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Theoretical Insights into Covalency Driven *f* Element Separations

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S2. Optimized Geometries in Cartesian Coordinates for $[M^{III}(DTPA)(H_2O)]^{2-}$ (M = Nd, Am).

Table S1. Gas Phase Optimized Geometry of $[Nd^{III}(DTPA)(H_2O)]^{2-}$ in Cartesian Coordinates.

Atom	X	Y	Z
O	-1.95375	-1.40462	-1.53828
O	1.675393	-2.01998	-1.03563
O	0.192477	-1.28762	-3.12695
O	-0.1577	-1.52256	1.420855
O	-1.09651	-2.28146	3.325853
O	-1.23699	1.74724	-1.22502
O	-1.89193	3.890577	-0.94753
O	1.713847	1.352866	-1.32641
O	3.634089	2.528965	-1.15276
N	-2.42706	-0.08982	0.813373
N	0.036108	1.766474	1.267092
N	2.378584	-0.20064	0.881364
C	-3.19914	-1.14537	-1.32466
C	-3.47703	-0.10168	-0.21722
C	-2.40739	1.143284	1.612037
C	-1.04184	1.473332	2.235872
C	1.349947	1.819392	1.954524
C	2.031716	0.458502	2.147798
C	2.759879	-1.6126	1.062393
C	2.600153	-2.43336	-0.23847
C	-2.50795	-1.29028	1.658267
C	-1.14485	-1.73963	2.208847
C	-0.23129	3.025214	0.53285

C	-1.22136	2.889507	-0.64725
C	3.417318	0.525637	0.129753
C	2.885677	1.580001	-0.86315
H	-0.69436	-1.66166	-2.95989
H	0.832221	-1.91586	-2.73794
O	-4.16515	-1.62102	-1.93559
O	3.329449	-3.42088	-0.40381
H	-2.87437	-2.12165	1.041775
H	-3.13509	1.080052	2.447822
H	-0.71512	0.644683	2.872013
H	-2.73338	1.972112	0.977563
H	-1.18926	2.351701	2.899427
H	-0.58958	3.817127	1.214686
H	0.707774	3.359982	0.077411
H	1.244549	2.295179	2.9501
H	2.006832	2.471081	1.369437
H	1.378891	-0.22261	2.702807
H	2.939201	0.635066	2.765733
H	2.081867	-2.06429	1.79476
H	3.798577	-1.71587	1.423396
H	3.969925	-0.2003	-0.48127
H	4.149212	0.999115	0.808383
H	-3.22015	-1.1529	2.491912
H	-4.47892	-0.29528	0.206893
H	-3.488	0.876134	-0.71416
Nd	0.022335	-0.22831	-0.64448

Table S2. Gas Phase Optimized Geometry of $[Am^{III}(DTPA)(H_2O)]^{2-}$ in Cartesian Coordinates.

Atom	X	Y	Z
O	-1.85084	-1.46006	-1.65927
O	1.846064	-1.64378	-1.45508
O	0.119903	-0.88464	-3.34729
O	0.203707	-1.70294	1.141288
O	-0.44656	-2.8265	2.984686
O	-1.55641	1.626285	-0.97654
O	-2.47883	3.60196	-0.39516
O	1.43927	1.753931	-1.33468
O	3.185688	3.169456	-1.10934
N	-2.29879	-0.57055	0.896367
N	-0.07882	1.561643	1.398931
N	2.48741	0.048935	0.602664
C	-3.0939	-1.43667	-1.31852

C	-3.42093	-0.61129	-0.05157
C	-2.37125	0.543282	1.848029
C	-1.00999	0.997214	2.400627
C	1.269044	1.743007	1.993179
C	2.156349	0.492677	1.964179
C	3.0711	-1.30447	0.57703
C	2.900675	-1.98998	-0.79671
C	-2.12765	-1.86372	1.569824
C	-0.66994	-2.16341	1.957075
C	-0.59241	2.839488	0.852066
C	-1.6502	2.687267	-0.2639
C	3.351309	1.004707	-0.11259
C	2.599886	2.090289	-0.91423
H	-0.70543	-1.37682	-3.16559
H	0.854635	-1.47726	-3.09842
O	-4.03142	-1.97703	-1.92123
O	3.743844	-2.82474	-1.15022
H	-2.42474	-2.65761	0.871719
H	-2.99628	0.272985	2.725429
H	-0.5122	0.157232	2.895082
H	-2.87014	1.385355	1.359686
H	-1.21242	1.759516	3.182445
H	-1.00218	3.481179	1.652326
H	0.243078	3.368227	0.378938
H	1.182553	2.079563	3.045573
H	1.768191	2.552257	1.45063
H	1.657542	-0.34192	2.466931
H	3.077541	0.732539	2.538553
H	2.530585	-1.92887	1.29712
H	4.143182	-1.29664	0.841708
H	3.939783	0.446898	-0.85328
H	4.069739	1.489049	0.572622
H	-2.77268	-1.94662	2.4639
H	-4.34219	-1.01867	0.403533
H	-3.6226	0.40954	-0.39828
Am	0.042375	-0.1526	-0.75175

S3. Optimized Geometries in Cartesian Coordinates for $[M^{III}(H_2O)_9]^{3+}$ (M = Nd, Am), $H_9O_4^+$, and H_8O_4.

