A. ELECTRONIC SUPPLEMENTARY INFORMATION

Approaching actinide (III) hydration from first principles.
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Electronic supplementary material includes:

- Optimized $[An^{III}(OH_2)_h]^{3+}$ ($h = 7$, 8, 9) and $[An^{III}(OH_2)_{h-1} \cdot OH_2]^{3+}$ ($h = 8$, 9) molecular structures as `.xyz` files.
- An^{III} atomic ion total energies (tab. A.1).
- Free OH_2 ligand total gas phase energy, total COSMO SCRF energy, and ZPE (tab. A.1).
- $[An^{III}(OH_2)_h]^{3+}$ ($h = 7$, 8, 9) total gas phase and total COSMO SCRF energies (tab.s A.2, A.3, A.4).
  $[An^{III}(OH_2)_{h-1} \cdot OH_2]^{3+}$ ($h = 8$, 9) total gas phase and total COSMO SCRF energies (tab.s A.5, A.6).
- $[An^{III}(OH_2)_h]^{3+}$ ($h = 7$, 8, 9) ZPEs, entropies $S°$, and mean An^{III}, O, H Mulliken net charges $q_{An^{III}}$, q_O, q_H, and An^{III} Mulliken 5f populations $n^{5f}_{An^{III}}$ (tab.s A.7, A.8, A.9).
  $[An^{III}(OH_2)_{h-1} \cdot OH_2]^{3+}$ ($h = 8$, 9) ZPEs, entropies $S°$, An^{III} Mulliken net charges and Mulliken 5f populations $n^{5f}_{An^{III}}$, and O and H Mulliken net charges q_O and q_H for the 1^{st} hydration sphere OH_2 ligands that coordinate the single 2^{nd} hydration sphere OH_2 ligand (tab.s A.10, A.11).

`.xyz` files are named according to fig. 2 in the paper and the corresponding actinide element, *e.g.* `1ac.xyz` for $[Ac^{III}(OH_2)_7]^{3+}$, `2th.xyz` for $[Th^{III}(OH_2)_8]^{3+}$, ... All `.xyz` files will be found in the `xyz.zip` archive.

Note that some structure visualization programs will not support actinide elements! In this case you will have to replace the actinide element by *e.g.* lanthanum in the corresponding `.xyz` files manually.

Table A.1.: An^{III} atomic ion total energies, in a.u.

	BP86-DFT An^{III}: [6s5p4d2f]	BP86-DFT An^{III}: [6s5p4d2f1g]	MP2 An^{III}: [6s5p4d2f]	HF An^{III}: [6s5p4d2f]
Ac	−28.2645689499	−28.2645689499	−28.0788227139	−27.9141245879
Th	−29.0092998197	−29.0092998197	−28.8250380942	−28.6588473303
Pa	−29.7229190823	−29.7229190823	−29.5378206972	−29.3723596279
U	−30.4108378387	−30.4108378382	−30.2238973250	−30.0611438983
Np	−31.0795132347	−31.0795132346	−30.8894791046	−30.7305751909
Pu	−31.7332875414	−31.7332875414	−31.5432114478	−31.3845121364
Am	−32.3742753746	−32.3742753747	−32.1897828938	−32.0258208559
Cm	−33.0041682416	−33.0041682416	−32.8267712403	−32.6568462385
Bk	−33.6293502263	−33.6293502262	−33.4551644290	−33.2804538293
Cf	−34.2449273184	−34.2449273183	−34.0748015042	−33.8971794581
Es	−34.8592452170	−34.8592452171	−34.6899850187	−34.5099333993
Fm	−35.4705453280	−35.4705453282	−35.3013732786	−35.1195520656
Md	−36.0761076952	−36.0761076952	−35.9097068240	−35.7263308929
No	−36.6833877140	−36.6833877138	−36.5176006724	−36.3329073838
Lr	−37.2904768691	−37.2904768690	−37.1257450996	−36.9398209687

free OH_2 system (ZPE: 0.0206953 a.u.)

