Electronic Supplementary information for:

Computational Study of the Interactions of Tetravalent Actinides (An = Th – Pu) with the α-Fe_{13} Keggin Cluster

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Supplementary Figures

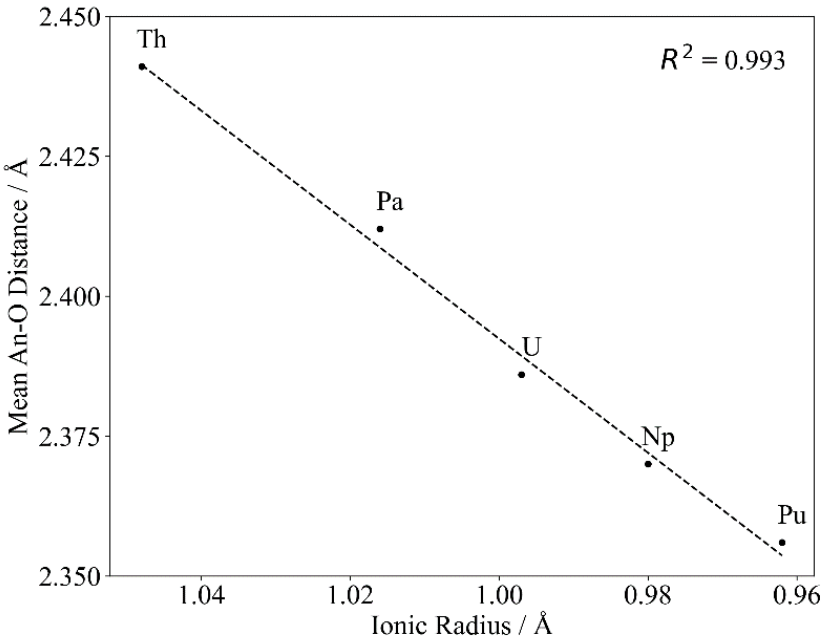

Figure S1: Mean An-O for the optimized 9 coordinate aquo complexes $[An(H_2O)_9]^{4+}$ calculated at PBE0/def2-SVP/ECP level, vs. ionic radius.[1]

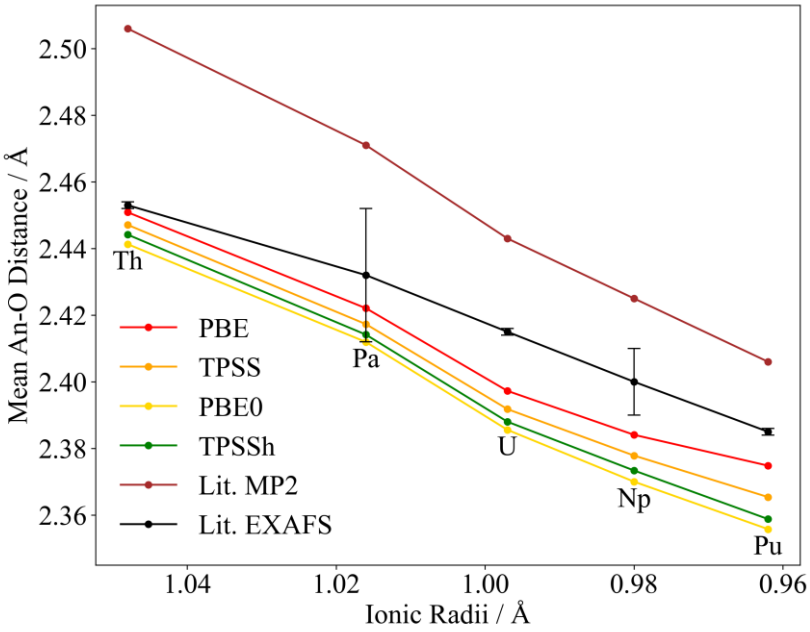

Figure S2: Mean An-O for the optimized 9 coordinate aquo complexes $[An(H_2O)_9]^{4+}$ compared to literature EXAFS and MP2 calculated distances for $[An(H_2O)_9]^{4+}\bullet H_2O$.[1,2]

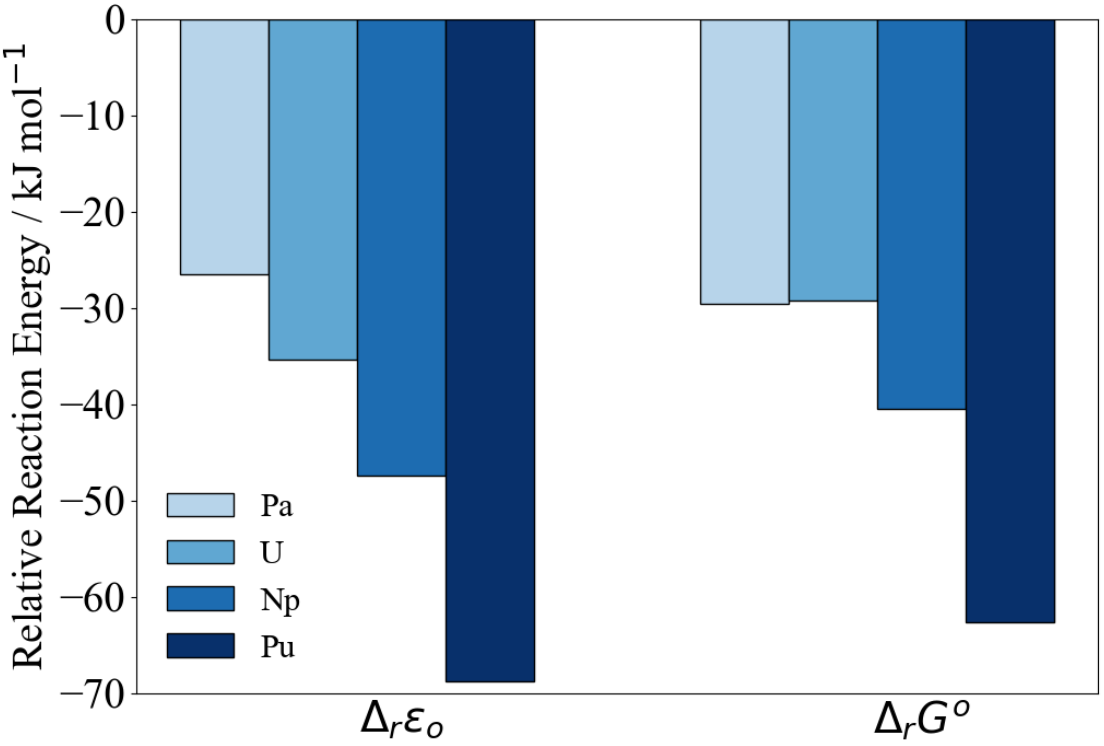

Figure S3: Reaction energy Δ_r according to Eqn (2) relative to Th (PBE/def2-TZVP/ECP).

