374	-4.453539	-1.777703
68	6	-4.275844	4.775588	-2.058824
69	8	-5.159293	4.654441	-1.153502
70	8	-4.270901	5.499913	-3.076197
71	6	4.824946	4.236043	-2.025385
72	8	4.688413	5.133028	-1.135733
73	8	5.572012	4.212675	-3.026020
74	6	4.327378	-4.763144	-2.015461
75	8	4.334768	-5.494972	-3.026991
76	8	5.192622	-4.644504	-1.092410
77	6	-4.878858	-4.223978	-1.973018
78	8	-4.745670	-5.104287	-1.066310
79	8	-5.637471	-4.208052	-2.964581
80	59	0.002160	0.017614	0.290184
81	1	-2.126434	-3.664850	1.729961
82	1	3.682607	-2.106801	1.746723
83	1	2.129186	3.685868	1.729317
84	1	-3.675917	2.138901	1.730041

E(RTPSSh) = -2386.8571702 Hartree
Zero-point correction = 0.668228 Hartree/particle
Sum of electronic and thermal Energies = -2386.141211 Hartree

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 Sum of electronic and thermal Enthalpies = -2386.140267 Hartree
 Sum of electronic and thermal Free Energies = -2386.275453 Hartree
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Table S76. [Nd(DOTA-$(gly)_4(H_2O)]^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.138647	-1.778470	2.037905
2	6	-0.205658	-2.213666	3.113458
3	6	1.226368	-2.382741	2.618850
4	7	1.791988	-1.127498	2.040033
5	6	2.207571	-0.201703	3.129700
6	6	2.384524	1.233949	2.648477
7	7	1.138955	1.801720	2.051104
8	6	0.206449	2.235319	3.127039
9	6	-1.226031	2.402938	2.633283
10	7	-1.788847	1.149845	2.046673
11	6	-2.209835	0.218796	3.128616
12	6	-2.385328	-1.213532	2.636924
13	6	-1.505027	-2.928527	1.176400
14	6	-2.243555	-2.420820	-0.055160
15	8	-1.949586	-1.268959	-0.513897
16	6	2.955136	-1.478474	1.190353
17	6	2.471500	-2.192741	-0.064569
18	8	1.324128	-1.898324	-0.535589
19	6	1.505194	2.947191	1.184021
20	6	2.237812	2.426667	-0.047778
21	8	1.958005	1.264839	-0.483998
22	6	-2.943764	1.505962	1.188123
23	6	-2.439343	2.222282	-0.059086
24	8	-1.285163	1.932912	-0.509914
25	1	-0.244380	-1.466256	3.909047
26	1	-0.542391	-3.160057	3.558926
27	1	1.265120	-3.153321	1.844758
28	1	1.852177	-2.737620	3.449228
29	1	1.445819	-0.245147	3.911287
30	1	3.145806	-0.541563	3.589974
31	1	3.169773	1.278866	1.890013
32	1	2.722755	1.854197	3.489763
33	1	0.246253	1.487005	3.921941
34	1	0.541872	3.181782	3.573490
35	1	-1.266302	3.177777	1.863665
36	1	-1.853265	2.751224	3.465371
37	1	-1.451676	0.257383	3.914031
38	1	-3.150340	0.555170	3.586832
39	1	-3.168610	-1.252907	1.875913
40	1	-2.725793	-1.840116	3.472559
41	1	-0.589468	-3.418408	0.829938
42	1	3.448809	-0.556574	0.867874
43	1	0.589831	3.439001	0.840033
44	1	-3.439712	0.587121	0.860307
45	8	-0.048145	-0.257888	-2.250508
46	1	0.573780	-0.998339	-2.369581
47	1	-0.928879	-0.639246	-2.412017
48	7	-3.236717	3.096465	-0.629898
49	7	3.117963	3.217699	-0.618597
50	7	3.274991	-3.066125	-0.625113
51	7	-3.137620	-3.208293	-0.606510
52	1	-4.172995	3.377559	-0.276330

53	1	3.394764	4.158177	-0.273319
54	1	4.202999	-3.356809	-0.255012
55	1	-3.427234	-4.139531	-0.243840
56	6	-3.912464	-2.962494	-1.816267
57	1	-3.255013	-2.841607	-2.683266
58	6	-3.001952	3.838933	-1.861766
59	1	-2.852278	3.157059	-2.704875
60	6	3.883429	2.962543	-1.832184
61	1	3.217754	2.809228	-2.687569
62	6	3.055381	-3.811884	-1.857981
63	1	2.941354	-3.133268	-2.709443
64	1	-4.512934	-2.053051	-1.714750
65	1	-2.110704	4.468247	-1.772113
66	1	4.502737	2.066974	-1.719433
67	1	2.150462	-4.423382	-1.785930
68	6	-4.275922	4.722279	-2.085149
69	8	-5.166337	4.598064	-1.187076
70	8	-4.269063	5.439794	-3.107298
71	6	4.782051	4.226284	-2.052395
72	8	4.640675	5.132294	-1.172670
73	8	5.526484	4.197490	-3.054820
74	6	4.319254	-4.718931	-2.044408
75	8	4.321747	-5.448141	-3.057794
76	8	5.192440	-4.596961	-1.129268
77	6	-4.836265	-4.214715	-1.998777
78	8	-4.696893	-5.104880	-1.102641
79	8	-5.593645	-4.192981	-2.991113
80	60	0.002926	0.018480	0.314767
81	1	-2.101204	-3.671861	1.722309
82	1	3.689812	-2.084641	1.737069
83	1	2.107654	3.690441	1.723323
84	1	-3.680680	2.114732	1.728980

E(RTPSSh) = -2387.4797495 Hartree
Zero-point correction = 0.668445 Hartree/particle
Sum of electronic and thermal Energies = -2386.763680 Hartree
Sum of electronic and thermal Enthalpies = -2386.762736 Hartree
Sum of electronic and thermal Free Energies = -2386.897563 Hartree

Table S77. [Sm(DOTA-(gly)$_4$(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.118796	-1.785883	2.045588
2	6	-0.187411	-2.227856	3.119247
3	6	1.244732	-2.383196	2.623098
4	7	1.795192	-1.119448	2.048144
5	6	2.212455	-0.199398	3.141589
6	6	2.377628	1.238482	2.665068
7	7	1.125773	1.793680	2.068588
8	6	0.196040	2.226753	3.146754
9	6	-1.237265	2.383555	2.654264
10	7	-1.785314	1.125472	2.064294
11	6	-2.206502	0.193472	3.144884
12	6	-2.370956	-1.237928	2.649034
13	6	-1.476093	-2.926338	1.167991
14	6	-2.212699	-2.399504	-0.055881
15	8	-1.924970	-1.234731	-0.486381
16	6	2.952866	-1.457250	1.186390