	BP86-DFT O, H: aug-cc-pVDZ	BP86-DFT O, H: cc-pVQZ	MP2 O, H: aug-cc-pVDZ	HF O, H: aug-cc-pVDZ
		gas phase total energies		
OH_2	−76.4492498605	−76.4738516109	−76.2607966147	−76.0399725424
		COSMO total energies		
OH_2	−76.4599864235	−76.4849883218	−76.2723112995	−76.0525018468

Table A.2.: $[An^{III}(OH_2)_7]^{3+}$ total energies, in a.u.

	BP86-DFT An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ	BP86-DFT An^{III}: [6s5p4d2f1g] O, H: cc-pVQZ	MP2 An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ	HF An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ
		gas phase total energies		
Ac	−564.0322983481	−564.2193273348	−562.5421728398	−560.7962411591
Th	−564.7901369775	−564.9770777319	−563.3026583788	−561.5548250565
Pa	−565.5167103326	−565.7035330244	−564.0289169410	−562.2811187999
U	−566.2174500468	−566.4041826964	−564.7283669629	−562.9823267670
Np	−566.8983649492	−567.0850313058	−565.4067752678	−563.6635736877
Pu	−567.5637657357	−567.7503740998	−566.0714014550	−564.3287419099
Am	−568.2159859356	−568.4025702846	−566.7279508650	−564.9809711930
Cm	−568.8569971329	−569.0435845447	−567.3755703762	−565.6228035411
Bk	−569.4919574975	−569.6785502228	−568.0137734298	−566.2560372510
Cf	−570.1179990942	−570.3046065974	−568.6439515833	−566.8829842759
Es	−570.7414287924	−570.9280401708	−569.2683883965	−567.5047829612
Fm	−571.3614407115	−571.5480812412	−569.8886286038	−568.1230684160
Md	−571.9768170473	−572.1634529085	−570.5069043686	−568.7395322605
No	−572.5928684325	−572.7795084819	−571.1237582374	−569.3548422460
Lr	−573.2087530404	−573.3954004387	−571.7408600804	−569.9704467772
		COSMO total energies		
Ac	−564.6549376626	−564.8411970742	−563.1668625175	−561.4193817314
Th	−565.4154675645	−565.6016584617	−563.9301001815	−562.1807943124
Pa	−566.1438155580	−566.3299129037	−564.6581099928	−562.9088800788
U	−566.8389481717	−567.0250797639	−565.3509067933	−563.6031217389
Np	−567.5294354812	−567.7154211970	−566.0399472913	−564.2953534672
Pu	−568.1958352627	−568.3817848033	−566.7054702687	−564.9614229794
Am	−568.8489479920	−569.0348963257	−567.3627874856	−565.6144127608
Cm	−569.4910587277	−569.6770295924	−568.0113821538	−566.2572136189
Bk	−570.1269233755	−570.3129144392	−568.6503847213	−566.8912365128
Cf	−570.7545080528	−570.9405273259	−569.2820471098	−567.5196672699
Es	−571.3786071503	−571.5646456096	−569.9070366399	−568.1420046513
Fm	−571.9999381461	−572.1860164158	−570.5285680705	−568.7615815309
Md	−572.6163113301	−572.8024000208	−571.1477517871	−569.3789407820
No	−573.2333238412	−573.4194314479	−571.7654923827	−569.9951226726
Lr	−573.8502094760	−574.0363397411	−572.3835194170	−570.6116400117

Table A.3.: $[An^{III}(OH_2)_8]^{3+}$ total energies, in a.u.