Table S3. Gas Phase Optimized Geometry of $[H_9O_4]^+$ in Cartesian Coordinates.

Atom	X	Y	Z
O	1.552533	1.543625	-0.43679
H	2.532389	1.438576	-0.19238
H	1.142866	2.430194	-0.15915
H	0.9782	0.73258	-0.22855
O	0.577327	3.828566	0.168836
H	0.325189	4.463172	-0.51499
O	4.033713	1.20563	0.081581
H	4.50907	1.412377	0.896686
O	0.03237	-0.4666	-0.00584
H	0.001728	-1.03917	0.771554
H	-0.36672	-0.95845	-0.73574
H	4.69125	1.154147	-0.62474
H	0.12152	4.089643	0.979517

Table S4. Gas Phase Optimized Geometry of H_8O_4 in Cartesian Coordinates.

Atom	X	Y	Z
O	1.213085	2.154085	0.090995
H	2.084285	1.999074	0.494636
H	0.678415	1.389262	0.371149
O	3.849709	0.919505	0.184668
H	4.790905	1.120625	0.139738
O	0.127722	-0.52025	-0.1366
H	0.35761	-1.31067	0.365277
H	0.746327	-0.49642	-0.88775
H	3.577252	0.696461	-0.72293
H	1.662968	1.376418	-1.45067
O	2.02498	0.636338	-1.99867
H	1.920114	0.89193	-2.92172

Table S5. Gas Phase Optimized Geometry of $[Nd(H_2O)_9]^{3+}$ in Cartesian Coordinates.

Atom	X	Y	Z
Nd	0.00178	-0.00028	-0.00489
O	1.94024	0.8637	1.41903
O	0.09479	2.56191	-0.07867
O	-1.76101	1.00114	1.53863
O	-1.83541	0.90179	-1.51639
O	-1.92302	-1.6774	0.11362
O	1.74039	-1.85911	0.03409

O	0.01289	-1.18136	2.28232
O	1.83177	0.75344	-1.59903
H	2.04035	1.76852	1.75513
H	2.7368	0.39145	1.7089
H	-2.71631	-1.64431	0.67133
H	-2.02766	-2.47065	-0.4356
H	1.76983	-2.59183	0.66966
H	2.52542	-1.97063	-0.52538
H	1.86392	0.59136	-2.55535
H	2.66054	1.21071	-1.38575
H	-2.6628	0.47056	-1.78293
H	-1.86731	1.78857	-1.90929
H	0.6368	3.11579	-0.66245
H	-0.40838	3.18148	0.47283
H	-0.55521	-1.9132	2.5705
H	0.58996	-0.98896	3.03815
H	-1.78903	0.90415	2.50373
H	-2.55289	1.51133	1.30604
O	-0.11019	-1.3627	-2.17006
H	-0.65417	-1.17624	-2.95149
H	0.3796	-2.1727	-2.38291

Table S6. Gas Phase Optimized Geometry of $[Am(H_2O)_9]^{3+}$ in Cartesian Coordinates.

Atom	X	Y	Z
Am	0.000356	-0.0191	0.007214
O	-0.03353	-1.546	-2.04886
O	-1.7518	0.857294	-1.61169
O	1.866466	0.659676	-1.62041
O	1.803719	-1.79578	0.195208
O	1.830827	0.927378	1.494312
O	0.138436	2.566965	-0.21954
O	-1.98585	-1.60288	0.091063
O	-0.1181	-1.20813	2.272703
O	-1.75015	1.114046	1.502792
H	1.918294	0.421078	-2.55954
H	2.675132	1.16419	-1.43859
H	2.612664	0.476512	1.850336
H	1.911978	1.852075	1.776963
H	2.620863	-1.8722	-0.32236
H	1.828524	-2.52167	0.838572
H	0.484958	-2.35577	-2.17788

H	-0.54624	-1.43039	-2.8645
H	0.692693	3.072347	-0.83457
H	-0.34759	3.231467	0.293419
H	-2.58103	0.436533	-1.88833
H	-1.74503	1.728129	-2.03961
H	-1.81068	1.035685	2.468043
H	-2.50405	1.666471	1.242146
H	-0.72402	-1.92603	2.514687
H	0.40808	-1.02984	3.068205
H	-2.09471	-2.41259	-0.4324
H	-2.80101	-1.51104	0.609292

S4. Calculated IR and Raman Intensities for $[M^{III}(DTPA)(H_2O)]^{2-}$ (M = Nd, Am).