17	6	2.456589	-2.160878	-0.068845
18	8	1.296650	-1.873716	-0.514293
19	6	1.482057	2.935683	1.193043
20	6	2.209517	2.404964	-0.036891
21	8	1.940339	1.231708	-0.449044
22	6	-2.935957	1.473320	1.197099
23	6	-2.420337	2.186741	-0.046421
24	8	-1.254527	1.908084	-0.473951
25	1	-0.231269	-1.488125	3.921842
26	1	-0.520621	-3.180213	3.554380
27	1	1.288856	-3.150348	1.846002
28	1	1.876317	-2.734832	3.450323
29	1	1.454816	-0.251319	3.926772
30	1	3.154929	-0.537119	3.594585
31	1	3.162750	1.292161	1.907197
32	1	2.708967	1.860941	3.507276
33	1	0.242202	1.480797	3.943641
34	1	0.528314	3.176308	3.588763
35	1	-1.284212	3.159840	1.886594
36	1	-1.869186	2.723086	3.486258
37	1	-1.451249	0.235397	3.933033
38	1	-3.150472	0.525084	3.599273
39	1	-3.155406	-1.281209	1.889555
40	1	-2.703216	-1.872237	3.481966
41	1	-0.557445	-3.406566	0.816846
42	1	3.440164	-0.530989	0.867645
43	1	0.563244	3.421470	0.850439
44	1	-3.425539	0.551851	0.867976
45	8	-0.040255	-0.196935	-2.184762
46	1	0.568540	-0.946791	-2.312742
47	1	-0.927021	-0.560537	-2.353641
48	7	-3.216779	3.047826	-0.637637
49	7	3.075247	3.195579	-0.629454
50	7	3.258847	-3.018079	-0.654788
51	7	-3.099525	-3.179444	-0.628395
52	1	-4.163604	3.318270	-0.304365
53	1	3.342644	4.145851	-0.304029
54	1	4.195713	-3.304417	-0.303431
55	1	-3.384655	-4.120579	-0.287849
56	6	-3.873820	-2.910978	-1.833707
57	1	-3.215838	-2.768678	-2.697008
58	6	-2.969587	3.782654	-1.871674
59	1	-2.791402	3.095615	-2.704940
60	6	3.832498	2.928661	-1.845673
61	1	3.160871	2.749798	-2.691357
62	6	3.026009	-3.753495	-1.891456
63	1	2.895737	-3.067704	-2.734809
64	1	-4.478508	-2.006665	-1.713493
65	1	-2.091037	4.427863	-1.770272
66	1	4.465903	2.044385	-1.722530
67	1	2.126342	-4.371741	-1.812391
68	6	-4.253483	4.642170	-2.128907
69	8	-5.158964	4.512056	-1.246851
70	8	-4.238780	5.349266	-3.158197
71	6	4.710549	4.200860	-2.098018
72	8	4.564361	5.120889	-1.233738
73	8	5.445760	4.164241	-3.106952
74	6	4.293640	-4.649554	-2.104746
75	8	4.285990	-5.370804	-3.123684
76	8	5.180015	-4.527571	-1.202324
77	6	-4.791297	-4.163569	-2.044255
78	8	-4.646275	-5.073114	-1.168639

79	8	-5.549514	-4.123485	-3.035299
80	62	0.006433	0.017458	0.356208
81	1	-2.071533	-3.679608	1.700753
82	1	3.693902	-2.066977	1.720254
83	1	2.085725	3.684185	1.723489
84	1	-3.678755	2.082065	1.729726

E(RTPSSh) = -2388.7016232 Hartree
Zero-point correction = 0.668791 Hartree/particle
Sum of electronic and thermal Energies = -2387.985379 Hartree
Sum of electronic and thermal Enthalpies = -2387.984435 Hartree
Sum of electronic and thermal Free Energies = -2388.118854 Hartree

Table S78. [Eu(DOTA-(gly)$_4$(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.112840	-1.790117	2.048511
2	6	-0.183723	-2.238775	3.121101
3	6	1.248337	-2.390316	2.624778
4	7	1.794889	-1.123132	2.053805
5	6	2.214882	-0.208254	3.150281
6	6	2.377872	1.230896	2.677849
7	7	1.124382	1.783798	2.082622
8	6	0.196749	2.216988	3.162330
9	6	-1.236503	2.372834	2.670462
10	7	-1.780317	1.114832	2.076514
11	6	-2.204309	0.181415	3.154506
12	6	-2.366627	-1.248259	2.654031
13	6	-1.467880	-2.923986	1.161697
14	6	-2.200976	-2.385068	-0.058700
15	8	-1.913485	-1.214409	-0.473839
16	6	2.949324	-1.456232	1.186269
17	6	2.445688	-2.150961	-0.070714
18	8	1.279717	-1.865931	-0.502164
19	6	1.478493	2.924638	1.204873
20	6	2.200449	2.389588	-0.026239
21	8	1.935774	1.211148	-0.426675
22	6	-2.927565	1.463069	1.205246
23	6	-2.404212	2.174844	-0.035731
24	8	-1.232410	1.900946	-0.450024
25	1	-0.228562	-1.503560	3.927841
26	1	-0.517722	-3.193642	3.550006
27	1	1.293558	-3.154784	1.845168
28	1	1.881291	-2.743049	3.450420
29	1	1.458882	-0.263346	3.936888
30	1	3.158450	-0.547740	3.599597
31	1	3.162957	1.287911	1.920261
32	1	2.707385	1.852458	3.521362
33	1	0.243923	1.470661	3.958856
34	1	0.529644	3.166474	3.603922
35	1	-1.284760	3.151340	1.905191
36	1	-1.869915	2.708006	3.503023
37	1	-1.450641	0.221631	3.944308
38	1	-3.149326	0.512150	3.607262
39	1	-3.151189	-1.290207	1.894639
40	1	-2.697057	-1.886775	3.484402
41	1	-0.548580	-3.401416	0.808772
42	1	3.436384	-0.528655	0.871451

43	1	0.559160	3.411044	0.864910
44	1	-3.416789	0.542117	0.874544
45	8	-0.034303	-0.167278	-2.153862
46	1	0.568926	-0.920821	-2.286549
47	1	-0.923780	-0.523948	-2.323561
48	7	-3.199023	3.029353	-0.638314
49	7	3.056782	3.180717	-0.631346
50	7	3.246772	-2.998504	-0.671811
51	7	-3.084777	-3.159155	-0.643295
52	1	-4.151795	3.293689	-0.317134
53	1	3.318250	4.136358	-0.316784
54	1	4.188343	-3.282960	-0.331236
55	1	-3.369942	-4.104613	-0.314403
56	6	-3.857544	-2.877797	-1.846690
57	1	-3.198661	-2.726276	-2.707721
58	6	-2.944622	3.759245	-1.873882
59	1	-2.749264	3.069087	-2.700667
60	6	3.804809	2.910723	-1.852646
61	1	3.126843	2.715255	-2.689506
62	6	3.007575	-3.724841	-1.912670
63	1	2.867430	-3.032932	-2.749428
64	1	-4.462715	-1.975063	-1.717538
65	1	-2.074211	4.414377	-1.766001
66	1	4.450347	2.035553	-1.727319
67	1	2.111800	-4.348434	-1.831676
68	6	-4.234305	4.603670	-2.151599
69	8	-5.148359	4.470106	-1.278941
70	8	-4.215180	5.303848	-3.185511
71	6	4.664726	4.190949	-2.126359
72	8	4.517779	5.116664	-1.268295
73	8	5.388369	4.154204	-3.143579
74	6	4.278329	-4.612312	-2.143419
75	8	4.266423	-5.325306	-3.168045
76	8	5.171290	-4.492403	-1.247211
77	6	-4.774165	-4.128644	-2.071618
78	8	-4.628489	-5.047798	-1.206161
79	8	-5.532104	-4.078172	-3.062330
80	63	0.009383	0.016121	0.378065
81	1	-2.065355	-3.680994	1.686745
82	1	3.691401	-2.070734	1.713122
83	1	2.085616	3.672788	1.731808
84	1	-3.671523	2.073471	1.734296