	BP86-DFT An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ	BP86-DFT An^{III}: [6s5p4d2f1g] O, H: cc-pVQZ	MP2 An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ	HF An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ
			gas phase energies	
Ac	−640.5272794757	−640.7407151084	−638.8580277321	−636.8855012194
Th	−641.2853876723	−641.4987432252	−639.6189556991	−637.6443528393
Pa	−642.0120433364	−642.2253211350	−640.3456022387	−638.3709087829
U	−642.7128434547	−642.9260411744	−641.0454428623	−639.0722551629
Np	−643.3938801964	−643.6069794322	−641.7242100999	−639.7535653019
Pu	−644.0592215924	−644.2722942668	−642.3889097230	−640.4187843615
Am	−644.7114648474	−644.9240786675	−643.0452765963	−641.0709975503
Cm	−645.3523044384	−645.5653460971	−643.6927250433	−641.7127640699
Bk	−645.9872320307	−646.2002339240	−644.3308580958	−642.3458914329
Cf	−646.6131165028	−646.8261249966	−644.9609016727	−642.9725665036
Es	−647.2363393933	−647.4493731470	−645.5852596792	−643.5943005023
Fm	−647.8562226534	−648.0692452091	−646.2053571802	−644.2123719197
Md	−648.4713446842	−648.6841969582	−646.8234330126	−644.8285684761
No	−649.0871759946	−649.3000431211	−647.4400553332	−645.4435595108
Lr	−649.7027066011	−649.9157139950	−648.0569095324	−646.0588966793
			COSMO total energies	
Ac	−641.1257314971	−641.3384300510	−639.4572612526	−637.4829651990
Th	−641.8896953914	−642.1022503269	−640.2246321698	−638.2483922682
Pa	−642.6178665842	−642.8303646611	−640.9527255927	−638.9764071894
U	−643.3203352469	−643.5327715222	−641.6541789573	−639.6793753981
Np	−644.0033058132	−644.2156559420	−642.3348178035	−640.3625359616
Pu	−644.6702066622	−644.8825431686	−643.0010247485	−641.0292679959
Am	−645.3240340644	−645.5363196855	−643.6589255025	−641.6830208658
Cm	−645.9662971909	−646.1786279504	−644.3077374021	−642.3261501377
Bk	−646.6026258369	−646.8149253314	−644.9472302693	−642.9606380626
Cf	−647.2299180912	−647.4422345527	−645.5786413820	−643.5886771381
Es	−647.8544520368	−648.0668053781	−646.2042730918	−644.2116828019
Fm	−648.4755386459	−648.6878912791	−646.8255379720	−644.8309173859
Md	−649.0919875853	−649.3043513179	−647.4449019083	−645.4484040710
No	−649.7089075344	−649.9212772182	−648.0625796407	−646.0644483394
Lr	−650.3256259483	−650.5380024412	−648.6805900284	−646.6809378314

Table A.4.: $[An^{III}(OH_2)_9]^{3+}$ total energies, in a.u.

	BP86-DFT An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ	BP86-DFT An^{III}: [6s5p4d2f1g] O, H: cc-pVQZ	MP2 An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ	HF An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ
		gas phase energies		
Ac	−717.0115526199	−717.2512495626	−715.1638328626	−712.9639491789
Th	−717.7693698363	−718.0090014867	−715.9244187222	−713.7227010150
Pa	−718.4957779303	−718.7353092904	−716.6510674110	−714.4486012382
U	−719.1961775383	−719.4356210473	−717.3507103259	−715.1494562396
Np	−719.8766796229	−720.1160484072	−718.0292605315	−715.8303400366
Pu	−720.5415944316	−720.7808827417	−718.6934140219	−716.4949006223
Am	−721.1932564328	−721.4320977782	−719.3490334675	−717.1465889716
Cm	−721.8336386587	−722.0728630661	−719.9958084726	−717.7877953121
Bk	−722.4679321624	−722.7071372108	−720.6333267779	−718.4202709690
Cf	−723.0932275825	−723.3324182817	−721.2627841061	−719.0463992549
Es	−723.7159127660	−723.9551148307	−721.8864982811	−719.6673135663
Fm	−724.3351682593	−724.5743610581	−722.5059938721	−720.2847514105
Md	−724.9496451757	−725.1886925854	−723.1232906360	−720.9002979420
No	−725.5648297604	−725.8038782626	−723.7393087841	−721.5145537289
Lr	−726.1797913185	−726.4189554927	−724.3554650706	−722.1290845129
		COSMO total energies		
Ac	−717.5971755471	−717.8360638940	−715.7501509931	−713.5485479406
Th	−718.3566430468	−718.5954761485	−716.5123651555	−714.3089348127
Pa	−719.0853039330	−719.3240478379	−717.2412407177	−715.0370651513
U	−719.7875757834	−720.0262436909	−717.9427368930	−715.7397800318
Np	−720.4698386066	−720.7084430695	−718.6230272167	−716.4224109043
Pu	−721.1364578514	−721.3749933040	−719.2888747478	−717.0886702887
Am	−721.7896532557	−722.0281366666	−719.9460013717	−717.7418708198
Cm	−722.4317100785	−722.6701983799	−720.5944415768	−718.3847458650
Bk	−723.0674135328	−723.3058892270	−721.2333560331	−719.0186220370
Cf	−723.6941765513	−723.9326447364	−721.8642688916	−719.6462083083
Es	−724.3181034500	−724.5565887223	−722.4892127430	−720.2683540955
Fm	−724.9385562584	−725.1770338837	−723.1098938637	−720.8869745897
Md	−725.5541523466	−725.7926490371	−723.7282984541	−721.5036259632
No	−726.1706460732	−726.4091345407	−724.3456162039	−722.1191819386
Lr	−726.7872523595	−727.0257087308	−724.9633983654	−722.7353055051