Table S7. Calculated IR and Raman Frequencies for $[Nd^{III}(DTPA)(H_2O)]^{2-}$

Frequency (cm-1)	IR Intensity	Raman Intensity
46.8	1.6771	0.5166
53.1	5.6281	0.8308
54.74	6.7204	1.441
60.01	6.2869	2.4217
71.97	2.4411	1.2956
78.25	6.693	0.7776
88.97	1.459	1.0138
103.58	2.8792	0.2588
107.77	3.9136	0.5734
113.14	2.4983	0.3906
124.52	2.4247	0.5912
133.21	5.0335	0.6318
139.96	2.0922	0.6333
146.6	6.8715	0.5053
154.43	5.2941	0.4223
157.64	13.3259	0.338
160.93	4.4443	0.4262
176.5	31.502	1.2021
182.82	4.3591	0.2645
192.06	32.4987	0.4396
198.45	21.6327	2.8906
204.85	12.2524	0.4793
216.19	120.8371	0.3173
222.61	39.3729	0.3872
229.67	98.6749	1.1196
236.35	2.1505	0.6287
251.81	12.2641	2.2849
267.59	4.6562	0.4568
275.62	1.821	0.9212
332.94	6.0646	4.238
341.16	3.5641	1.223
355.38	20.4413	1.4584
367.28	25.2407	1.0103
379.18	7.2352	8.5167
385.32	3.5163	8.3547
424.65	4.4426	1.4658
432.29	3.8831	0.2157
436.98	8.6788	2.5984
447.93	7.2001	6.1937

458.99	9.1868	1.1147
464.75	6.1765	0.7035
499.46	73.6089	0.1128
508.87	10.2656	0.3685
531.28	7.423	1.2769
557.52	5.2045	0.6946
573.94	13.0479	0.215
596.4	9.3539	0.3016
609.48	16.6006	0.4913
616.75	7.3008	0.9332
645.5	9.5194	0.5701
656.08	7.8023	0.8959
705.42	3.744	0.9369
707.18	6.8928	1.2615
712.81	17.3238	1.2961
730.79	26.7928	0.8528
735.56	167.4993	3.2358
739.65	244.2024	2.9348
757.72	129.8491	3.7792
805.53	17.0142	1.8964
846.05	16.0238	2.9759
859.76	56.0888	2.0503
897.99	10.3132	5.0201
920.28	48.9702	4.5984
921.19	41.2985	13.1861
924.31	54.3128	0.6885
929.89	18.7633	5.2908
931.81	25.9783	1.8202
966.62	12.6944	3.7831
975.64	15.7467	2.7903
979.55	14.8395	3.9285
987.49	15.5291	2.5315
1001.89	11.4162	0.9181
1005.11	26.7007	0.4036
1015.89	30.7323	3.8546
1035.17	9.7005	4.3418
1061.62	11.7595	1.3827
1064.46	1.2093	6.5848
1109.19	63.6204	2.1475
1117.67	25.2549	2.4049
1137.95	14.3402	4.1469
1142.31	13.8648	3.4425
1172.75	3.6416	4.4719

1196.66	1.8731	2.4907
1229.05	14.1672	0.932
1248.86	3.9538	5.7715
1255.22	31.8568	5.7292
1257.89	1.5684	5.2238
1263.83	20.7001	3.9946
1284.57	8.4879	3.9085
1307.29	4.3001	1.6689
1312.57	5.5475	6.0141
1321.9	18.7112	3.895
1325.9	41.3552	3.7089
1327.09	19.4215	4.8928
1338.64	7.8803	5.4272
1341.39	18.4226	8.2681
1342.78	38.6783	12.5161
1362.2	195.6571	8.8738
1364.34	22.8013	5.6594
1369.59	95.2187	15.2222
1383.21	8.7695	1.385
1384.97	452.1817	13.5561
1392.15	155.6258	13.0511
1396.12	51.917	7.3825
1416.71	20.101	1.4684
1422.72	11.5436	2.4633
1448.92	1.8952	4.2358
1451.87	1.3312	4.8429
1453.85	0.6463	3.282
1455.73	0.1707	8.1571
1459.35	0.1512	9.3996
1475.74	2.0306	3.9998
1482.61	8.8142	14.3958
1490.01	6.8261	8.6129
1492.39	9.5172	3.643
1628.28	37.9769	1.8089
1654.25	224.1382	7.1209
1659.04	619.1469	7.1998
1665.11	1546.909	6.4423
1666.45	1489.418	19.6266
1690.69	7.3313	15.7095
2899.47	87.4048	122.7267
2909.16	9.0204	22.5233
2926.17	142.2715	177.1362
2941.03	166.2224	216.3004

2973.31	24.2311	40.0097
2978.41	18.6203	38.3312
2980.16	103.645	98.6705
2981.76	244.3829	327.251
2987.02	160.7533	379.2539
3080.31	9.2645	52.1993
3081.83	9.0435	47.6367
3096.55	1.7971	15.8012
3098.23	2.6755	17.451
3104.31	1.0779	34.8214
3107.27	0.6257	99.6403
3113.58	3.5904	47.9496
3118.67	10.9371	14.3786
3126.07	14.2553	7.9652
3572.37	131.3989	93.6342
3648.45	389.3191	27.5998