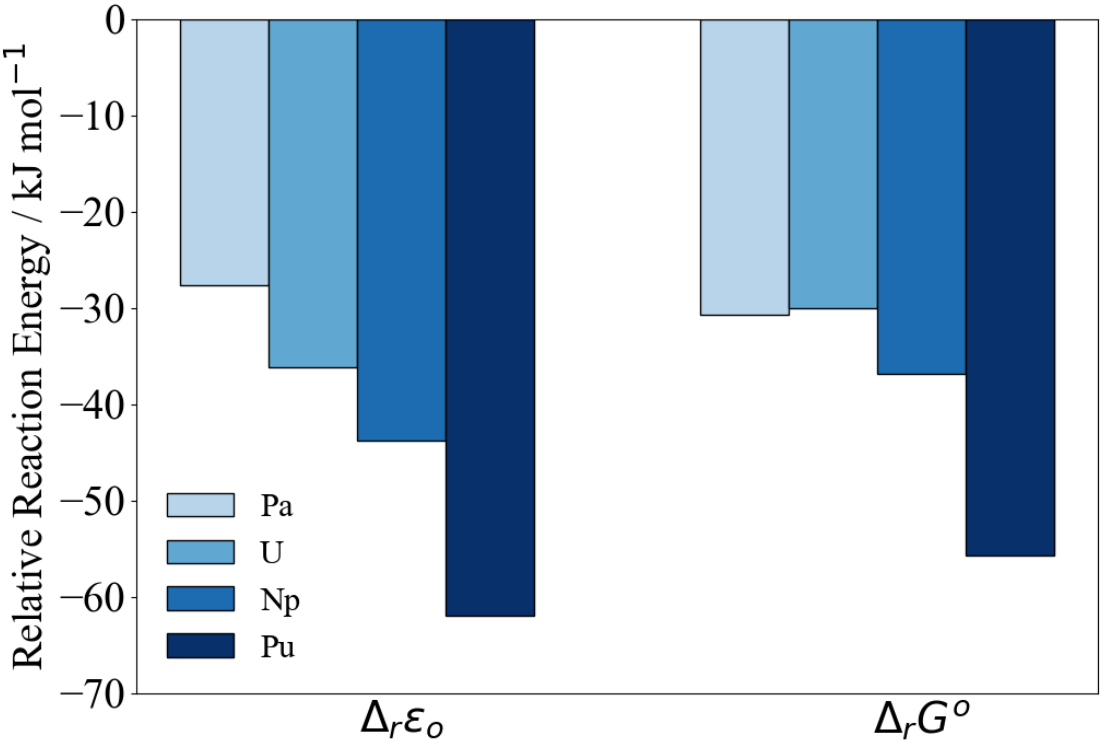

Figure S4: Reaction energy Δ_r according to Eqn (2) relative to Th (TPSS/def2-TZVP/ECP).

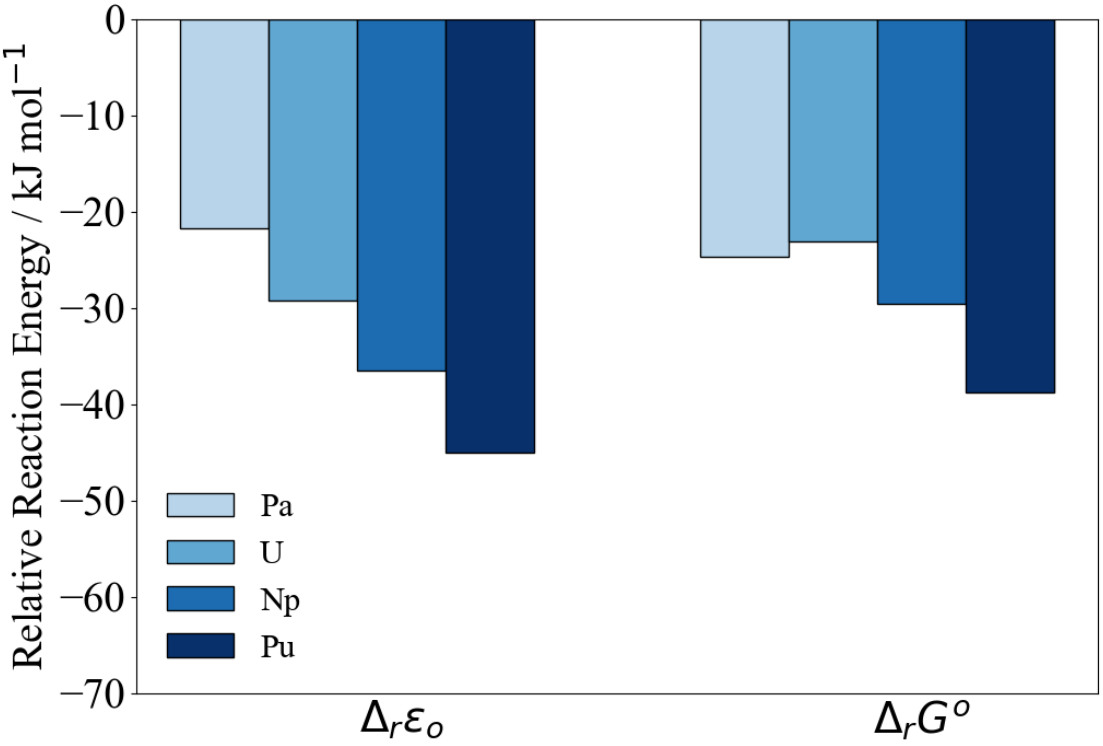

Figure S5: Reaction energy Δ_r according to Eqn (2) relative to Th (PBE0/def2-TZVP/ECP).

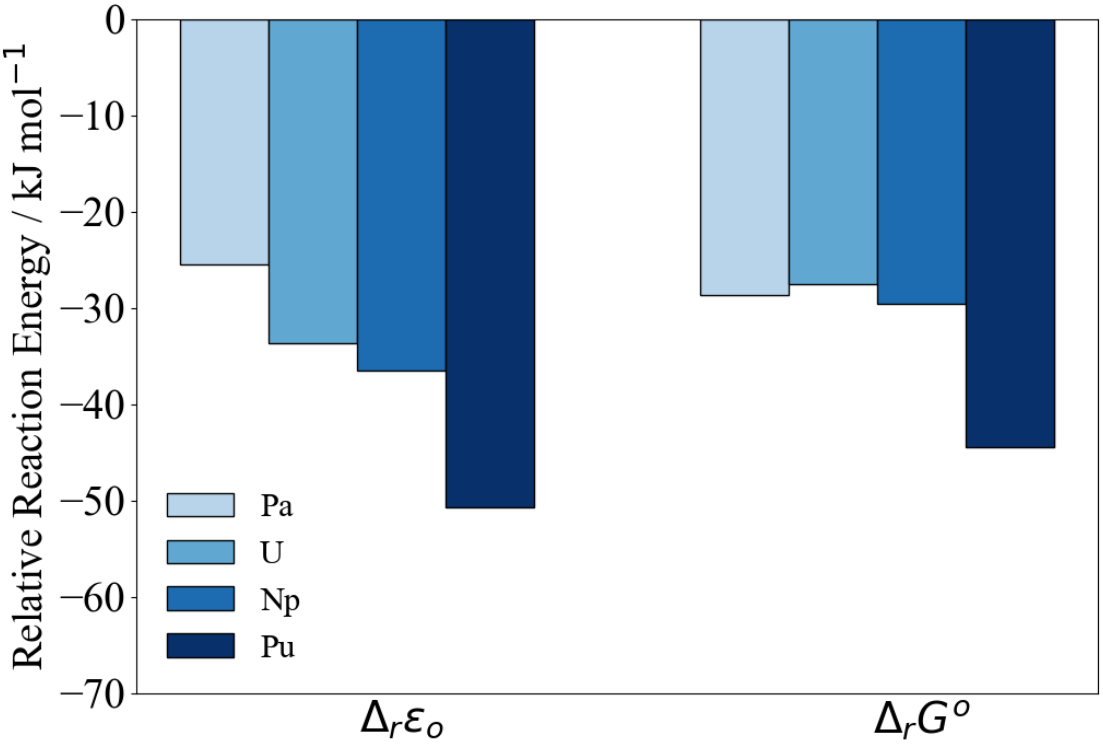

Figure S6: Reaction energy Δ_r according to Eqn (2) relative to Th (TPSSh/def2-TZVP/ECP).