Table A.5.: $[An^{III}(OH_2)_7 \cdot OH_2]^{3+}$ total energies, in a.u.

	BP86-DFT An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ	BP86-DFT An^{III}: [6s5p4d2f1g] O, H: cc-pVQZ	MP2 An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ	HF An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ
		gas phase energies		
Ac	−640.5318416364	−640.7447152849	−638.8538056941	−636.8763040691
Th	−641.2900534608	−641.5028360377	−639.6146754683	−637.6352313844
Pa	−642.0169969475	−642.2296504725	−640.3412806360	−638.3618398330
U	−642.7180977463	−642.9306527415	−641.0410816119	−639.0633360106
Np	−643.3993798892	−643.6118549347	−641.7198473431	−639.7448974384
Pu	−644.0651270766	−644.2775262816	−642.3847910100	−640.4103473153
Am	−644.7176787735	−644.9300381834	−643.0416280967	−641.0628318830
Cm	−645.3590162485	−645.5713618476	−643.6895359356	−641.7049587475
Bk	−645.9942487313	−646.2065903861	−644.3280015626	−642.3384347520
Cf	−646.6205970502	−646.8329319776	−644.9584706921	−642.9656701248
Es	−647.2442728022	−647.4566083072	−645.5831500521	−643.5876559842
Fm	−647.8645379784	−648.0768727083	−646.2036506940	−644.2061721311
Md	−648.4801727148	−648.6924989069	−646.8221866893	−644.8228752787
No	−649.0964652245	−649.3087812760	−647.4392762576	−645.4384051042
Lr	−649.7125764645	−649.9248987872	−648.0565918683	−646.0542369946
		COSMO total energies		
Ac	−641.1245260726	−641.3365982213	−639.4483358660	−637.4691328524
Th	−641.8842958770	−642.0963021433	−640.2107074516	−638.2295868389
Pa	−642.6126183263	−642.8245162183	−640.9386374789	−638.9575342248
U	−643.3150964290	−643.5269157083	−641.6397653025	−639.6603709689
Np	−643.9989039080	−644.2106373433	−642.3211785939	−640.3446214862
Pu	−644.6658815527	−644.8775586542	−642.9872890576	−641.0112427829
Am	−645.3197597151	−645.5314114747	−643.6454028466	−641.6650134573
Cm	−645.9623771435	−646.1740316697	−644.2945298618	−642.3083627103
Bk	−646.5987392686	−646.8104009804	−644.9340826525	−642.9429246468
Cf	−647.2255964608	−647.4372817337	−645.5648936510	−643.5705029053
Es	−647.8504463680	−648.0621437684	−646.1907189187	−644.1936339886
Fm	−648.4717397203	−648.6834451239	−646.8122150791	−644.8131472489
Md	−649.0885487882	−649.3002567918	−647.4318931127	−645.4309940260
No	−649.7058742460	−649.9175804150	−648.0499840886	−646.0475261782
Lr	−650.3230364566	−650.5347596176	−648.6683228009	−646.6643803446

Table A.6.: $[An^{III}(OH_2)_8 \cdot OH_2]^{3+}$ total energies, in a.u.