Table S8. Calculated IR and Raman Frequencies for $[Am^{III}(DTPA)(H_2O)]^{2-}$

Frequency	IR Intensity	Raman Intensity
47.05	2.1343	0.5894
51.07	5.8283	0.5147
53.87	9.8418	1.6835
59.62	5.9812	1.8094
65.99	1.4964	1.6248
76.1	8.225	1.4122
86.03	0.2629	0.8397
101.75	4.703	0.4962
105.88	1.4876	0.5397
108.59	10.1356	0.2642
121.9	6.1307	0.3643
123.89	4.4052	0.7318
138.96	4.347	0.4305
143.59	29.5679	0.9994
144.3	5.3346	0.285
148.33	12.3456	0.3447
158.58	3.8134	0.5121
170.57	14.747	0.6113
177.14	16.4381	0.8667
189.65	37.2606	1.2749
192.92	23.1408	0.5037
198.7	8.4987	0.4076
208.37	95.1608	0.8818

219.5	43.775	0.8336
221.93	33.6574	1.5716
236.13	2.9775	0.7791
252.75	7.2578	2.8502
268.96	3.6603	0.5271
273.57	2.0769	1.2069
333.02	5.1734	3.6322
340.95	2.9566	1.1563
354.48	18.8963	1.4377
366.77	23.1009	0.9799
378.42	6.9988	7.6484
386.01	3.4472	8.4702
424.55	2.9721	1.3851
431.38	2.8603	0.1564
435.82	9.709	2.1692
446.09	6.9643	5.6496
458.47	9.3403	0.8044
464.6	7.0722	0.8058
497.95	70.4224	0.1226
508.87	10.1442	0.4141
530.83	7.3024	1.2039
555.57	5.7466	0.7935
573.19	12.87	0.255
596.05	9.5016	0.2812
609.17	15.9285	0.5008
616.03	7.0323	0.8764
645.29	8.5765	0.5454
656.19	7.9795	0.8231
704.55	13.6583	0.7609
707.69	4.8045	1.5192
713.05	26.1058	1.283
727.96	314.0513	5.6673
731.54	66.0577	1.3854
737.36	39.5916	1.3319
753.77	137.2855	4.3627
804.95	19.238	1.8811
845.68	18.5121	3.1798
859.51	55.078	1.8977
898.82	10.0971	4.8893
918.73	40.7181	5.5709
921.73	41.7268	14.1416
925.67	57.1659	0.6439
929.58	23.3009	6.004

932.15	23.2711	1.655
966.14	14.2552	4.1192
975.39	16.53	2.735
980.16	13.4391	4.6205
987.28	15.8666	2.0306
1001.94	10.1389	0.8779
1005.49	26.9183	0.4
1014.56	32.19	3.8143
1034.92	10.5181	4.4926
1061.8	12.5671	0.8551
1065.17	1.4443	7.1479
1110.01	61.858	2.0482
1117.3	23.7661	2.5191
1137.5	15.2146	4.3074
1145.81	13.6185	3.4589
1173.38	4.0747	4.5368
1196.8	1.9955	2.2592
1229.23	14.1091	1.1251
1249.3	4.2942	5.9261
1257.04	33.4834	6.0098
1258.32	2.6772	5.4952
1262.86	18.2889	4.2423
1284.73	8.4363	4.2819
1308.29	3.4942	1.7543
1312.58	5.8699	6.6492
1321.69	18.3174	4.6109
1326.03	47.4765	4.0373
1326.74	12.1656	5.2328
1339.41	5.7884	7.7618
1340.91	23.7595	6.5071
1343.04	33.0111	12.8991
1362.99	195.8973	12.037
1364.76	34.8695	6.1183
1369.49	109.5817	13.7146
1383.41	7.9769	1.2958
1384.12	398.9305	17.6321
1393.23	165.569	13.8007
1396.56	71.4168	8.4001
1416.28	18	1.5936
1423.46	12.6185	2.5148
1448.56	2.1262	3.9102
1451.7	1.3903	4.9206
1454.01	0.5582	3.4629

1455.5	0.2337	8.2117
1459.18	0.1527	9.6044
1475.35	2.4196	4.8743
1482.46	8.3134	13.3876
1489.28	6.4562	9.6154
1491.58	8.8619	2.5368
1626.65	34.5835	1.9189
1653.86	204.6204	6.8735
1657.01	645.7925	5.9455
1663.14	1424.005	14.5212
1666.51	1686.306	12.1361
1690.25	3.2032	16.6129
2901.97	88.1799	125.4524
2906.06	12.9352	31.3464
2922.57	152.5763	188.949
2943.03	160.0366	209.2034
2967.94	28.5437	45.8622
2976.76	210.5948	291.7828
2979.56	20.382	41.615
2982.01	134.0069	178.5775
2988.57	172.1445	356.2166
3078.91	9.2367	51.2206
3079.9	9.3151	46.8394
3097.26	1.7113	12.679
3098.83	2.6471	13.4703
3104.12	0.5336	35.11
3107.37	0.368	94.4565
3111.75	1.9633	66.4638
3118.74	10.4508	6.4195
3124.44	16.6723	4.1266
3568.66	148.3387	88.4267
3652.86	352.0463	26.8171

S5. Mulliken Population Analysis of the M, N, and O atoms for $[M^{III}(DTPA)(H_2O)]^{2-}$ (M = Nd, Am).