E(RTPSSh) = -2389.2984991 Hartree
Zero-point correction = 0.668880 Hartree/particle
Sum of electronic and thermal Energies = -2388.582213 Hartree
Sum of electronic and thermal Enthalpies = -2388.581269 Hartree
Sum of electronic and thermal Free Energies = -2388.715821 Hartree

Table S79. [Gd(DOTA-(gly)$_4$(H$_2$O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.109892	-1.796873	2.049431
2	6	-0.184094	-2.251781	3.122055
3	6	1.248537	-2.399823	2.627353
4	7	1.792556	-1.129376	2.061359
5	6	2.213620	-0.219675	3.161512
6	6	2.375200	1.220846	2.693653

7	7	1.120895	1.772178	2.098696
8	6	0.193808	2.204262	3.179138
9	6	-1.238799	2.359850	2.686252
10	7	-1.778199	1.102571	2.086853
11	6	-2.206234	0.167013	3.161155
12	6	-2.365961	-1.260722	2.655269
13	6	-1.461774	-2.924636	1.153802
14	6	-2.189846	-2.374826	-0.064448
15	8	-1.902277	-1.198933	-0.465072
16	6	2.945332	-1.457945	1.190245
17	6	2.437085	-2.143005	-0.069959
18	8	1.266081	-1.860491	-0.489709
19	6	1.474036	2.913014	1.220758
20	6	2.191172	2.375836	-0.012097
21	8	1.931494	1.193165	-0.403435
22	6	-2.921242	1.452158	1.210789
23	6	-2.389253	2.163572	-0.026521
24	8	-1.211938	1.893932	-0.427982
25	1	-0.230799	-1.520847	3.932622
26	1	-0.519319	-3.208979	3.544694
27	1	1.295674	-3.161415	1.845101
28	1	1.881551	-2.754044	3.452246
29	1	1.458135	-0.278032	3.948423
30	1	3.157631	-0.560918	3.608492
31	1	3.160854	1.281181	1.936972
32	1	2.702382	1.841105	3.538965
33	1	0.240862	1.456678	3.974518
34	1	0.526732	3.153172	3.621850
35	1	-1.287272	3.141093	1.923845
36	1	-1.874507	2.690282	3.518888
37	1	-1.455105	0.205066	3.953516
38	1	-3.152821	0.496638	3.611343
39	1	-3.149409	-1.300825	1.894666
40	1	-2.696070	-1.903514	3.482391
41	1	-0.541469	-3.399719	0.800642
42	1	3.433296	-0.529155	0.880818
43	1	0.554672	3.401027	0.883530
44	1	-3.409893	0.532052	0.877233
45	8	-0.026740	-0.143906	-2.126135
46	1	0.572098	-0.900301	-2.262506
47	1	-0.918277	-0.495338	-2.296360
48	7	-3.181289	3.013338	-0.639187
49	7	3.038065	3.168706	-0.627902
50	7	3.238594	-2.979652	-0.685356
51	7	-3.069743	-3.143701	-0.661357
52	1	-4.139362	3.272519	-0.329659
53	1	3.292108	4.129531	-0.323088
54	1	4.184980	-3.260507	-0.354981
55	1	-3.355084	-4.093062	-0.343530
56	6	-3.839214	-2.850979	-1.864204
57	1	-3.178169	-2.691214	-2.722084
58	6	-2.918120	3.740388	-1.874655
59	1	-2.707830	3.048552	-2.696343
60	6	3.775370	2.898587	-1.855740
61	1	3.090404	2.684728	-2.682260
62	6	2.996539	-3.693873	-1.932731
63	1	2.844325	-2.993846	-2.760594
64	1	-4.445225	-1.949858	-1.728108
65	1	-2.054201	4.402794	-1.759308
66	1	4.436053	2.034783	-1.730207
67	1	2.107085	-4.326535	-1.852480
68	6	-4.211073	4.573556	-2.170674

69	8	-5.134085	4.436157	-1.308095
70	8	-4.185402	5.269826	-3.207063
71	6	4.612092	4.189225	-2.151733
72	8	4.464736	5.118849	-1.297987
73	8	5.320571	4.155548	-3.179645
74	6	4.273713	-4.566996	-2.182857
75	8	4.261635	-5.266948	-3.216377
76	8	5.171334	-4.450168	-1.290918
77	6	-4.754411	-4.100343	-2.103453
78	8	-4.609527	-5.027820	-1.246763
79	8	-5.510322	-4.040861	-3.095148
80	64	0.011763	0.013071	0.398213
81	1	-2.062028	-3.684802	1.670985
82	1	3.687091	-2.077671	1.711390
83	1	2.084497	3.659708	1.745874
84	1	-3.666713	2.063747	1.736282

E(RTPSSh) = -2389.8883268 Hartree
Zero-point correction = 0.668996 Hartree/particle
Sum of electronic and thermal Energies = -2389.171989 Hartree
Sum of electronic and thermal Enthalpies = -2389.171045 Hartree
Sum of electronic and thermal Free Energies = -2389.305377 Hartree

Table S80. [Tb(DOTA-(gly)$_4$(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.106082	-1.797955	2.051189
2	6	-0.182641	-2.254266	3.124950
3	6	1.250566	-2.397648	2.631740
4	7	1.789202	-1.124738	2.066133
5	6	2.209944	-0.216191	3.167114
6	6	2.367241	1.224529	2.699508
7	7	1.111153	1.769949	2.102659
8	6	0.184213	2.203294	3.182474
9	6	-1.248031	2.354137	2.688237
10	7	-1.780804	1.094361	2.088096
11	6	-2.210672	0.159904	3.162347
12	6	-2.365175	-1.267905	2.656152
13	6	-1.453756	-2.923260	1.150996
14	6	-2.178283	-2.367393	-0.066132
15	8	-1.890845	-1.188149	-0.457223
16	6	2.940862	-1.449908	1.192493
17	6	2.428494	-2.131343	-0.067596
18	8	1.253860	-1.850741	-0.479040
19	6	1.462011	2.908506	1.220874
20	6	2.178918	2.365070	-0.008952
21	8	1.921104	1.178954	-0.391069
22	6	-2.921097	1.440570	1.207227
23	6	-2.382262	2.149892	-0.027817
24	8	-1.201748	1.880504	-0.420093
25	1	-0.232336	-1.525381	3.937247
26	1	-0.517184	-3.213122	3.544300
27	1	1.300856	-3.159029	1.849492
28	1	1.884893	-2.749373	3.456632
29	1	1.455407	-0.276996	3.954810
30	1	3.155180	-0.556029	3.612501
31	1	3.153759	1.287439	1.943957
32	1	2.690660	1.846845	3.544678