	BP86-DFT An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ	BP86-DFT An^{III}: [6s5p4d2f1g] O, H: cc-pVQZ	MP2 An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ	HF An^{III}: [6s5p4d2f] O, H: aug-cc-pVDZ
			gas phase energies	
Ac	−717.0222580402	−717.2615562762	−715.1638328626	−712.9639491789
Th	−717.7806340041	−718.0198486773	−715.9244187222	−713.7227010150
Pa	−718.5075969712	−718.7466968030	−716.6510674110	−714.4486012382
U	−719.2086537297	−719.4476700541	−717.3507103259	−715.1494562396
Np	−719.8898381608	−720.1287783482	−718.0292605315	−715.8303400366
Pu	−720.5554376583	−720.7943101312	−718.6934140219	−716.4949006223
Am	−721.2078180479	−721.4466560430	−719.3490334675	−717.1465889716
Cm	−721.8489435877	−722.0877666494	−719.9958084726	−717.7877953121
Bk	−722.4839532339	−722.7227758885	−720.6333267779	−718.4202709690
Cf	−723.1100240112	−723.3488486064	−721.2627841061	−719.0463992549
Es	−723.7334340852	−723.9722653482	−721.8864982811	−719.6673135663
Fm	−724.3534093127	−724.5922499318	−722.5059938721	−720.2847514105
Md	−724.9686954208	−725.2075267068	−723.1232906360	−720.9002979420
No	−725.5846463757	−725.8234767883	−723.7393087841	−721.5145537289
Lr	−726.2003941658	−726.4392161004	−724.3554650706	−722.1290845129
			COSMO total energies	
Ac	−717.6000417410	−717.8423100791	−715.7447407068	−713.5394505783
Th	−718.3600644612	−718.6021390736	−716.5075082722	−714.3001100926
Pa	−719.0886996299	−719.3316833752	−717.2360394222	−715.0285243024
U	−719.7914638583	−720.0363450301	−717.9377836865	−715.7317571833
Np	−720.4742809767	−720.7176757746	−718.6183763481	−716.4148747809
Pu	−721.1413802542	−721.3849830935	−719.2847108360	−717.0817279151
Am	−721.7952366327	−722.0333179334	−719.9426883531	−717.7355434329
Cm	−722.4378067717	−722.6758831948	−720.5917390600	−718.3789031300
Bk	−723.0740893910	−723.3165512539	−721.2312560976	−719.0134340845
Cf	−723.7015261555	−723.9438183443	−721.8628778543	−719.6417174450
Es	−724.3259928028	−724.5682533351	−722.4883538824	−720.2645063979
Fm	−724.9470818074	−725.1893023495	−723.1096744257	−720.8837862750
Md	−725.5635924491	−725.8017131066	−723.7290908366	−721.5012870538
No	−726.1807028118	−726.4188290757	−724.3470203737	−722.1176073657
Lr	−726.7976537908	−727.0395920215	−724.9651808373	−722.7342215206

Table A.7.: $[An^{III}(OH_2)_7]^{3+}$ ZPEs in a.u., entropies S° in $kJ\,K^{-1}\,mol^{-1}$, mean An^{III}, O, H Mulliken net charges $q_{An^{III}}$, q_O, q_H, and An^{III} 5f Mulliken populations $n^{5f}_{An^{III}}$.