Table S9. Mulliken orbital population of the Nd, N and O atoms for $[Nd^{III}(DTPA)(H_2O)]^{2-}$. The atomic labels can be found in Figure 1. The O population analysis only refers to atoms bonded to Nd. Only the MOs which contain 4*f* occupations > 5% are shown. MO energies are in Hartrees.

mo no. 105	eig	-0.10258			
iatm, sym,	total pop,	s	p	d	f
1 O	0.01	0	0.01	0	
2 O	0.006	0	0.005	0	
3 O	0.003	0	0.003	0	
4 O	0.044	0.005	0.039	0	
5 O	0.005	0	0.005	0	
6 O	0.007	0.001	0.006	0	
1 N	0.003	0	0.002	0	
2 N	0.03	0.001	0.029	0	
3 N	0.003	0	0.003	0	
1 Nd	0.817	-0.003	-0.001	0.003	0.817
mo no. 106	eig	-0.1005			
iatm, sym,	total pop,	s	p	d	f
1 O	0.038	0.002	0.036	0	
2 O	0.013	0.001	0.012	0	
3 O	0.002	0	0.015	0	
4 O	0.015	0	0.015	0	
5 O	0.01	0.002	0.009	0	
6 O	0.006	0	0.006	0	
1 N	0.005	0.001	0.004	0	
2 N	0	0	0	0	
3 N	0.002	0	0.002	0	
1 Nd	0.85	0	0	0.003	0.848
mo no. 107	eig	-0.09746			
iatm, sym,	total pop,	s	p	d	f
1 O	0.006	0	0.006	0	
2 O	0.002	0	0.002	0	
3 O	0.003	0	0.003	0	
4 O	0.009	0	0.008	0	
5 O	0.013	0	0.012	0	
6 O	0.008	0.001	0.007	0	
1 N	0.002	0	0.002	0	
2 N	0.004	0	0.003	0	

3 N	0.002	0	0.002	0		
1 Nd	0.909	0	0	0.005	0.904	

Table S10. Mulliken orbital population of the Am, N and O atoms for $[Am^{III}(DTPA)(H_2O)]^{2-}$. The atomic labels can be found in Figure 1. The O population analysis only refers to atoms bonded to Am. Only the MOs which contain 5*f* occupations > 5% are shown. MO energies are in Hartrees.

mo no. 105	eig	-0.094			
iatm, sym,	total pop,	s	p	d	f
1 O	0.016	0.001	0.015	0	
2 O	0.033	0.002	0.031	0	
3 O	0.038	0	0.038	0	
4 O	0.032	-0.001	0.033	0	
5 O	0.063	0.002	0.061	0	
6 O	0.01	0	0.01	0	
1 N	0.005	-0.001	0.005	0	
2 N	0.381	0.004	0.374	0.003	
3 N	-0.009	-0.005	-0.005	0.001	
1 Am	0.096	0.018	0.009	0.02	0.048
mo no. 106	eig	-0.088			
iatm, sym,	total pop,	s	p	d	f
1 O	0.061	0.006	0.055	0	
2 O	0.219	0.005	0.214	0	
3 O	0.032	0.003	0.029	0	
4 O	0.199	0.007	0.192	0	
5 O	0.034	-0.001	0.035	0	
6 O	0.005	-0.001	0.006	0	
1 N	0.019	0.001	0.018	0	
2 N	0.009	0.002	0.006	0.001	
3 N	0.005	-0.002	0.006	0	
1 Am	0.105	0.006	-0.005	0.027	0.077
mo no. 107	eig	-0.087			
iatm, sym,	total pop,	s	p	d	f
1 O	0.165	0.004	0.161	0	
2 O	0.085	0	0.085	0	
3 O	0.022	0.001	0.021	0	
4 O	0.06	0.001	0.059	0	
5 O	0.027	0.001	0.026	0	
6 O	0.003	0	0.003	0	
1 N	-0.004	-0.002	-0.002	0	