33	1	0.232762	1.457638	3.979632
34	1	0.515538	3.153886	3.622680
35	1	-1.298238	3.135226	1.925790
36	1	-1.886621	2.681963	3.519625
37	1	-1.462081	0.200202	3.957059
38	1	-3.159339	0.488222	3.609033
39	1	-3.147840	-1.310393	1.894880
40	1	-2.693375	-1.912894	3.482253
41	1	-0.532293	-3.396540	0.798825
42	1	3.428106	-0.520388	0.884562
43	1	0.542176	3.394407	0.882299
44	1	-3.407675	0.519667	0.873264
45	8	-0.020497	-0.124972	-2.099761
46	1	0.570337	-0.887314	-2.237488
47	1	-0.914726	-0.465235	-2.277375
48	7	-3.169517	2.999047	-0.647296
49	7	3.025114	3.153929	-0.630690
50	7	3.228040	-2.963785	-0.690870
51	7	-3.056021	-3.132343	-0.670958
52	1	-4.129983	3.258326	-0.345540
53	1	3.278008	4.117357	-0.333419
54	1	4.177525	-3.242890	-0.367815
55	1	-3.341793	-4.084084	-0.360644
56	6	-3.822681	-2.831235	-1.873513
57	1	-3.159661	-2.665814	-2.728797
58	6	-2.896360	3.725070	-1.881163
59	1	-2.680265	3.032512	-2.700726
60	6	3.762744	2.874661	-1.856252
61	1	3.077982	2.653962	-2.681119
62	6	2.980302	-3.673863	-1.939489
63	1	2.820338	-2.971152	-2.763619
64	1	-4.428646	-1.930825	-1.732647
65	1	-2.032704	4.386703	-1.759596
66	1	4.423675	2.012067	-1.723867
67	1	2.093506	-4.309818	-1.855971
68	6	-4.186231	4.559116	-2.187785
69	8	-5.116615	4.421908	-1.333121
70	8	-4.151469	5.255688	-3.223729
71	6	4.599034	4.163202	-2.162265
72	8	4.450482	5.099847	-1.316429
73	8	5.308280	4.121482	-3.189382
74	6	4.258905	-4.541481	-2.201330
75	8	4.242267	-5.238227	-3.236927
76	8	5.162247	-4.424039	-1.315230
77	6	-4.737508	-4.078818	-2.123320
78	8	-4.593594	-5.012629	-1.273340
79	8	-5.492074	-4.012217	-3.115577
80	65	0.010808	0.012014	0.411526
81	1	-2.055625	-3.685403	1.663310
82	1	3.683505	-2.071450	1.710172
83	1	2.072570	3.657376	1.742696
84	1	-3.668915	2.053051	1.728275

E(RTPSSh) = -2390.4781089 Hartree
Zero-point correction = 0.669161 Hartree/particle
Sum of electronic and thermal Energies = -2389.761678 Hartree
Sum of electronic and thermal Enthalpies = -2389.760733 Hartree
Sum of electronic and thermal Free Energies = -2389.894718 Hartree

Table S81. [Dy(DOTA-(gly)$_4$(H_2O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.110806	-1.793172	2.053889
2	6	-0.191656	-2.255525	3.128503
3	6	1.241312	-2.401958	2.636563
4	7	1.781367	-1.129504	2.071379
5	6	2.207190	-0.224401	3.172986
6	6	2.367656	1.215793	2.705802
7	7	1.112388	1.762156	2.108001
8	6	0.188122	2.200957	3.187689
9	6	-1.243245	2.355028	2.692895
10	7	-1.776592	1.096030	2.091669
11	6	-2.212855	0.164236	3.165385
12	6	-2.369969	-1.262627	2.658245
13	6	-1.459850	-2.914151	1.149046
14	6	-2.178150	-2.349056	-0.067141
15	8	-1.886842	-1.167149	-0.447701
16	6	2.929574	-1.457139	1.194384
17	6	2.408749	-2.132448	-0.065261
18	8	1.230924	-1.849524	-0.466410
19	6	1.466179	2.896945	1.222594
20	6	2.178767	2.344560	-0.005478
21	8	1.917781	1.156036	-0.377969
22	6	-2.912443	1.445211	1.206383
23	6	-2.363549	2.149681	-0.026653
24	8	-1.180495	1.876319	-0.408557
25	1	-0.240148	-1.528212	3.942327
26	1	-0.530463	-3.214176	3.544846
27	1	1.290634	-3.163398	1.854341
28	1	1.874981	-2.754479	3.461560
29	1	1.453615	-0.283592	3.961771
30	1	3.152065	-0.567924	3.616260
31	1	3.154877	1.277297	1.950900
32	1	2.691077	1.838093	3.550920
33	1	0.234369	1.456441	3.986085
34	1	0.523040	3.151212	3.625837
35	1	-1.291474	3.136694	1.930935
36	1	-1.882313	2.683082	3.523763
37	1	-1.466182	0.202215	3.962061
38	1	-3.161658	0.496021	3.609159
39	1	-3.152135	-1.302991	1.896366
40	1	-2.699595	-1.908190	3.483268
41	1	-0.539340	-3.389627	0.797636
42	1	3.420285	-0.529211	0.887502
43	1	0.547961	3.385411	0.883643
44	1	-3.401669	0.526000	0.871950
45	8	-0.017692	-0.108990	-2.077183
46	1	0.564409	-0.877690	-2.216621
47	1	-0.915664	-0.439087	-2.255096
48	7	-3.143223	2.999434	-0.654658
49	7	3.025302	3.126781	-0.634913
50	7	3.202887	-2.962543	-0.698231
51	7	-3.054920	-3.107482	-0.681226
52	1	-4.105483	3.261877	-0.361539
53	1	3.280897	4.091805	-0.345268
54	1	4.154755	-3.243321	-0.383551
55	1	-3.344403	-4.060636	-0.378413
56	6	-3.817686	-2.796029	-1.883670
57	1	-3.152224	-2.628203	-2.736600
58	6	-2.857707	3.721975	-1.887795

59	1	-2.637332	3.026951	-2.704124
60	6	3.759985	2.836730	-1.859761
61	1	3.073144	2.611692	-2.681704
62	6	2.945651	-3.667705	-1.947763
63	1	2.780570	-2.961848	-2.768217
64	1	-4.420516	-1.894194	-1.738700
65	1	-1.992849	4.380959	-1.760628
66	1	4.419063	1.973525	-1.722268
67	1	2.059005	-4.303316	-1.860373
68	6	-4.142274	4.559573	-2.206735
69	8	-5.080593	4.426015	-1.360234
70	8	-4.096182	5.254801	-3.243148
71	6	4.598796	4.120953	-2.176845
72	8	4.453359	5.064448	-1.338136
73	8	5.306605	4.069613	-3.204522
74	6	4.221899	-4.535119	-2.221960
75	8	4.197001	-5.228863	-3.259370
76	8	5.132065	-4.420308	-1.342532
77	6	-4.736494	-4.038852	-2.143016
78	8	-4.596863	-4.978630	-1.298910
79	8	-5.489258	-3.963349	-3.135957
80	66	0.010484	0.011491	0.425251
81	1	-2.066781	-3.675537	1.656415
82	1	3.670433	-2.084061	1.708092
83	1	2.081089	3.644636	1.740925
84	1	-3.659286	2.062360	1.723252

E(RTPSSh) = -2391.0642526 Hartree
Zero-point correction = 0.669323 Hartree/particle
Sum of electronic and thermal Energies = -2390.347727 Hartree
Sum of electronic and thermal Enthalpies = -2390.346783 Hartree
Sum of electronic and thermal Free Energies = -2390.480435 Hartree