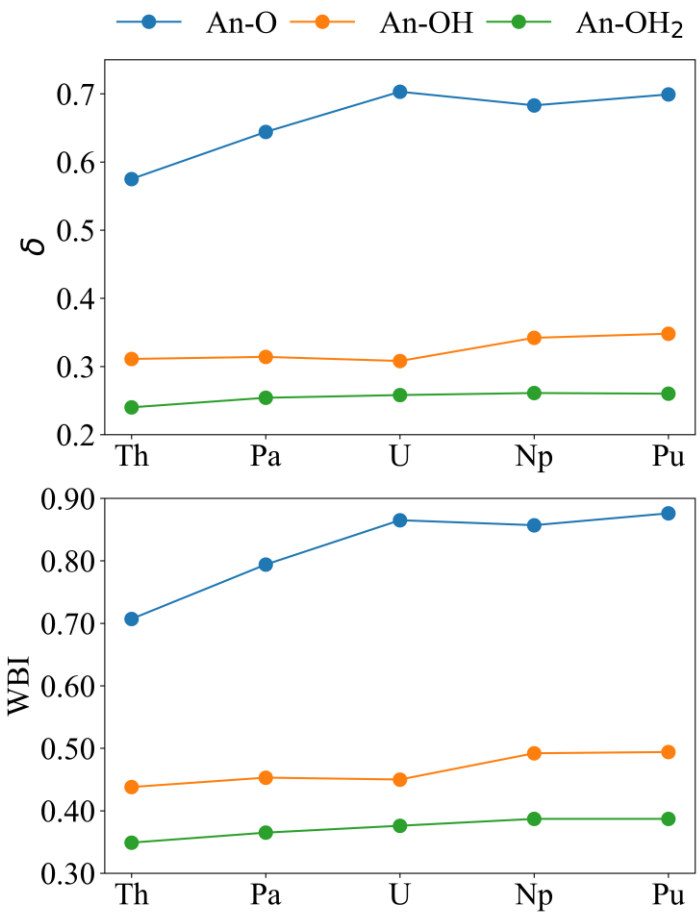

Figure S7: PBE0/Def2TZVP/SARC-DKH2+SO delocalisation indices and Wiberg bond indices (a.u.) for the An-O, An-OH, and An-OH_2 bonds in $[\{An(H_2O)_5\}\{Na(H_2O)_2\}_5Fe_{13}O_{28}H_{12}]^{4+}$.

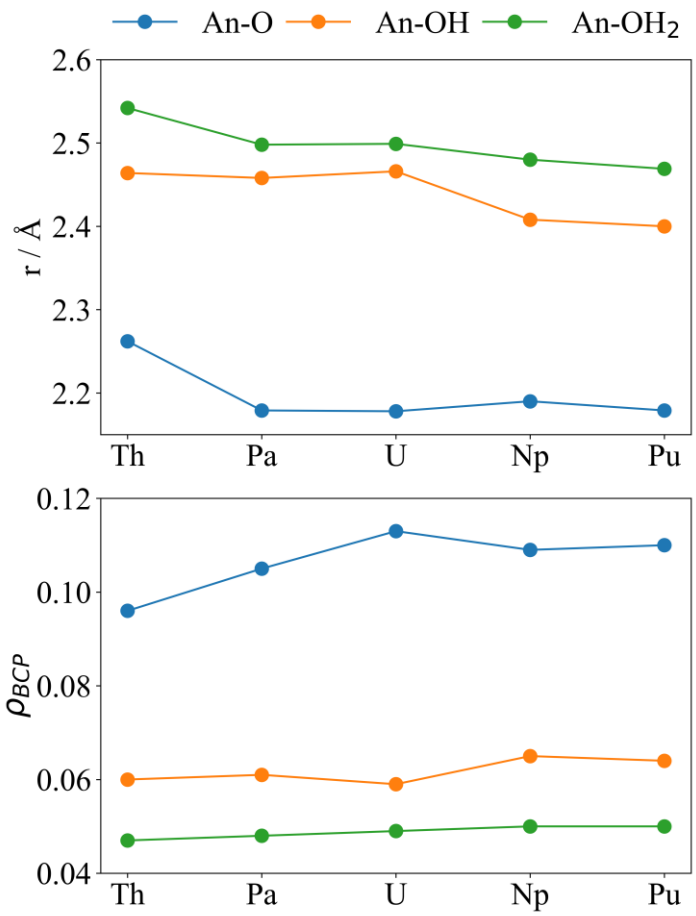

Figure S8: PBE0/def2-TZVP/ECP optimised distances *r* (Å, upper) and PBE0/Def2TZVP/SARC-DKH2+SO ρ_{BCP} (a.u., lower) for $[\{An(H_2O)_5\}\{Na(H_2O)_2\}_5Fe_{13}O_{28}H_{12}]^{4+}$.

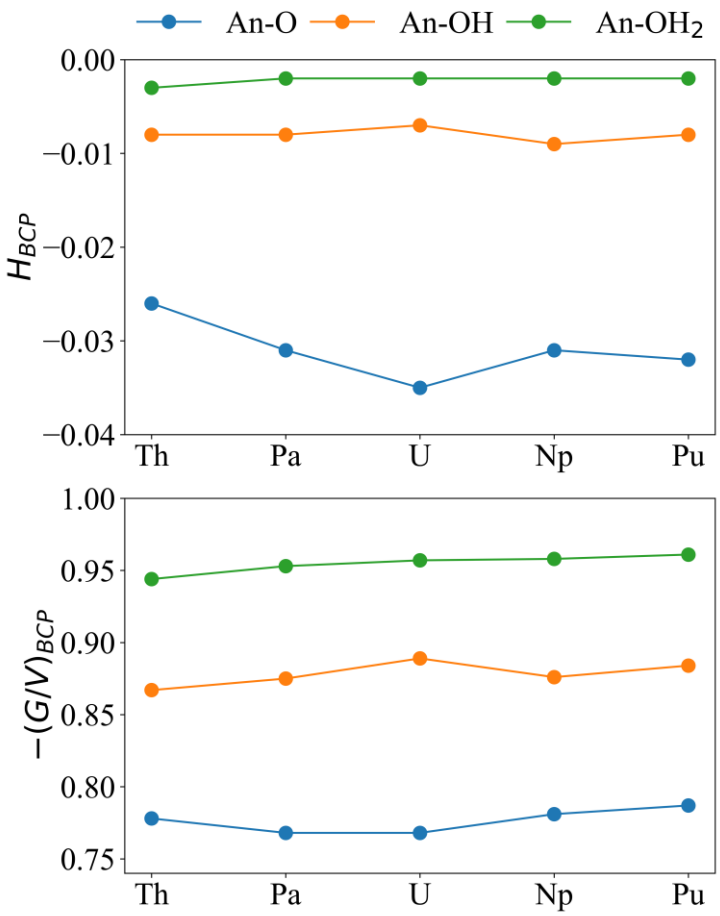

Figure S9: PBE0/Def2TZVP/SARC-DKH2+SO H_{BCP} and $-(G/V)_{BCP}$ (a.u.) for the An-O, An-OH, and An-OH_2 bonds in $[\{An(H_2O)_5\}\{Na(H_2O)_2\}_5Fe_{13}O_{28}H_{12}]^{4+}$.

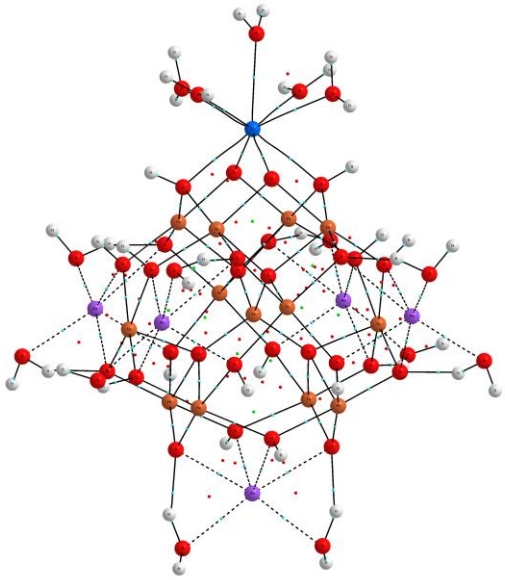

Figure S10: Left, full molecular graph of the Pu substituted Fe_{13} Keggin cluster and right, close up of the "square-window" binding. BCPs in light blue, Ring Critical Points (RCP) in red, Cage Critical Points (CCP) in green. Weak critical points are connected to nuclei with dashed paths. Default cutoff values were used within AIMStudio.