2	N	0.003	0	0.002	0	
3	N	0.076	-0.004	0.079	0	
1	Am	0.172	0.007	0.003	0.005	0.157

mo no. 110	eig		-0.082			
iatm, sym,		total pop,	s	p	d	f
1	O	0.122	0.005	0.116	0.001	
2	O	0.144	0	0.144	0	
3	O	0.035	0	0.036	0	
4	O	0.009	0	0.008	0	
5	O	0.083	0.002	0.081	0	
6	O	0.001	0	0.001	0	
1	N	0.002	-0.001	0.002	0	
2	N	0.005	0	0.004	0	
3	N	0.194	0.004	0.188	0.001	
1	Am	0.12	0.004	0.011	0.03	0.075

mo no. 111	eig		-0.079			
iatm, sym,		total pop,	s	p	d	f
1	O	0.013	-0.003	0.016	0	
2	O	0.157	0.005	0.152	0	
3	O	0.015	0	0.014	0	
4	O	0.051	0.003	0.048	0	
5	O	0.205	0.002	0.203	0	
6	O	0.005	0.001	0.004	0	
1	N	0.043	-0.001	0.043	0.001	
2	N	0.01	-0.006	0.015	0.001	
3	N	0.009	0	0.008	0	
1	Am	0.077	0.012	-0.004	0.011	0.059

mo no. 112	eig		-0.078			
iatm, sym,		total pop,	s	p	d	f
1	O	0.148	0.001	0.147	0	
2	O	0.057	0	0.057	0	
3	O	0.2	0.009	0.192	0	
4	O	0.016	0	0.016	0	
5	O	0.001	-0.002	0.002	0	
6	O	0.001	0	0.001	0	
1	N	0.022	-0.001	0.023	0	
2	N	0.026	0.006	0.02	0	
3	N	-0.004	0	-0.004	0	
1	Am	0.14	0.006	-0.003	0.007	0.131

mo no. 114	eig	-0.072			
iatm, sym,	total pop,	s	p	d	f
1 O	0.093	0.002	0.091	0	
2 O	0.149	0	0.149	0	
3 O	0.138	0.002	0.136	0	
4 O	0.055	0.001	0.053	0	
5 O	0.054	0.002	0.052	0	
6 O	0.001	0	0.001	0	
1 N	0.036	0.001	0.035	0	
2 N	0.001	0.001	0	0	
3 N	0.004	-0.002	0.005	0	
1 Am	0.063	0.002	0.003	0.007	0.052

mo no. 115	eig	-0.075			
iatm, sym,	total pop,	s	p	d	f
1 O	0.058	-0.001	0.059	0	
2 O	0.073	-0.002	0.074	0	
3 O	0.082	-0.003	0.085	0	
4 O	0.216	0.009	0.207	0	
5 O	0.02	0.001	0.019	0	
6 O	0.002	0	0.002	0	
1 N	0.029	-0.002	0.031	0	
2 N	0	0.001	-0.001	0	
3 N	0.087	0.009	0.077	0.001	
1 Am	0.101	0.006	0.005	0.012	0.078

mo no. 117	eig	-0.065			
iatm, sym,	total pop,	s	p	d	f
1 O	0.028	-0.001	0.028	0	
2 O	0.002	-0.002	0.004	0	
3 O	0.314	0.004	0.31	0	
4 O	0.067	0.004	0.062	0	
5 O	0.038	-0.001	0.038	0	
6 O	0.001	0	0.001	0	
1 N	0.125	0.013	0.111	0.001	
2 N	0.012	0	0.011	0	
3 N	0.049	0.01	0.039	0	
1 Am	0.096	0	0.003	0.006	0.086

mo no. 118	eig	-0.055			
iatm, sym,	total pop,	s	p	d	f
1 O	0.012	0.001	0.011	0	
2 O	0.012	0.003	0.009	0	

3 O	0.005	0	0.005	0	
4 O	0.019	0	0.019	0	
5 O	0.004	0.001	0.003	0	
6 O	0.001	0	0.001	0	
1 N	0.002	0	0.002	0	
2 N	0.001	-0.001	0.002	0	
3 N	0.002	0	0.001	0	
1 Am	0.532	0.002	-0.001	0.001	0.529

mo no. 119	eig	-0.053			
iatm, sym,	total pop,	s	p	d	f
1 O	0.047	0.013	0.033	0.001	
2 O	0.008	0	0.007	0	
3 O	0.002	0	0.002	0	
4 O	0.005	0.002	0.003	0	
5 O	0.015	0	0.015	0	
6 O	0.001	0	0.001	0	
1 N	0.038	0.008	0.028	0.002	
2 N	-0.005	0	-0.005	0	
3 N	0.002	0	0.002	0	
1 Am	0.073	-0.001	0.008	0.005	0.06

mo no. 121	eig	-0.049			
iatm, sym,	total pop,	s	p	d	f
1 O	0.013	0.005	0.007	0	
2 O	0.017	0.002	0.015	0	
3 O	0.007	0.001	0.005	0	
4 O	0.02	0	0.021	0	
5 O	0.028	0.007	0.021	0	
6 O	0.002	0	0.002	0	
1 N	0.035	0.011	0.023	0.001	
2 N	-0.002	0.001	-0.003	0	
3 N	0.006	0.001	0.003	0.001	
1 Am	0.098	-0.002	0.015	0.003	0.083

mo no. 122	eig	-0.047			
iatm, sym,	total pop,	s	p	d	f
1 O	0.003	0	0.003	0	
2 O	0.009	0	0.008	0	
3 O	0.052	0.009	0.042	0	
4 O	0.01	0.001	0.009	0	
5 O	0.015	0.002	0.013	0	
6 O	0	0	0	0	