Table S82. [Ho(DOTA-$(gly)_4(H_2O)]^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.109895	-1.790774	2.057366
2	6	-0.193486	-2.256302	3.132723
3	6	1.239455	-2.402260	2.641634
4	7	1.777462	-1.129092	2.076173
5	6	2.206427	-0.225928	3.177918
6	6	2.366129	1.213999	2.710643
7	7	1.109951	1.758178	2.112685
8	6	0.187661	2.199669	3.192761
9	6	-1.243426	2.353374	2.698039
10	7	-1.774489	1.093852	2.095908
11	6	-2.214269	0.163533	3.169218
12	6	-2.370378	-1.262806	2.661198
13	6	-1.457622	-2.908739	1.148561
14	6	-2.171216	-2.337047	-0.066914
15	8	-1.879916	-1.151741	-0.437234
16	6	2.922778	-1.456543	1.195594
17	6	2.394843	-2.127599	-0.063048
18	8	1.213541	-1.844596	-0.454311
19	6	1.463352	2.890435	1.223998
20	6	2.172080	2.331515	-0.003039
21	8	1.910868	1.140098	-0.366091
22	6	-2.907251	1.443204	1.206920

23	6	-2.350893	2.143914	-0.024522
24	8	-1.164970	1.869464	-0.396816
25	1	-0.242430	-1.530363	3.947779
26	1	-0.533876	-3.215457	3.546564
27	1	1.289542	-3.163950	1.859734
28	1	1.873648	-2.753619	3.466662
29	1	1.454373	-0.285292	3.968176
30	1	3.152049	-0.570500	3.618719
31	1	3.153314	1.275830	1.955758
32	1	2.688669	1.837286	3.555310
33	1	0.233915	1.456189	3.992148
34	1	0.523832	3.150330	3.629003
35	1	-1.291989	3.135496	1.936592
36	1	-1.883424	2.680050	3.528672
37	1	-1.469420	0.201249	3.967650
38	1	-3.163960	0.496228	3.610346
39	1	-3.151976	-1.303090	1.898753
40	1	-2.699713	-1.909737	3.485206
41	1	-0.536971	-3.384376	0.798046
42	1	3.414413	-0.529056	0.889187
43	1	0.545398	3.379486	0.885506
44	1	-3.397222	0.524517	0.872449
45	8	-0.015624	-0.094624	-2.056095
46	1	0.559527	-0.868188	-2.197292
47	1	-0.916437	-0.416955	-2.233822
48	7	-3.125426	2.992142	-0.660721
49	7	3.016173	3.109612	-0.640643
50	7	3.184817	-2.954651	-0.704915
51	7	-3.044231	-3.091956	-0.690366
52	1	-4.089708	3.255639	-0.375501
53	1	3.272145	4.076668	-0.358367
54	1	4.139320	-3.235447	-0.398303
55	1	-3.334301	-4.047311	-0.394942
56	6	-3.803129	-2.773070	-1.893351
57	1	-3.134995	-2.600536	-2.743267
58	6	-2.830097	3.711320	-1.893536
59	1	-2.604246	3.013964	-2.706358
60	6	3.747281	2.811307	-1.865664
61	1	3.058012	2.580161	-2.683836
62	6	2.918855	-3.656306	-1.954633
63	1	2.747686	-2.948190	-2.771902
64	1	-4.406231	-1.872002	-1.744933
65	1	-1.965623	4.369845	-1.761567
66	1	4.407225	1.949426	-1.724098
67	1	2.033158	-4.292569	-1.862708
68	6	-4.111471	4.549164	-2.224405
69	8	-5.056490	4.418151	-1.385044
70	8	-4.056592	5.241951	-3.262042
71	6	4.584463	4.093645	-2.194473
72	8	4.441213	5.042938	-1.361997
73	8	5.288913	4.035318	-3.224096
74	6	4.193621	-4.522176	-2.240663
75	8	4.161150	-5.213528	-3.279439
76	8	5.110393	-4.408599	-1.367998
77	6	-4.721206	-4.014340	-2.163002
78	8	-4.583627	-4.959344	-1.324434
79	8	-5.471219	-3.932697	-3.157503
80	67	0.010399	0.011564	0.438265
81	1	-2.067678	-3.670561	1.651453
82	1	3.663641	-2.086679	1.705312
83	1	2.081176	3.638143	1.738773
84	1	-3.654219	2.063340	1.719975

```
 E(RTPSSh) = -2391.6480766 Hartree
 Zero-point correction = 0.669492 Hartree/particle
 Sum of electronic and thermal Energies = -2390.931451 Hartree
 Sum of electronic and thermal Enthalpies = -2390.930507 Hartree
 Sum of electronic and thermal Free Energies = -2391.063757 Hartree
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Table S83. [Er(DOTA-$(gly)_4(H_2O)]^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.103969	-1.789551	2.062889
2	6	-0.188452	-2.254840	3.138842
3	6	1.244598	-2.396983	2.647968
4	7	1.777153	-1.122132	2.081292
5	6	2.207126	-0.219110	3.182484
6	6	2.362231	1.220683	2.714248
7	7	1.103626	1.759052	2.116116
8	6	0.182395	2.201387	3.196512
9	6	-1.248962	2.350714	2.702179
10	7	-1.774909	1.088902	2.100442
11	6	-2.215077	0.159647	3.174234
12	6	-2.366775	-1.266928	2.666404
13	6	-1.448008	-2.906313	1.151495
14	6	-2.157876	-2.331282	-0.064249
15	8	-1.870066	-1.142154	-0.425364
16	6	2.920195	-1.447123	1.197096
17	6	2.386645	-2.116108	-0.059983
18	8	1.201356	-1.836045	-0.441687
19	6	1.453549	2.889234	1.223535
20	6	2.159239	2.325087	-0.002580
21	8	1.900550	1.130122	-0.355741
22	6	-2.906109	1.434943	1.208388
23	6	-2.345223	2.132220	-0.022595
24	8	-1.156012	1.859791	-0.385925
25	1	-0.239340	-1.529675	3.954490
26	1	-0.527613	-3.214999	3.551291
27	1	1.296952	-3.159214	1.866766
28	1	1.880597	-2.745204	3.472884
29	1	1.456811	-0.280100	3.974304
30	1	3.154396	-0.561977	3.621000
31	1	3.149013	1.284397	1.959124
32	1	2.682542	1.846308	3.557984
33	1	0.230845	1.459519	3.997291
34	1	0.517401	3.153556	3.630274
35	1	-1.300001	3.132476	1.940540
36	1	-1.890414	2.675244	3.532478
37	1	-1.471608	0.199624	3.973873
38	1	-3.166196	0.491028	3.613196
39	1	-3.147984	-1.309625	1.903707
40	1	-2.693893	-1.915496	3.489963
41	1	-0.526263	-3.380882	0.802720
42	1	3.410555	-0.519111	0.890551
43	1	0.534586	3.376696	0.885821
44	1	-3.395067	0.515365	0.875190
45	8	-0.014466	-0.081962	-2.036319
46	1	0.556257	-0.858474	-2.178968
47	1	-0.917110	-0.399536	-2.213175
48	7	-3.117824	2.975657	-0.667280

49	7	2.998622	3.100955	-0.648883
50	7	3.174318	-2.938803	-0.710016
51	7	-3.024125	-3.086007	-0.697061
52	1	-4.084551	3.237664	-0.389153
53	1	3.252837	4.070619	-0.374163
54	1	4.132041	-3.217173	-0.411292
55	1	-3.311485	-4.044414	-0.408833