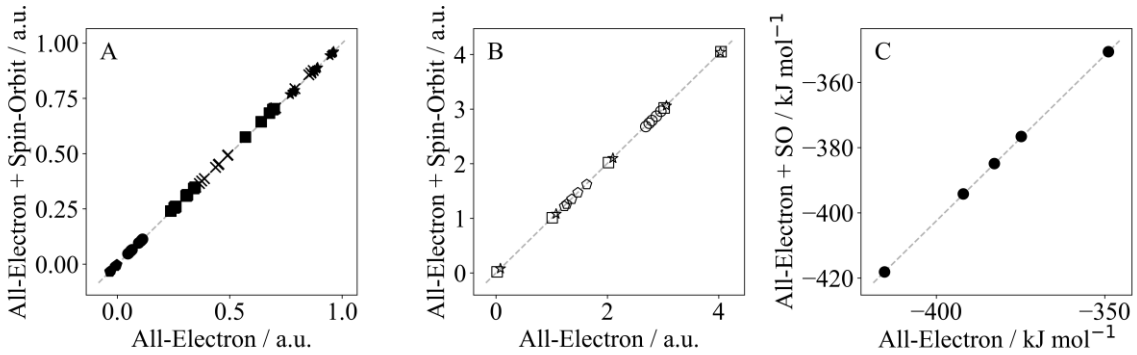

Figure S11: Comparison of various quantities derived from all-electron calculations including and neglecting spin-orbit coupling. A; QTAIM critical point properties and Bonding Indices: ρ_{BCP} = circles, $-(G/V)_{BCP}$ = stars, H_{BCP} = pentagons, δ = squares, and WBI = crosses. B; Charges and Spin: QTAIM charge = circles, QTAIM spin density = squares, NPA charge = pentagons, NPA spin density = stars. C; Electronic reaction energy. The identity line y = x is marked with a dashed grey line.

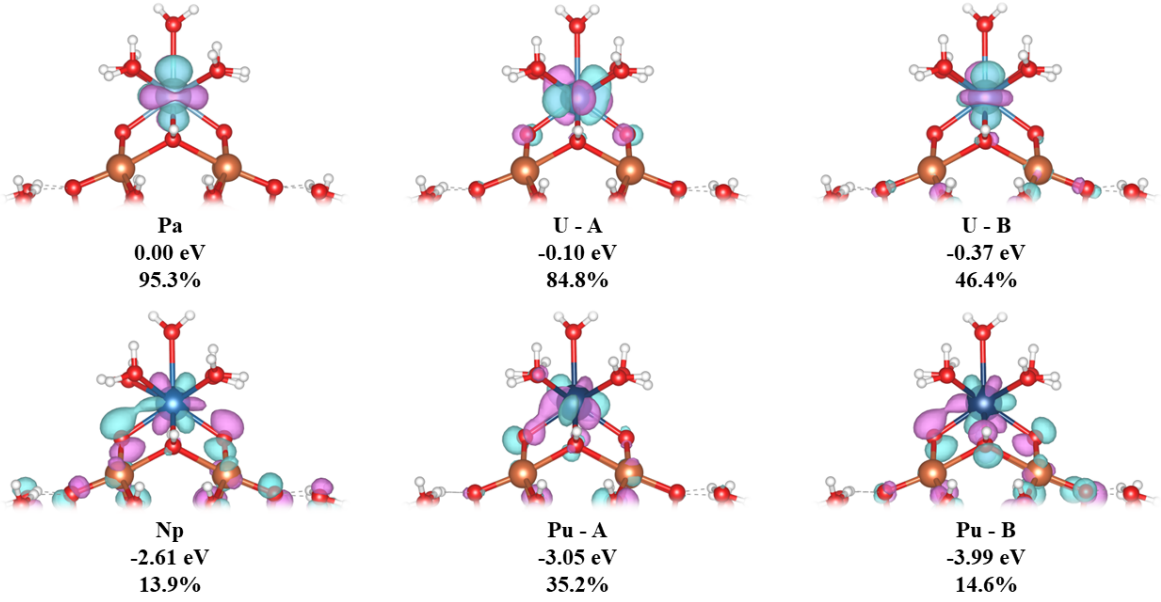

Figure S12: Three-dimensional representations, *ΔE vs* the HOMO, and %An(f) of selected α spin MOs, showing An(5f)-O(2p) overlap in Np and Pu (isovalue = 0.05). These MOs were obtained including SO.

Supplementary Tables

Table S1: Mean An-O distances (Å) for the optimized 9 coordinate aquo complexes $[An(H_2O)_9]^{4+}$. Ionic radii for tetravalent actinides with 8-fold coordination. Correlation of An-O distance with ionic radii given in parentheses (R^2).

An	Ionic Radii[1]	Lit. EXAFS[2] (*0.995*)	Lit. MP2[2] (*0.997*)	PBE (*0.982*)	TPSS (*0.992*)	PBE0 (*0.993*)	TPSSh (*0.993*)
Th	1.048	2.453	2.506	2.451	2.447	2.441	2.444
Pa	1.016	2.432	2.471	2.422	2.417	2.412	2.414
U	0.997	2.415	2.443	2.397	2.392	2.386	2.388
Np	0.980	2.40	2.425	2.384	2.378	2.370	2.373
Pu	0.962	2.385	2.406	2.375	2.365	2.356	2.359

Table S2: Energy components used to calculate Th substitution energy. Electronic energy calculated at the PBE0/def2-TZVP/ECP, PBE0/Def2TZVP/SARC-DKH2, and PBE0/Def2TZVP/SARC-DKH2+SO levels and G_{corr} calculated using the RRHO model with frequencies at the PBE0/def2-SVP/ECP level. Energy units given in Hartree.

	Fe_{13}	$[Th(H_2O)_9]^{4+}$	Th-Fe_{13}	$[Na(H_2O)_6]^{+}$
$\mathcal{E}_0$ ECP	-20429.461081	-1094.844441	-20903.866774	-620.520604
$\mathcal{E}_0$ SARC-DKH2	-20523.919177	-27103.083755	-47006.151069	-620. 984741
$\mathcal{E}_0$ SARC-DKH2+SO	-20524.157791	-27110.131633	-47013.438006	-620.984943
G_{corr}	0.413785	0.174763	0.483264	0.110297

Table S3: Energy components used to calculate Pa substitution energy. Electronic energy calculated at the PBE0/def2-TZVP/ECP, PBE0/Def2TZVP/SARC-DKH2, and PBE0/Def2TZVP/SARC-DKH2+SO levels and G_{corr} calculated using the RRHO model with frequencies at the PBE0/def2-SVP/ECP level. Energy units given in Hartree.

	Fe_{13}	$[Pa(H_2O)_9]^{4+}$	Pa-Fe_{13}	$[Na(H_2O)_6]^{+}$
$\mathcal{E}_0$ ECP	-20429.461081	-1128.513287	-20937.543881	-620.520604
$\mathcal{E}_0$ SARC-DKH2	-20523.919177	-27859.183331	-47762.260532	-620. 984741
$\mathcal{E}_0$ SARC-DKH2+SO	-20524.157791	-27866.446800	-47769.763075	-620.984943
G_{corr}	0.413785	0.175543	0.482885	0.110297

Table S4: Energy components used to calculate U substitution energy. Electronic energy calculated at the PBE0/def2-TZVP/ECP, PBE0/Def2TZVP/SARC-DKH2, and PBE0/Def2TZVP/SARC-DKH2+SO levels and G_{corr} calculated using the RRHO model with frequencies at the PBE0/def2-SVP/ECP level. Energy units given in Hartree.