1	N	0.002	0	0.001	0	
2	N	0.001	-0.001	0.002	0	
3	N	0.029	0.003	0.026	0.001	
1	Am	0.197	0.002	0.001	0.003	0.192

mo no. 123	eig		-0.044			
iatm, sym,		total pop,	s	p	d	f
1	O	0.006	0	0.006	0	
2	O	0.028	0.006	0.022	0	
3	O	0.014	0.001	0.013	0	
4	O	0.018	0.004	0.014	0	
5	O	0.008	0.001	0.007	0	
6	O	0.001	0	0.001	0	
1	N	0.001	0	0.001	0	
2	N	0.007	0.001	0.006	0.001	
3	N	0.027	0.007	0.019	0.001	
1	Am	0.33	0.003	0	0.006	0.321

mo no. 124	eig		-0.039			
iatm, sym,		total pop,	s	p	d	f
1	O	0.032	0.002	0.03	0	
2	O	0.016	0	0.015	0	
3	O	0.012	0	0.012	0	
4	O	0.038	0	0.037	0	
5	O	0.004	0	0.004	0	
6	O	0.003	0	0.003	0	
1	N	0.001	0	0.001	0	
2	N	0.007	0.001	0.007	0	
3	N	0.003	0	0.003	0	
1	Am	0.726	0	0.002	0.004	0.72

mo no. 125	eig		-0.028			
iatm, sym,		total pop,	s	p	d	f
1	O	0.001	0	0.001	0	
2	O	0.004	0	0.004	0	
3	O	0.061	0	0.06	0	
4	O	0.007	0	0.007	0	
5	O	0.024	0	0.023	0	
6	O	0.007	0	0.006	0	
1	N	0.004	0.001	0.002	0	
2	N	0.005	0	0.004	0	
3	N	0.001	0	0	0	
1	Am	0.85	0	0	0.002	0.849

mo no. 126	eig	-0.026			
iatm, sym,	total pop,	s	p	d	f
1 O	0.024	0.001	0.023	0	
2 O	0.008	0	0.008	0	
3 O	0.029	0.001	0.028	0	
4 O	0.006	0	0.006	0	
5 O	0.002	0	0.001	0	
6 O	0.002	0	0.002	0	
1 N	0	0	0	0	
2 N	0.019	0.003	0.016	0	
3 N	0.004	0.001	0.004	0	
1 Am	0.824	0.001	0.001	0.006	0.816

mo no. 127	eig	-0.022			
iatm, sym,	total pop,	s	p	d	f
1 O	0.031	0	0.031	0	
2 O	0.016	0	0.016	0	
3 O	0.032	0	0.032	0	
4 O	0.012	0	0.012	0	
5 O	0.015	0	0.015	0	
6 O	0	0	0	0	
1 N	0.004	0	0.004	0	
2 N	0.001	0	0.001	0	
3 N	0.032	0.004	0.028	0	
1 Am	0.78	0.002	0	0.006	0.771

mo no. 128	eig	-0.017			
iatm, sym,	total pop,	s	p	d	f
1 O	0.007	0.001	0.006	0	
2 O	0.013	0	0.012	0	
3 O	0.012	0	0.011	0	
4 O	0.025	0	0.025	0	
5 O	0.03	0	0.029	0	
6 O	0.004	0	0.004	0	
1 N	0.055	0.006	0.049	0	
2 N	0.006	0	0.006	0	
3 N	0.016	0.001	0.015	0	
1 Am	0.737	0.001	0.001	0.005	0.731

S6. AIM Approach for the M, N, and O atoms for $[M^{III}(DTPA)(H_2O)]^{2-}$ (M = Nd, Am).

Table S11. Characteristics of the Nd-N bond critical points for $[Nd^{III}(DTPA)(H_2O)]^{2-}$. The atomic labels can be found in Figure 1. The values *V* and *G* refer to the potential and kinetic energy density, respectively.

		Nd-N1	Nd-N2	Nd-N3	Average
M-N	ρ	0.0250	0.0304	0.0276	0.0277
	$\nabla^2\rho$	0.0726	0.0880	0.0798	0.0802
	ε	0.0774	0.1423	0.0415	0.0871
	V	-0.0174	-0.0221	-0.0197	-0.0197
	G	0.0178	0.0221	0.0198	0.0199
	H	0.000397	-0.000057	0.000147	0.000162

Table S12. Characteristics of the Nd-O bond critical points for $[Nd^{III}(DTPA)(H_2O)]^{2-}$. The atomic labels can be found in Figure 1. The values *V* and *G* refer to the potential and kinetic energy density, respectively.