	ZPE	S°	$q_{An^{III}}$	$n^{5f}_{An^{III}}$	q_O	q_H
Ac	0.1705798	0.62	1.95	0.14	−0.31	0.23
Th	0.1707299	0.61	1.93	0.11	−0.31	0.23
Pa	0.1708613	0.61	1.92	0.09	−0.32	0.24
U	0.1710865	0.61	1.92	0.08	−0.32	0.24
Np	0.1712933	0.60	1.92	0.08	−0.32	0.24
Pu	0.1715774	0.60	1.92	0.07	−0.33	0.24
Am	0.1715976	0.59	1.94	0.07	−0.34	0.24
Cm	0.1717498	0.59	1.96	0.06	−0.34	0.25
Bk	0.1718795	0.59	1.98	0.06	−0.35	0.25
Cf	0.1719837	0.58	2.01	0.06	−0.36	0.25
Es	0.1721296	0.58	2.03	0.06	−0.36	0.25
Fm	0.1722105	0.58	2.05	0.05	−0.37	0.25
Md	0.1724054	0.58	2.09	0.05	−0.38	0.25
No	0.1725651	0.57	2.12	0.05	−0.39	0.26
Lr	0.1726641	0.57	2.16	0.05	−0.39	0.26

Table A.8.: $[An^{III}(OH_2)_8]^{3+}$ ZPEs in a.u., entropies S° in $kJ\,K^{-1}\,mol^{-1}$, mean An^{III}, O, H Mulliken net charges $q_{An^{III}}$, q_O, q_H, and An^{III} 5f Mulliken populations $n^{5f}_{An^{III}}$.

	ZPE	S°	$q_{An^{III}}$	$n^{5f}_{An^{III}}$	q_O	q_H
Ac	0.1950423	0.66	0.95	0.14	−0.34	0.23
Th	0.1954356	0.65	0.94	0.11	−0.35	0.24
Pa	0.1955726	0.65	0.93	0.10	−0.35	0.24
U	0.1957126	0.65	0.91	0.08	−0.35	0.24
Np	0.1961571	0.64	0.88	0.08	−0.36	0.24
Pu	0.1962281	0.63	0.86	0.07	−0.36	0.24
Am	0.1965359	0.63	0.82	0.07	−0.37	0.25
Cm	0.1966545	0.63	0.77	0.06	−0.37	0.25
Bk	0.1969403	0.62	0.71	0.06	−0.38	0.25
Cf	0.1968592	0.62	0.64	0.06	−0.39	0.25
Es	0.1971556	0.62	0.58	0.06	−0.40	0.25
Fm	0.1973633	0.61	0.51	0.05	−0.42	0.26
Md	0.1975285	0.61	0.42	0.05	−0.43	0.26
No	0.1975998	0.61	0.35	0.05	−0.44	0.26
Lr	0.1977131	0.61	0.27	0.05	−0.46	0.26

Table A.9.: $[An^{III}(OH_2)_9]^{3+}$ ZPEs in a.u., entropies S° in $kJ\,K^{-1}\,mol^{-1}$, mean An^{III}, O, H Mulliken net charges $q_{An^{III}}$, q_O, q_H, and An^{III} 5f Mulliken populations $n^{5f}_{An^{III}}$.

	ZPE	S°	$q_{An^{III}}$	$n^{5f}_{An^{III}}$	q_O	q_H
Ac	0.2195926	0.70	0.82	0.14	−0.39	0.24
Th	0.2197343	0.71	0.83	0.11	−0.39	0.24
Pa	0.2199736	0.69	0.84	0.09	−0.39	0.24
U	0.2199540	0.69	0.84	0.08	−0.40	0.25
Np	0.2200157	0.69	0.84	0.08	−0.40	0.25
Pu	0.2206238	0.68	0.85	0.07	−0.40	0.25
Am	0.2210587	0.67	0.83	0.07	−0.41	0.25
Cm	0.2210846	0.67	0.83	0.06	−0.42	0.25
Bk	0.2212451	0.67	0.81	0.06	−0.42	0.26
Cf	0.2214606	0.66	0.78	0.06	−0.43	0.26
Es	0.2213519	0.66	0.75	0.06	−0.43	0.26
Fm	0.2216677	0.66	0.70	0.05	−0.44	0.26
Md	0.2220093	0.65	0.64	0.05	−0.45	0.26
No	0.2221944	0.64	0.56	0.05	−0.47	0.27
Lr	0.2221178	0.65	0.46	0.05	−0.49	0.27