	Fe_{13}	$[U(H_2O)_9]^{4+}$	U-Fe_{13}	$[Na(H_2O)_6]^{+}$
$\mathcal{E}_0$ ECP	-20429.461081	-1164.034512	-20973.067953	-620.520604
$\mathcal{E}_0$ SARC-DKH2	-20523.919177	-28629.674785	-48532.755009	-620. 984741
$\mathcal{E}_0$ SARC-DKH2+SO	-20524.157791	-28637.104434	-48540.423883	-620.984943
G_{corr}	0.413785	0.175884	0.486704	0.110297

Table S5: Energy components used to calculate Np substitution energy. Electronic energy calculated at the PBE0/def2-TZVP/ECP, PBE0/Def2TZVP/SARC-DKH2, and PBE0/Def2TZVP/SARC-DKH2+SO levels and G_{corr} calculated using the RRHO model with frequencies at the PBE0/def2-SVP/ECP level. Energy units given in Hartree.

	Fe_{13}	$[Np(H_2O)_9]^{4+}$	Np-Fe_{13}	$[Na(H_2O)_6]^{+}$
$\mathcal{E}_0$ ECP	-20429.461081	-1201.515171	-21010.551418	-620.520604
$\mathcal{E}_0$ SARC-DKH2	-20523.919177	-29415.337443	-49318.421189	-620.984741
$\mathcal{E}_0$ SARC-DKH2+SO	-20524.157791	-29422.968708	-49326.291708	-620.984943
G_{corr}	0.413785	0.176303	0.487478	0.110297

Table S6: Energy components used to calculate Pu substitution energy. Electronic energy calculated at the PBE0/def2-TZVP/ECP, PBE0/Def2TZVP/SARC-DKH2, and PBE0/Def2TZVP/SARC-DKH2+SO levels and G_{corr} calculated using the RRHO model with frequencies at the PBE0/def2-SVP/ECP level. Energy units given in Hartree.

	Fe_{13}	$[Pu(H_2O)_9]^{4+}$	Pu-Fe_{13}	$[Na(H_2O)_6]^{+}$
$\mathcal{E}_0$ ECP	-20429.461081	-1240.964737	-21050.00420	-620.520604
$\mathcal{E}_0$ SARC-DKH2	-20523.919177	-30215.893594	-50118.986237	-620. 984741
$\mathcal{E}_0$ SARC-DKH2+SO	-20524.157791	-30223.679217	-50127.011318	-620.984943
G_{corr}	0.413785	0.176750	0.487620	0.110297

Table S7: Electronic energy calculated at the PBE/def2-TZVP/ECP, TPSS/def2-TZVP/ECP, and TPSSh/def2-TZVP/ECP level using the PBE0/def2-SVP/ECP optimised geometries. Energy units given in Hartree.

An	PBE		TPSS		TPSSh	
	$[An(H_2O)_9]^{4+}$	An-Fe_{13}	$[An(H_2O)_9]^{4+}$	An-Fe_{13}	$[An(H_2O)_9]^{4+}$	An-Fe_{13}
Th	-1094.857044	-20903.779094	-1095.499856	-20911.618467	-1095.431278	-20910.862162
Pa	-1128.544945	-20937.477078	-1129.174569	-20945.303698	-1129.099416	-20944.540019
U	-1164.065067	-20973.0005763	-1164.676237	-20980.808597	-1164.603269	-20980.046965
Np	-1201.561232	-21010.501341	-1202.163032	-21018.2983422	-1202.084929	-21017.529717
Pu	-1241.017286	-21049.965540	-1241.600100	-21057.742282	-1241.520698	-21056.970879
	$[Na(H_2O)_6]^{+}$	Fe_{13}	$[Na(H_2O)_6]^{+}$	Fe_{13}	$[Na(H_2O)_6]^{+}$	Fe_{13}
	-620.502489	-20429.344750	-621.152018	-20437.186202	-621.095804	-20436.441209

Table S8: PBE0/Def2TZVP/SARC-DKH+SO NPA and Mulliken populations for the 6d- and 5f-orbitals. n_{excess}(f) is the difference between the NPA calculated n(f) and the formal value An(IV) (*e.g.* 0 for Th(IV) to 4 for Pu(IV)). These data are analogous to the all-electron equivalents in table 3 of the main text.

An	NPA			Mulliken		
	n(d)	n(f)	n_{excess}(f)	n(d)	n(f)	n_{excess}(f)
Th	1.08	0.09	0.09	0.94	0.41	0.41
Pa	1.14	1.62	0.62	0.97	1.47	0.47
U	1.16	2.62	0.62	0.98	2.54	0.54
Np	1.19	3.64	0.64	1.01	3.57	0.57
Pu	1.19	4.72	0.72	1.03	4.62	0.62

Table S9: PBE0/Def2TZVP/SARC-DKH+SO NPA, Mulliken, and QTAIM charges and spin-density on the actinide centre.

An	NPA		Mulliken		QTAIM		
	q(M)	ρ_s(M)	q(M)	ρ_s(M)	q(M)	ρ_s(M)	$Z - \lambda$
Th	1.62	0.08	2.94	0.02	2.96	0.02	4.52
Pa	1.47	1.08	2.87	1.09	2.87	1.01	4.56
U	1.35	2.10	2.78	2.13	2.79	2.02	4.54
Np	1.26	3.06	2.69	3.12	2.74	3.02	4.51
Pu	1.22	4.04	2.62	3.12	2.68	4.05	4.48

Table S10: PBE0/Def2TZVP/SARC-DKH2 An-O dataset including QTAIM bond order metrics δ and WBI. Distances in Å, QTAIM data in a.u.

An	r	ρ	$\nabla^2\rho$	H	-G/V	δ	WBI
Th	2.262	0.095	0.262	0.780	-0.026	0.570	0.704
Pa	2.179	0.105	0.288	0.770	-0.031	0.640	0.790
U	2.178	0.112	0.321	0.769	-0.034	0.698	0.861
Np	2.190	0.108	0.325	0.783	-0.031	0.677	0.852
Pu	2.179	0.110	0.342	0.789	-0.032	0.692	0.871

Table S11: PBE0/Def2TZVP/SARC-DKH2 An-OH dataset including QTAIM bond order metrics δ and WBI. Distances in Å, QTAIM data in a.u.

An	r	ρ	$\nabla^2\rho$	H	-G/V	δ	WBI
Th	2.464	0.060	0.181	0.869	-0.008	0.309	0.437
Pa	2.458	0.060	0.194	0.877	-0.008	0.312	0.452
U	2.466	0.059	0.196	0.891	-0.007	0.306	0.449
Np	2.408	0.065	0.215	0.878	-0.009	0.340	0.490
Pu	2.400	0.064	0.221	0.886	-0.008	0.346	0.492

Table S12: PBE0/Def2TZVP/SARC-DKH2 An-OH_2 dataset including QTAIM bond order metrics δ and WBI. Distances in Å, QTAIM data in a.u.

An	r	ρ	$\nabla^2\rho$	H	-G/V	δ	WBI
Th	2.542	0.047	0.165	0.946	-0.003	0.238	0.349
Pa	2.498	0.048	0.183	0.955	-0.002	0.253	0.365
U	2.499	0.049	0.192	0.959	-0.002	0.257	0.375
Np	2.480	0.050	0.199	0.960	-0.002	0.260	0.387
Pu	2.469	0.050	0.203	0.962	-0.002	0.259	0.387

Table S13: PBE0/Def2TZVP/SARC-DKH2+SO An-O dataset including QTAIM bond order metrics δ and WBI. Distances in Å, QTAIM data in a.u.

An	r	ρ	$\nabla^2\rho$	H	-G/V	δ	WBI
Th	2.262	0.096	0.260	0.778	-0.026	0.575	0.707
Pa	2.179	0.105	0.286	0.768	-0.031	0.644	0.794
U	2.178	0.113	0.319	0.768	-0.035	0.703	0.865
Np	2.190	0.109	0.323	0.781	-0.031	0.683	0.857
Pu	2.179	0.110	0.339	0.787	-0.032	0.699	0.876

Table S14: PBE0/Def2TZVP/SARC-DKH2+SO An-OH dataset including QTAIM bond order metrics δ and WBI. Distances in Å, QTAIM data in a.u.