		Nd-O1	Nd-O2	Nd-O3	Nd-O4	Nd-O5	Average
M-O	ρ	0.0493	0.0479	0.0532	0.0540	0.0476	0.0504
	$\nabla^2\rho$	0.1894	0.1785	0.2029	0.2029	0.1781	0.1904
	ε	0.0635	0.0300	0.0767	0.1041	0.0493	0.0647
	V	-0.0486	-0.0463	-0.0536	-0.0543	-0.0462	-0.0498
	G	0.0480	0.0455	0.0521	0.0525	0.0453	0.0487
	H	-0.000610	-0.000841	-0.00141	-0.00180	-0.000809	-0.00109

Table S13. Characteristics of the Am-N bond critical points for $[Am^{III}(DTPA)(H_2O)]^{2-}$. The atomic labels can be found in Figure 1. The values *V* and *G* refer to the potential and kinetic energy density, respectively.

		Am-N1	Am-N2	Am-N3	Average
M-N	ρ	0.0248	0.0330	0.0301	0.0293
	$\nabla^2\rho$	0.0737	0.0992	0.0879	0.0870
	ε	0.0077	0.0768	0.0888	0.0578
	V	-0.0190	-0.0270	-0.0236	-0.0232
	G	0.0187	0.0259	0.0228	0.0225
	H	-0.000296	-0.001085	-0.000820	-0.000734

Table S14. Characteristics of the Am-O bond critical points for $[Am^{III}(DTPA)(H_2O)]^{2-}$. The atomic labels can be found in Figure 1. The values *V* and *G* refer to the potential and kinetic energy density, respectively.

		Am-O1	Am-O2	Am-O3	Am-O4	Am-O5	Average
M-O	ρ	0.0509	0.0526	0.0542	0.0576	0.0495	0.0530
	$\nabla^2\rho$	0.2088	0.2076	0.2191	0.2306	0.1956	0.2124
	ε	0.0433	0.0856	0.1154	0.0122	0.0674	0.0648
	V	-0.0553	-0.0557	-0.0592	-0.0628	-0.0522	-0.0570
	G	0.0538	0.0538	0.0570	0.0602	0.0506	0.0551
	H	-0.001543	-0.001900	-0.002208	-0.002574	-0.001664	-0.00198

S7. NBO Bond Analysis for the M, N, and O atoms for $[M^{III}(DTPA)(H_2O)]^{2-}$ (M = Nd, Am).

Table S15. Significant Metal-Ligand interaction energies in $[M^{III}(DTPA)(H_2O)]^{2-}$ (M = Nd, Am) based on Second Order Perturbation Analysis (Alpha Spin).

L → M Interaction	Interaction energy (kcal·mol^{-1})	Metal electronic configuration
LP(1)O1 → LP*(4)Nd	4.98	99.61%s 0.01%p 0.23%d 0.13%f
LP(1)O2 → LP*(4)Nd	5.21	
LP(1)O3 → LP*(4)Nd	5.44	
LP(1)O4 → LP*(4)Nd	5.36	
LP(1)O5 → LP*(4)Nd	5.24	
LP(1)N1 → LP*(4)Nd	1.77	
LP(1)N2 → LP*(4)Nd	2.02	
LP(1)N3 → LP*(4)Nd	1.78	
LP(2)O6 → LP*(4)Nd	2.34	
LP(3)O1 → LP*(7)Am	9.65	82.52%s 2.40%p 14.92%d 0.16%f
LP(3)O2 → LP*(7)Am	7.87	
LP(3)O3 → LP*(7)Am	10.76	
LP(3)O4 → LP*(7)Am	9.87	
LP(3)O5 → LP*(7)Am	7.93	
LP(1)N1 → LP*(7)Am	0.72	
LP(1)N2 → LP*(7)Am	1.55	
LP(1)N3 → LP*(7)Am	1.02	
LP(2)O6 → LP*(7)Am	4.15	

Table S16. Significant Metal-Ligand interaction energies in $[M^{III}(DTPA)(H_2O)]^{2-}$ (M = Nd, Am) based on Second Order Perturbation Analysis (Beta Spin).

L → M Interaction	Interaction energy (kcal·mol^{-1})	Metal electronic configuration

LP(1)O1 → LP*(1)Nd	5.17	99.46%s 0.01%p 0.18%d 0.33%f
LP(1)O2 → LP*(1)Nd	5.28	
LP(1)O3 → LP*(1)Nd	5.59	
LP(1)O4 → LP*(1)Nd	5.39	
LP(1)O5 → LP*(1)Nd	5.38	
LP(1)N1 → LP*(1)Nd	1.73	
LP(1)N2 → LP*(1)Nd	1.95	
LP(1)N3 → LP*(1)Nd	1.71	
LP(2)O6 → LP*(1)Nd	2.38	
LP(3)O1 → LP*(1)Am	9.30	81.82%s 2.55%p 15.99%d 0.13%f
LP(3)O2 → LP*(1)Am	7.55	
LP(3)O3 → LP*(1)Am	10.40	
LP(3)O4 → LP*(1)Am	9.53	
LP(3)O5 → LP*(1)Am	7.60	
LP(1)N1 → LP*(1)Am	0.66	
LP(1)N2 → LP*(1)Am	1.43	
LP(1)N3 → LP*(1)Am	0.94	
LP(2)O6 → LP*(1)Am	4.04	