Table A.10.: $[An^{III}(OH_2)_7 \cdot OH_2]^{3+}$ ZPEs in a.u., entropies S° in $kJ\,K^{-1}\,mol^{-1}$, mean An^{III}, O, H Mulliken net charges $q_{An^{III}}$, q_O, q_H, and An^{III} 5f Mulliken populations $n^{5f}_{An^{III}}$. O and H Mulliken net charges given refer to the 1^{st} hydration sphere OH_2 ligands that coordinate the single 2^{nd} hydration sphere OH_2 ligand.

	ZPE	S°	$q_{An^{III}}$	$n^{5f}_{An^{III}}$	q_O	q_H
Ac	0.1961511	0.66	1.97	0.14	−0.33	0.25
Th	0.1962431	0.65	1.96	0.11	−0.34	0.26
Pa	0.1964863	0.65	1.95	0.10	−0.34	0.26
U	0.1965906	0.64	1.94	0.08	−0.35	0.26
Np	0.1967915	0.64	1.95	0.08	−0.35	0.26
Pu	0.1968881	0.64	1.95	0.08	−0.35	0.27
Am	0.1971415	0.63	2.00	0.07	−0.37	0.27
Cm	0.1972491	0.63	2.00	0.06	−0.38	0.28
Bk	0.1973940	0.62	2.02	0.06	−0.39	0.28
Cf	0.1976388	0.62	2.04	0.06	−0.40	0.28
Es	0.1977064	0.62	2.06	0.06	−0.40	0.28
Fm	0.1978401	0.62	2.09	0.05	−0.41	0.29
Md	0.1979855	0.61	2.12	0.05	−0.42	0.29
No	0.1980935	0.61	2.16	0.05	−0.43	0.29
Lr	0.1982053	0.61	2.19	0.05	−0.43	0.30

Table A.11.: $[An^{III}(OH_2)_8 \cdot OH_2]^{3+}$ ZPEs in a.u., entropies S° in $kJ\,K^{-1}\,mol^{-1}$, mean An^{III}, O, H Mulliken net charges $q_{An^{III}}$, q_O, q_H, and An^{III} 5f Mulliken populations $n^{5f}_{An^{III}}$. O and H Mulliken net charges given refer to the 1^{st} hydration sphere OH_2 ligands that coordinate the single 2^{nd} hydration sphere OH_2 ligand.

	ZPE	S°	$q_{An^{III}}$	$n^{5f}_{An^{III}}$	q_O	q_H
Ac	0.2206256	0.72	2.06	0.14	−0.37	0.26
Th	0.2209147	0.72	2.05	0.11	−0.37	0.27
Pa	0.2210555	0.71	2.03	0.10	−0.38	0.27
U	0.2212424	0.71	2.03	0.09	−0.38	0.27
Np	0.2214455	0.70	2.03	0.08	−0.39	0.28
Pu	0.2217663	0.68	2.04	0.07	−0.40	0.28
Am	0.2220872	0.67	2.05	0.07	−0.41	0.28
Cm	0.2219589	0.69	2.07	0.06	−0.41	0.28
Bk	0.2220506	0.69	2.10	0.06	−0.42	0.29
Cf	0.2222724	0.69	2.14	0.06	−0.42	0.29
Es	0.2223784	0.68	2.17	0.06	−0.43	0.29
Fm	0.2225145	0.68	2.22	0.05	−0.43	0.29
Md	0.2230010	0.66	2.29	0.05	−0.45	0.30
No	0.2231152	0.65	2.35	0.05	−0.45	0.30
Lr	0.2228761	0.67	2.43	0.05	−0.46	0.30