An	r	ρ	$\nabla^2\rho$	H	-G/V	δ	WBI
Th	2.464	0.060	0.180	0.867	-0.008	0.311	0.438
Pa	2.458	0.061	0.193	0.875	-0.008	0.314	0.453
U	2.466	0.059	0.195	0.889	-0.007	0.308	0.450
Np	2.408	0.065	0.214	0.876	-0.009	0.342	0.492
Pu	2.400	0.064	0.220	0.884	-0.008	0.348	0.494

Table S15: PBE0/Def2TZVP/SARC-DKH2+SO An-OH_2 dataset including QTAIM bond order metrics δ and WBI. Distances in Å, QTAIM data in a.u.

An	r	ρ	$\nabla^2\rho$	H	-G/V	δ	WBI
Th	2.542	0.047	0.164	0.944	-0.003	0.240	0.349
Pa	2.498	0.048	0.182	0.953	-0.002	0.254	0.365
U	2.499	0.049	0.192	0.957	-0.002	0.258	0.376
Np	2.480	0.050	0.199	0.958	-0.002	0.261	0.387
Pu	2.469	0.050	0.202	0.961	-0.002	0.260	0.387

Thermodynamic Corrections and Reaction Energies

There is good correlation between the reaction energy and ionic radius of the tetravalent actinide (figure 4), although that at the $\Delta_r G^o$ level is noticeably lower with $R^2 = 0.85$ compared to $\Delta_r H^o$ where $R^2 = 0.96$. We suspected that the disagreement comes from the entropic corrections, specifically the vibrational entropy. To investigate further, an in-house version of Turbomole's freeH script was written allowing a deeper look at how these thermodynamic corrections are calculated and potential sources of error. The thermodynamic corrections were calculated according to the standard rigid-rotor harmonic approximation (RRHO) equations which can be found in standard textbooks[3-5] and are nicely summarised by Ochterski.[6] Gibbs free energy corrections G_{corr} can be calculated *via* equation 1 found in the Turbomole freeH output file:

$$G_{corr} = \varepsilon_{ZPE} - RT \ln q_{trans} q_{rot} q_{vib} \quad (1)$$

where q_{vib} is calculated according to

$$q_{vib} = \prod_i \frac{1}{1 - e^{\Theta_i / T}} \quad (2)$$

where Θ_i are the rotational temperatures of vibrational modes i calculated according to

$$\Theta_i = \frac{hcv_i}{k_B} \quad (3)$$

where h, c, and k_B are Planck's constant, the speed of light, and the Boltzmann constant respectively. The dependence of q_{vib} on the vibrational frequency v_i is worth noting as it is important to the discussion below. To verify our implementation of the thermodynamics, we compare the energy corrections calculated using freeH and our script in tables S16 for the non-aquo actinide complexes. The agreement is very good, all energies are within 0.1 kJ mol^{-1} and the entropy term is within 0.001 kJ mol^{-1} K^{-1}.

Table S16: Thermodynamic corrections to the electronic energy for $[An(H_2O)_9]^{4+}$ calculated using our in-house script and Turbomole's freeH given in parentheses. Values are given to the same decimal place as freeH with ε_{ZPE}, U_{corr}, H_{corr}, and G_{corr} in kJ mol^{-1} and S_{corr} is in kJ mol^{-1} K^{-1}.

An	ε_{ZPE}	U_{corr}	H_{corr}	G_{corr}	S_{corr}
Th	589.8 (589.8)	654.92 (654.91)	657.40 (657.39)	458.84 (458.78)	0.66596 (0.66613)
Pa	591.2 (591.2)	655.58 (655.56)	658.05 (658.04)	462.61 (462.54)	0.65554 (0.65570)
U	591.5 (591.5)	655.49 (655.47)	657.96 (657.95)	464.51 (464.45)	0.64886 (0.64902)
Np	592.7 (592.7)	656.00 (655.99)	658.48 (658.47)	466.32 (466.26)	0.64451 (0.64468)
Pu	593.4 (593.4)	656.18 (656.17)	658.66 (658.65)	468.05 (467.99)	0.63931 (0.63948)

The second method of calculating G_{corr} follows reference 6, and most standard texts:

$$G_{corr} = (U_{corr} + RT) - TS_{tot} \quad (4)$$

where R is the molar gas constant, U_{corr} is the sum of translational, rotational, and vibrational contributions to the internal thermal energy,

$$U_{corr} = U_{trans} + U_{rot} + R\sum_i \Theta_i \left(\frac{1}{2} + \frac{1}{e^{\Theta_i/T} - 1}\right) \quad (5)$$

and S_{tot} is the total entropy of the system defined as the sum of electronic, translational, rotational, and vibrational contributions.

$$S_{tot} = S_{elec} + S_{trans} + S_{rot} + R\sum_i \left(\frac{\Theta_i/T}{e^{\Theta_i/T} - 1} - \ln\left(1 - e^{-\Theta_i/T}\right)\right) \quad (6)$$

Here we have explicitly written out the form of the vibrational contributions for discussion below. In this formulation the total entropy includes electronic contributions which are not included in freeH as that method does not include the electronic partition function q_{elec}. Assuming that the ground electronic state is well separated from the first excited state such that the separation is much larger than k_BT, q_{elec} is simply the ground state spin multiplicity of the system (ω_0).[4-6] The electronic contributions are important for open-shell systems, especially those described here with a maximum multiplicity of 70 in $[\{Pu(H_2O)_5\}\{Na(H_2O)_2\}_5Fe_{13}O_{28}H_{12}]^{4+}$. In this case, at 298.15 K, TS_{elec} is on the order of ~10 kJ mol^{-1} (table S17) and should not be ignored in the calculation of G_{corr}. The thermodynamic corrections in

tables S2 – S6 include electronic contributions as do all reaction energies throughout this work. It should be noted that equations 1 and 4 are equivalent if q_{elec} is included in equation 1, so Turbomole's implementation is correct for closed-shell systems.

Table S17: Breakdown showing the ground state electronic multiplicity, electronic entropy S_{elec}, and $-TS_{elec}$ at 298.15 K for structures in this work.

Structure	Ground State Multiplicity ($q_{elec} = \omega_0$)	S_{elec} / kJ mol^{-1} K^{-1}	$-TS_{elec}$ / kJ mol^{-1}
$[Na(H_2O)_6]^+$	1	0.0000	-0.00
$[Th(H_2O)_9]^{4+}$	1	0.0000	-0.00
$[Pa(H_2O)_9]^{4+}$	2	0.0058	-1.72
$[U(H_2O)_9]^{4+}$	3	0.0091	-2.72
$[Np(H_2O)_9]^{4+}$	4	0.0115	-3.44
$[Pu(H_2O)_9]^{4+}$	5	0.0134	-3.99
Fe_{13}	66	0.0348	-10.39
Th-Fe_{13}	66	0.0348	-10.39
Pa-Fe_{13}	67	0.0350	-10.42
U-Fe_{13}	68	0.0351	-10.46
Np-Fe_{13}	69	0.0352	-10.50
Pu-Fe_{13}	70	0.0353	-10.53

Whichever set of equations are used, we can see that low frequency modes contribute significantly to G_{corr}. In equations 2, 5, and 6, as the vibrational frequency tends to zero the denominator of the vibrational terms vanish and the quantities diverge leading to increasingly large corrections. It is well known that low frequencies suffer more from anharmonic effects and are therefore not well described by the harmonic oscillator model. It follows that errors in these frequencies contribute significantly to errors in the free energy calculation.[7]

Truhlar and co-workers devised a quasi-RRHO (QRRHO) approach whereby modes below a threshold frequency are raised to that threshold.[8] This helps to reduce the impact of error in low frequency modes in the free energy calculation. We implemented this QRRHO approach and recalculated the thermodynamic corrections and reaction energies. Setting the threshold frequency to 100cm^{-1} (common in the literature) improves R^2 at the $\Delta_r G^o$ level from $R^2 = 0.91$ (harmonic approximation) to 0.93, though we note this is still lower than 0.97 achieved at the electronic level.