56	6	-3.778318	-2.763508	-1.902092
57	1	-3.106692	-2.584049	-2.747834
58	6	-2.816419	3.690978	-1.900899
59	1	-2.584043	2.991094	-2.709690
60	6	3.726603	2.795998	-1.874152
61	1	3.035311	2.557383	-2.688434
62	6	2.901256	-3.638344	-1.959424
63	1	2.722241	-2.928872	-2.773836
64	1	-4.385668	-1.865612	-1.751902
65	1	-1.954214	4.351864	-1.766008
66	1	4.389241	1.936842	-1.728691
67	1	2.018076	-4.277342	-1.862549
68	6	-4.097537	4.524717	-2.242920
69	8	-5.047644	4.394442	-1.409238
70	8	-4.037531	5.213772	-3.282767
71	6	4.559372	4.078157	-2.214685
72	8	4.416354	5.032890	-1.388445
73	8	5.260555	4.014345	-3.246223
74	6	4.176466	-4.499737	-2.256851
75	8	4.138078	-5.189222	-3.296655
76	8	5.099536	-4.384866	-1.391039
77	6	-4.690123	-4.006972	-2.182930
78	8	-4.553057	-4.955751	-1.348572
79	8	-5.435199	-3.923094	-3.180926
80	68	0.010093	0.012507	0.450496
81	1	-2.059709	-3.669117	1.650864
82	1	3.662660	-2.078340	1.703083
83	1	2.072762	3.638574	1.734203
84	1	-3.654230	2.056705	1.717744

E(RTPSSh) = -2392.2309506 Hartree
Zero-point correction = 0.669633 Hartree/particle
Sum of electronic and thermal Energies = -2391.514239 Hartree
Sum of electronic and thermal Enthalpies = -2391.513295 Hartree
Sum of electronic and thermal Free Energies = -2391.646308 Hartree

Table S84. [Tm(DOTA-(gly)$_4$(H$_2$O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.094948	-1.790110	2.069052
2	6	-0.179157	-2.252268	3.145838
3	6	1.254368	-2.387702	2.655258
4	7	1.778894	-1.110419	2.086756
5	6	2.207304	-0.205857	3.187018
6	6	2.354733	1.233952	2.717183
7	7	1.092691	1.763799	2.118739
8	6	0.171369	2.205055	3.199261
9	6	-1.260601	2.346582	2.705219
10	7	-1.778673	1.081189	2.104289
11	6	-2.217065	0.152297	3.178860
12	6	-2.361143	-1.275122	2.671881

13	6	-1.432837	-2.907176	1.155971
14	6	-2.140141	-2.331519	-0.060669
15	8	-1.858980	-1.138341	-0.413696
16	6	2.920858	-1.430888	1.199701
17	6	2.384181	-2.099588	-0.055926
18	8	1.194702	-1.825643	-0.429235
19	6	1.436863	2.892533	1.222172
20	6	2.141090	2.324673	-0.002806
21	8	1.887803	1.125757	-0.346299
22	6	-2.909104	1.421110	1.209075
23	6	-2.345543	2.115719	-0.021891
24	8	-1.152708	1.848008	-0.376833
25	1	-0.233673	-1.527568	3.961715
26	1	-0.514754	-3.213995	3.557485
27	1	1.310740	-3.150830	1.875220
28	1	1.892783	-2.730931	3.480347
29	1	1.458673	-0.269828	3.980257
30	1	3.156892	-0.544498	3.623722
31	1	3.141012	1.300934	1.961825
32	1	2.671507	1.862808	3.559794
33	1	0.223711	1.465168	4.001676
34	1	0.502866	3.159628	3.630332
35	1	-1.315902	3.127654	1.943171
36	1	-1.904162	2.667791	3.535119
37	1	-1.475186	0.196589	3.979808
38	1	-3.170416	0.480278	3.615455
39	1	-3.141893	-1.322399	1.908987
40	1	-2.684650	-1.925493	3.495399
41	1	-0.508976	-3.379106	0.809474
42	1	3.408133	-0.501427	0.892904
43	1	0.515969	3.376514	0.885010
44	1	-3.395193	0.499489	0.877598
45	8	-0.013352	-0.073194	-2.020128
46	1	0.553208	-0.852688	-2.162842
47	1	-0.917350	-0.386716	-2.196964
48	7	-3.118089	2.952881	-0.674639
49	7	2.974562	3.099952	-0.657304
50	7	3.171895	-2.916504	-0.713024
51	7	-2.997899	-3.088527	-0.702140
52	1	-4.087512	3.211647	-0.403105
53	1	3.225208	4.072524	-0.389888
54	1	4.133090	-3.190253	-0.421324
55	1	-3.279930	-4.050503	-0.420707
56	6	-3.747509	-2.764857	-1.909769
57	1	-3.072374	-2.575593	-2.750563
58	6	-2.812330	3.664986	-1.909086
59	1	-2.572358	2.963098	-2.713888
60	6	3.700618	2.789512	-1.882379
61	1	3.008187	2.542195	-2.693052
62	6	2.893997	-3.614948	-1.962020
63	1	2.705660	-2.904773	-2.773708
64	1	-4.362550	-1.872468	-1.758038
65	1	-1.953595	4.329826	-1.771487
66	1	4.367294	1.934221	-1.732601
67	1	2.014911	-4.258828	-1.860382
68	6	-4.094709	4.492360	-2.261640
69	8	-5.049112	4.361458	-1.433006
70	8	-4.031259	5.177809	-3.303672
71	6	4.527081	4.072710	-2.234138
72	8	4.382291	5.032597	-1.414236
73	8	5.225506	4.004617	-3.267289
74	6	4.171698	-4.468861	-2.270073

75	8	4.129533	-5.156774	-3.310774
76	8	5.100358	-4.350273	-1.410775
77	6	-4.648200	-4.013556	-2.203017
78	8	-4.508423	-4.966180	-1.373535
79	8	-5.388144	-3.929453	-3.204798
80	69	0.009325	0.013462	0.461491
81	1	-2.044542	-3.671871	1.652422
82	1	3.666087	-2.061228	1.702654
83	1	2.055674	3.644936	1.728753
84	1	-3.659696	2.043044	1.714530

E(RTPSSh) = -2392.8104773 Hartree
Zero-point correction = 0.669742 Hartree/particle
Sum of electronic and thermal Energies = -2392.093690 Hartree
Sum of electronic and thermal Enthalpies = -2392.092746 Hartree
Sum of electronic and thermal Free Energies = -2392.225725 Hartree

Table S85. [Yb(DOTA-(gly)$_4$(H$_2$O)]$^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.083885	-1.789656	2.078282
2	6	-0.167210	-2.245994	3.156426
3	6	1.266837	-2.373360	2.666182
4	7	1.781403	-1.093724	2.094220
5	6	2.207029	-0.185553	3.192233
6	6	2.345028	1.253705	2.718825
7	7	1.078952	1.772537	2.119406
8	6	0.156886	2.212720	3.199406
9	6	-1.275742	2.344085	2.705395
10	7	-1.784412	1.073714	2.107224
11	6	-2.219334	0.145940	3.183840
12	6	-2.354043	-1.283039	2.679518
13	6	-1.414265	-2.908574	1.165117
14	6	-2.119206	-2.334787	-0.053531
15	8	-1.847075	-1.137208	-0.398856
16	6	2.922444	-1.409223	1.204451
17	6	2.383044	-2.079720	-0.048811
18	8	1.188891	-1.813668	-0.413377