Table S18: Reaction energy without thermodynamic corrections and with various methods of calculating the Gibbs energy correction. Correlation with actinide 8-coordinate ionic radii given in italic. Energies given in kJ mol^{-1}.

An	$\Delta_r\varepsilon_0$ *(0.97)*	$\Delta_r G^o$ (RRHO) *(0.91)*	$\Delta_r G^o$ (QRRHO 60cm^{-1}) *(0.91)*	$\Delta_r G^o$ (QRRHO 100cm^{-1}) *(0.93)*
Th	-348.9	-335.7	-335.0	-331.1
Pa	-374.8	-364.7	-362.3	-358.1
U	-382.8	-363.5	-361.8	-359.1
Np	-392.0	-371.8	-370.7	-368.5
Pu	-415.4	-396.0	-394.6	-393.1

Since the species in this work were optimised without symmetry constraints it is difficult to directly compare individual vibrational modes. However, if we consider groups of modes of similar frequency we can make general statements about the ordering of the frequencies from Th – Pu. Figure S12 and S13 show the calculated vibrational frequencies of the $[An(H_2O)_9]^{4+}$ and $[\{An(H_2O)_5\}Na_5Fe_{13}O_{28}H_{12}]^{4+}$ species. The $[An(H_2O)_9]^{4+}$ vibrational modes are well behaved in the sense that, for a given frequency range, there is consistency in the ordering of the vibrational frequency from Th – Pu. Figure S12 A and B correspond with general motion of the water molecules (twisting, scissoring and rocking). Modes in S12 C are O-H stretches. The modes for the substituted cluster are not so clearly ordered and their nature is difficult to classify due to the low symmetry, particularly in the lower frequencies < 400 cm^{-1}. Similar to the non-aquo complexes, figure S13 lower frequency modes correspond to water and μ_2-OH rocking and twisting motion, but the exact motion must be examined on a case-by-case basis. High frequency modes correspond with OH stretches on the cluster or on the water molecules. These optimised structures lie on a flat potential energy surface due primarily to the explicit water molecules. It is noticeable that in the low frequency region the Pa modes are slightly smaller than the group. This is in contrast the non-aquo complexes where Th or Pu set the limits to the frequency of a mode. As this is happening in the lower frequency range the calculated G_{corr} values are smaller for Pa than expected leading to a more stable (more negative $\varepsilon_0 + G_{corr}$) Pa cluster than expected given the periodic trend, more negative reaction energy, and ultimately lowering R^2 at the $\Delta_r G^o$ level.

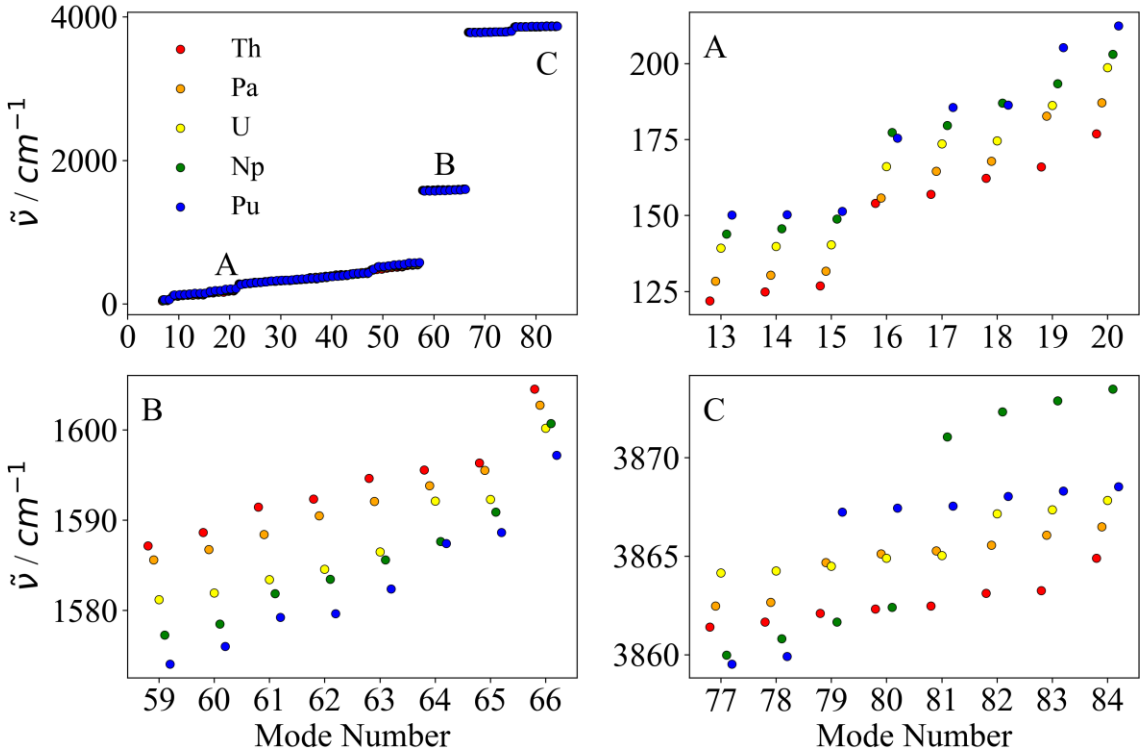

Figure S13: 3N-6 vibrational frequencies of the optimised $[An(H_2O)_9]^{4+}$ species. A, B, and C are zoomed in sections of the plot in the top right.

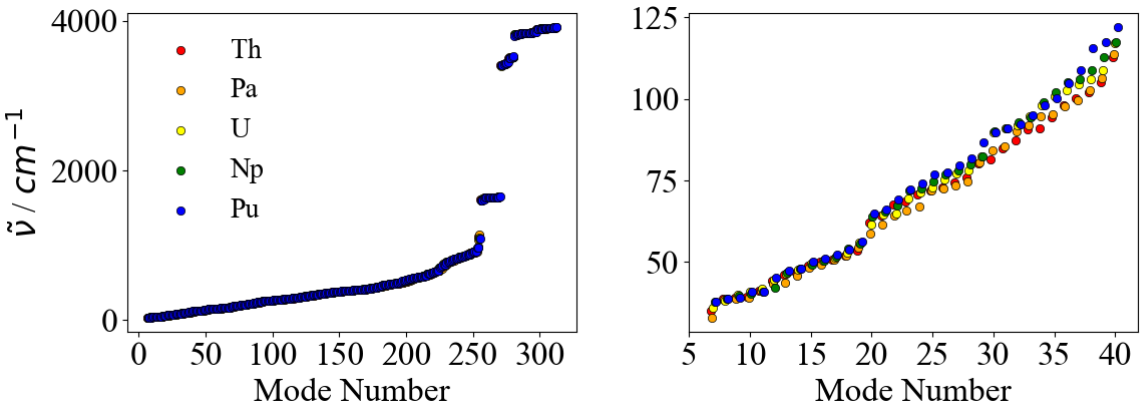

Figure S14: 3N-6 vibrational frequencies of the optimised $[\{An(H_2O)_5\}\{Na(H_2O)_2\}_5Fe_{13}O_{28}H_{12}]^{4+}$ species. Right plot is zoomed in on frequencies < 100 cm^{-1}.

Table S19: Summary of the number of frequencies < 100 cm^{-1} and the lowest calculated frequency for $[\{An(H_2O)_5\}\{Na(H_2O)_2\}_5Fe_{13}O_{28}H_{12}]^{4+}$.