19	6	1.415859	2.899177	1.217577
20	6	2.119618	2.326917	-0.005349
21	8	1.872793	1.123500	-0.337681
22	6	-2.914841	1.404823	1.208966
23	6	-2.350045	2.096173	-0.022950
24	8	-1.153363	1.834303	-0.369359
25	1	-0.226277	-1.520635	3.971409
26	1	-0.497885	-3.209090	3.568799
27	1	1.328117	-3.138162	1.888168
28	1	1.908283	-2.709816	3.491677
29	1	1.459987	-0.252290	3.986784
30	1	3.159219	-0.518162	3.627849
31	1	3.130673	1.323829	1.963102
32	1	2.657520	1.887359	3.559390
33	1	0.213981	1.475751	4.004217
34	1	0.483646	3.170520	3.626852
35	1	-1.336068	3.123130	1.941658
36	1	-1.921850	2.662602	3.534310
37	1	-1.478746	0.196564	3.985643
38	1	-3.175109	0.469763	3.618179

39	1	-3.133987	-1.336657	1.916228
40	1	-2.673481	-1.934599	3.503661
41	1	-0.487764	-3.377427	0.821815
42	1	3.405201	-0.478010	0.896144
43	1	0.492299	3.377633	0.880127
44	1	-3.396611	0.480026	0.880345
45	8	-0.013273	-0.067125	-2.003621
46	1	0.549535	-0.849611	-2.144856
47	1	-0.918514	-0.376642	-2.180796
48	7	-3.124220	2.924455	-0.684820
49	7	2.946209	3.101688	-0.668921
50	7	3.171536	-2.890117	-0.712727
51	7	-2.965252	-3.096708	-0.704479
52	1	-4.096634	3.178965	-0.419654
53	1	3.192722	4.077577	-0.409566
54	1	4.137054	-3.157142	-0.428780
55	1	-3.239144	-4.063114	-0.430096
56	6	-3.708338	-2.775416	-1.916817
57	1	-3.028331	-2.573744	-2.750770
58	6	-2.816470	3.632330	-1.921231
59	1	-2.568973	2.927946	-2.721562
60	6	3.671621	2.785428	-1.892918
61	1	2.978960	2.529565	-2.700734
62	6	2.890064	-3.587876	-1.961342
63	1	2.689757	-2.877346	-2.769847
64	1	-4.335420	-1.891328	-1.765794
65	1	-1.962108	4.302594	-1.782864
66	1	4.342080	1.934012	-1.738151
67	1	2.017250	-4.239434	-1.854731
68	6	-4.101855	4.450769	-2.283720
69	8	-5.059517	4.317989	-1.459108
70	8	-4.037321	5.131812	-3.328528
71	6	4.492070	4.069606	-2.255184
72	8	4.344194	5.034768	-1.442032
73	8	5.189417	3.996993	-3.288726
74	6	4.172586	-4.430324	-2.281020
75	8	4.128681	-5.115560	-3.323363
76	8	5.106406	-4.306321	-1.428074
77	6	-4.591469	-4.033165	-2.224589
78	8	-4.448375	-4.988112	-1.398352
79	8	-5.322820	-3.952698	-3.232924
80	70	0.007533	0.015465	0.472145
81	1	-2.025450	-3.675053	1.659526
82	1	3.671361	-2.037241	1.704828
83	1	2.033110	3.656202	1.719151
84	1	-3.668708	2.026374	1.710023

E(RTPSSh) = -2393.393626 Hartree
Zero-point correction = 0.669830 Hartree/particle
Sum of electronic and thermal Energies = -2392.676781 Hartree
Sum of electronic and thermal Enthalpies = -2392.675837 Hartree
Sum of electronic and thermal Free Energies = -2392.808797 Hartree

Table S86. [Lu(DOTA-$(gly)_4(H_2O)]^-$ [TSAP, Δ(δδδδ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.067987	-1.793740	2.084429
2	6	-0.149079	-2.243147	3.163420

3	6	1.285747	-2.358996	2.673532
4	7	1.787871	-1.075417	2.099368
5	6	2.208673	-0.163817	3.196217
6	6	2.334064	1.275703	2.721004
7	7	1.062828	1.781357	2.121122
8	6	0.139124	2.217315	3.201277
9	6	-1.294598	2.335861	2.708070
10	7	-1.791120	1.060193	2.110885
11	6	-2.220455	0.131230	3.188523
12	6	-2.342981	-1.298962	2.685292
13	6	-1.388433	-2.914355	1.170211
14	6	-2.093159	-2.343220	-0.049561
15	8	-1.832293	-1.140903	-0.387812
16	6	2.928732	-1.382612	1.206842
17	6	2.388604	-2.055243	-0.044745
18	8	1.189220	-1.800468	-0.400704
19	6	1.389668	2.907428	1.215164
20	6	2.094244	2.334213	-0.006680
21	8	1.858454	1.125714	-0.328859
22	6	-2.922256	1.380848	1.210073
23	6	-2.358153	2.072349	-0.021818
24	8	-1.156784	1.820921	-0.359983
25	1	-0.214312	-1.517914	3.978021
26	1	-0.472751	-3.208621	3.575684
27	1	1.353864	-3.124641	1.896936
28	1	1.930603	-2.687914	3.499352
29	1	1.463354	-0.235986	3.991911
30	1	3.164127	-0.488834	3.630322
31	1	3.118954	1.351869	1.965102
32	1	2.640584	1.913762	3.560382
33	1	0.202953	1.482580	4.007618
34	1	0.459096	3.178601	3.625932
35	1	-1.362330	3.113969	1.944024
36	1	-1.943541	2.648628	3.536900
37	1	-1.481113	0.188865	3.990995
38	1	-3.179202	0.448395	3.621134
39	1	-3.122370	-1.359900	1.921999
40	1	-2.656356	-1.953257	3.509545
41	1	-0.458217	-3.377269	0.829112
42	1	3.405007	-0.448351	0.897902
43	1	0.462275	3.378518	0.878101
44	1	-3.397287	0.452092	0.883032
45	8	-0.012671	-0.060316	-1.989801
46	1	0.553447	-0.840385	-2.130720
47	1	-0.916926	-0.374533	-2.163642
48	7	-3.136862	2.889919	-0.691425
49	7	2.910346	3.112472	-0.678827
50	7	3.180652	-2.856128	-0.715723
51	7	-2.927215	-3.111225	-0.708526
52	1	-4.113110	3.135717	-0.431935
53	1	3.147688	4.092577	-0.426692
54	1	4.150482	-3.114322	-0.438241
55	1	-3.191203	-4.082106	-0.440114
56	6	-3.666865	-2.793662	-1.923972
57	1	-2.984438	-2.579300	-2.752753
58	6	-2.830697	3.596082	-1.929232
59	1	-2.572759	2.891151	-2.725773
60	6	3.635127	2.795992	-1.903167
61	1	2.942652	2.528058	-2.707209
62	6	2.898450	-3.554146	-1.964028
63	1	2.686819	-2.843967	-2.769977
64	1	-4.306476	-1.918641	-1.772772

65	1	-1.983834	4.275558	-1.789869
66	1	4.314944	1.952619	-1.745318
67	1	2.032563	-4.214238	-1.853699
68	6	-4.122836	4.399956	-2.300451
69	8	-5.082614	4.260830	-1.479352
70	8	-4.060757	5.077442	-3.347697
71	6	4.441112	4.086403	-2.275881
72	8	4.285808	5.054976	-1.468200
73	8	5.136053	4.014590	-3.311082
74	6	4.187178	-4.383576	-2.292895
75	8	4.143583	-5.067360	-3.336183
76	8	5.124791	-4.252108	-1.445265
77	6	-4.531573	-4.061333	-2.243311
78	8	-4.381690	-5.018113	-1.420441
79	8	-5.257237	-3.985444	-3.256073
80	71	0.007043	0.016729	0.481936
81	1	-1.996696	-3.684779	1.662128
82	1	3.682688	-2.006948	1.704267
83	1	2.003535	3.670053	1.712406
84	1	-3.681081	1.999187	1.707624

E(RTPSSh) = -2393.9470345 Hartree
Zero-point correction = 0.669892 Hartree/particle
Sum of electronic and thermal Energies = -2393.230156 Hartree
Sum of electronic and thermal Enthalpies = -2393.229212 Hartree
Sum of electronic and thermal Free Energies = -2393.362131 Hartree

Table S87. $[Gd(DOTA)(H_2O)]^-$ [SAP, Δ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.026330	-1.769162	1.187635
2	6	0.056250	-2.581535	1.806477
3	6	1.243719	-1.728570	2.240553
4	7	1.864260	-0.976961	1.119874
5	6	2.704379	0.123900	1.665822
6	6	1.870627	1.325230	2.101333
7	7	1.064280	1.910242	0.999650
8	6	-0.002327	2.777378	1.568951
9	6	-1.180564	1.967246	2.102761
10	7	-1.825852	1.117414	1.068737
11	6	-2.652467	0.071008	1.728353
12	6	-1.807235	-1.086739	2.251649
13	6	-1.908110	-2.647558	0.377537
14	6	-2.707827	-1.922430	-0.719401
15	8	-2.133600	-0.869067	-1.221976
16	8	-3.782403	-2.409035	-1.086221
17	6	2.696566	-1.878571	0.282762
18	6	1.921341	-2.690721	-0.770707
19	8	2.418335	-3.744602	-1.180576
20	8	0.815921	-2.151553	-1.192590
21	6	1.926612	2.696631	0.080150
22	6	2.683166	1.847463	-0.961146
23	8	3.754191	2.281523	-1.404434
24	8	2.088796	0.756891	-1.328889
25	6	-2.671427	1.944146	0.169313
26	6	-1.890440	2.662638	-0.948611
27	8	-2.348645	3.721060	-1.397664
28	8	-0.828255	2.049977	-1.366347

29	1	-1.260522	-3.350794	-0.155545
30	1	-2.593294	-3.223465	1.014261
31	1	3.296304	-2.559461	0.902014
32	1	3.379776	-1.245845	-0.291671
33	1	2.638405	3.322061	0.635878
34	1	1.267536	3.351687	-0.497629
35	1	-3.360514	1.264760	-0.341459
36	1	-3.262633	2.675479	0.736935
37	1	0.379198	-3.319627	1.070558
38	1	-0.319904	-3.136596	2.679304
39	1	1.990195	-2.378648	2.723283
40	1	0.925124	-1.006137	2.997044
41	1	3.297594	-0.224743	2.524853
42	1	3.409816	0.423613	0.889319
43	1	2.541952	2.087049	2.527776
44	1	1.185837	1.030683	2.901512
45	1	0.393524	3.403610	2.382944
46	1	-0.341250	3.451478	0.780752
47	1	-0.845121	1.315824	2.914437
48	1	-1.917487	2.658648	2.540494
49	1	-3.373234	-0.296174	0.995858
50	1	-3.228915	0.494577	2.564789
51	1	-1.103964	-0.721471	3.005248
52	1	-2.466811	-1.806614	2.761124
53	64	0.002269	0.009593	-0.547794
54	1	-1.176955	-0.820271	-2.853689
55	8	-0.242384	-0.611968	-3.071175
56	1	0.236391	-1.437215	-2.836619

E(RTPSSh) = -1557.8519089 Hartree
Zero-point correction = 0.449472 Hartree/particle
Sum of electronic and thermal Energies = -1557.373114 Hartree
Sum of electronic and thermal Enthalpies = -1557.372169 Hartree
Sum of electronic and thermal Free Energies = -1557.459884 Hartree

Table S88. $[Gd(DOTA)(H_2O)]^-$ [TSAP, Λ(λλλλ)], TPSSh/LCRECP/6-31G(d,p), aqueous solution (0 Imaginary Frequencies)

Center Number	Atomic Number	Coordinates (Angstroms) X	Y	Z
1	7	-1.173496	-1.691745	1.253317
2	6	-1.986231	-0.999227	2.282347
3	6	-2.725706	0.205261	1.711720
4	7	-1.806076	1.203393	1.104498
5	6	-1.194786	2.048947	2.157233
6	6	0.048498	2.774014	1.655489
7	7	1.089136	1.839993	1.149318
8	6	1.847117	1.250175	2.278213
9	6	2.614155	0.002018	1.857706
10	7	1.726035	-1.053283	1.302807
11	6	1.050392	-1.787829	2.399123
12	6	-0.160890	-2.566778	1.900202
13	6	-2.022691	-2.525586	0.371256
14	6	-1.277493	-2.974658	-0.899744
15	8	-0.211041	-2.285095	-1.192152
16	6	-2.569512	2.015532	0.127704
17	6	-2.931621	1.195310	-1.128840
18	8	-2.198400	0.143971	-1.333166
19	6	1.982021	2.584816	0.230445
20	6	1.274726	2.921117	-1.098552

21	8	0.256132	2.170827	-1.388405
22	6	2.544117	-1.969595	0.475423
23	6	2.985336	-1.318740	-0.847758
24	8	2.276232	-0.292172	-1.224038
25	1	-1.313979	-0.679910	3.084018
26	1	-2.713313	-1.688890	2.737967
27	1	-3.414685	-0.122727	0.930300
28	1	-3.332877	0.669976	2.503113
29	1	-0.936705	1.402037	3.000847
30	1	-1.916846	2.788018	2.536990
31	1	-0.221603	3.438716	0.831842
32	1	0.454072	3.405369	2.460383
33	1	1.135132	1.003354	3.071206
34	1	2.550859	1.981823	2.703899
35	1	3.338393	0.260066	1.081726
36	1	3.184457	-0.383781	2.716196
37	1	0.742103	-1.058890	3.154022
38	1	1.747523	-2.479767	2.896168
39	1	0.155045	-3.303714	1.158384
40	1	-0.608268	-3.123069	2.737844
41	1	-2.413619	-3.405683	0.901608
42	1	-2.872317	-1.925621	0.034225
43	1	-3.481814	2.434415	0.575573
44	1	-1.939274	2.844634	-0.206708
45	1	2.360950	3.506215	0.694762
46	1	2.837669	1.950452	-0.018088
47	1	3.425683	-2.331108	1.023617
48	1	1.933850	-2.835825	0.204207
49	8	0.367032	-0.486098	-3.045638
50	1	0.131239	-1.409371	-2.802585
51	1	1.322926	-0.430163	-2.824388
52	64	-0.004957	0.009227	-0.482257
53	8	-1.743366	-3.908090	-1.558656
54	8	3.926239	-1.819853	-1.469661
55	8	1.733431	3.836284	-1.791503
56	8	-3.850154	1.600945	-1.849785

E(RTPSSh) = -1557.8543614 Hartree
Zero-point correction = 0.449268 Hartree/particle
Sum of electronic and thermal Energies = -1557.375798 Hartree
Sum of electronic and thermal Enthalpies = -1557.374854 Hartree
Sum of electronic and thermal Free Energies = -1557.462228 Hartree

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