An	#Frequencies < 100 cm^{-1}	Lowest Frequency / cm^{-1}
Th	30	35.1
Pa	31	32.9
U	28	35.9
Np	28	37.9
Pu	28	37.8

In conclusion, we have implemented a RRHO and QRRHO model to investigate the source of lower correlation between the reaction energy and ionic radii of the actinides at the $\Delta_r G^o$ level. In doing so, we have found that Turbomole's freeH script does not include the electronic partition function q_{elec} and therefore ignores electronic contributions to the total entropy leading to error in the calculation of G_{corr} for open-shell systems. The correlation improves when using the QRHHO model which is evidence that the vibrational contribution to G_{corr} from low frequency modes is a reason for the lower correlation. Through careful investigation of vibrational modes, we conclude that the low vibrational frequencies in Pa-Fe_{13} do not behave as expected leading to a lower Gibbs energy resulting in lower correlation between the reaction energy and ionic radii of the actinides.

Mulliken Analysis

In table 3 of the main text, we note that the n(d) = n_{excess}(d) as the tetravalent actinides discussed are formally d^0. To investigate the source of the d populations, we performed Mulliken analysis on all Kohn-Sham orbitals and filtered them to obtain a set of occupied orbitals with An(d) character. This was achieved using a batch script which interfaces with Multiwfn[9] to run the orbital composition analysis with Mulliken partitioning (Multiwfn command 8-1) on all MOs and write to an output file. The output file was then analysed using a python script which finds orbitals with An(d) contribution and sums up the total contribution. This method is reliant on the printing threshold of Multiwfn (*compthrs*) of which the default is 0.5%. To verify this method, we calculated the total populations at different printing thresholds. Table S20 shows that as the threshold is lowered the computed total population approaches the true total populations (Multiwfn command 7-5-1).

Table S20: Total populations calculated from the summation of Kohn-Sham MO occupations. This example is for the Th substituted cluster with different printing thresholds (*compthr*).

Printing Threshold	*n*(s)	*n*(p)	*n*(d)	*n*(f)	#occupied orbitals with d character	#occupied orbitals with f character
Total Population	11.89	29.82	30.94	14.41	-	-
0.001	11.89	29.82	30.94	14.41	507	442
0.01	11.89	29.81	30.91	14.39	398	329
0.1	11.87	29.71	30.78	14.24	204	122
0.5	11.89	29.49	30.28	14.04	75	28

The default threshold is usually reasonable as the most involved orbitals will be printed, although even with this setting the number of occupied spin-orbitals with d contribution is large. With a threshold of 0.001% the total population is captured exactly (to two decimal places) showing that 507 spin-orbitals contribute to n_{excess}(d). Taking Th as an example, table S21 shows that the highest individual contribution from these 507 spin-orbitals is 2.59% and 3.13% for the α- and β-MOs respectively. The source of n_{excess}(d) is a large number of MOs with minor contributions, by contrast to n_{excess}(f) where the majority of the contribution comes from far fewer MOs such as those in figure 5 with contributions an order of magnitude larger. With such a large spread of small contributions it is difficult to draw conclusions about 6d covalency.

Table S21: Source of n_{excess}(d) in $[\{An(H_2O)_5\}\{Na(H_2O)_2\}_5Fe_{13}O_{28}H_{12}]^{4+}$.

An	#occupied orbitals with d character	Highest α orbital %	Highest β orbital %
Th	507	2.59	3.13
Pa	508	2.57	3.41
U	503	1.97	2.78
Np	502	2.35	2.49
Pu	508	2.24	2.58

References for Supplementary Information

1. G. R. Choppin and E. N. Rizkalla, Lanthanides/Actinides: Chemistry, in *Handbook on the Physics and Chemistry of Rare Earths*, ed. K. A. Gscheidner Jr., L. Eyering, G. R. Choppin and G. H. Lander, Elsevier Science, Amsterdam, The Netherlands, 1994, vol. 18, ch. 128, pp 560.
2. N. L. Banik, V. Vallet, F. Réal, R. M. Belmecheri, B. Schimmelpfennig, J. Rothe, R. Marsac, P. Lindqvist-Reis, C. Walther, M. A. Denecke and C. M. Marquardt, *Dalton Trans.*, 2016, **45**, 453.
3. D. A. McQuarrie and J. D. Simon, in *Molecular Thermodynamics*, ed. A. McGuire, University Science Books, Sausalito, 1st edn., 1999, ch. 4.
4. C. J. Cramer, in *Essentials of Computational Chemistry: Theories and Models*, John Wiley & Sons, Chinchester, England, 2nd edn., 2004, ch. 10.
5. F. Jensen, in *Introduction to Computational Chemistry*, John Wiley & Sons, Chinchester, England, 2nd edn., 2007, ch. 13.
6. J. W. Ochterski, *Thermochemistry in Gaussian*, 2000, https://gaussian.com/thermo/.
7. M. E. Simms, M. M. Sibley, D. M. Driscoll, V. Kertesz, J. T. Damron, A. S. Ivanov, F. D. White and N. A. Thiele, *Inorg. Chem.*, 2023, **62**, 20834–20843.
8. R. F. Ribeiro, A. V. Marenich, C. J. Cramer and D. G. Truhlar, *J. Phys. Chem. B*, **2011**, **115**, 14556–14562.
9. T. Lu and F. Chen, *J. Comput. Chem.*, 2012, **33**, 580-592.

Atomic Coordinates and Total Energies

The nine coordinate aqueous actinide species have been provided optimised using PBE, TPSS, PBE0, and TPSSh functionals in combination with the def2-SVP basis. The energies for these structures have been provided at the def2-TZVP/Functional level where the functional is given in brackets. All other structures were optimised using the def2-SVP/PBE0, and where applicable both the def2-TZVP/ECP/PBE0 and Def2TZVP/SARC-DKH2/PBE0 single point electronic energy has been provided. Energies can be correlated with supporting information tables S2 – S7 above.

$[Th(H_2O)_9]^{4+}$ E = -1094.858261 H (PBE)

28
ThH18O9

Th	-0.002799900	-0.003654000	-0.002751600
O	-0.004591600	0.001897600	2.446793300
H	0.526584200	0.560375300	3.051651000
H	-0.606545600	-0.521473800	3.015967000
O	-1.425191300	-0.692718000	-1.871832200
H	-1.158629700	-0.882153300	-2.795346600
H	-2.378816000	-0.910031700	-1.812193700
O	-2.088513900	-0.985512000	0.838987700
H	-2.897225100	-0.495981500	1.095781500
H	-2.279569100	-1.934356900	0.990208100
O	1.490950900	-0.536354000	-1.867865200
H	1.785489200	0.106685300	-2.545964200
H	1.856114100	-1.404588600	-2.137816500
O	-0.092395000	1.772228300	-1.696160400
H	0.432000200	2.599629400	-1.702018000
H	-0.655246100	1.794880600	-2.497705200
O	2.162380300	-0.789430800	0.851203300
H	2.388449500	-0.960504400	1.788826500
H	2.969891200	-0.984139500	0.331859200
O	1.371596800	1.899503300	0.695160900
H	1.075982400	2.801743900	0.936959400
H	2.337759100	1.875980800	0.857065500
O	-1.544026600	1.764521600	0.702861100
H	-1.861705200	1.966329800	1.607390700
H	-1.916484000	2.459183900	0.120691700
O	0.122837500	-2.445747500	-0.108277100
H	0.791346500	-2.987080800	0.360930500
H	-0.399643000	-3.069233200	-0.654406700

$[Pa(H_2O)_9]^{4+}$ E = -1128.546352 H (PBE)

28
PaH18O9

Pa	-0.002258100	-0.003793600	-0.002452200
O	-0.005120700	0.000704000	2.421242000
H	0.528078900	0.565080200	3.018201700
H	-0.613166900	-0.512754500	2.992144200
O	-1.427840200	-0.727697600	-1.821454800
H	-1.166970800	-0.905515300	-2.748523400
H	-2.367899800	-0.990378300	-1.740496800
O	-2.029852900	-1.012187500	0.848662200
H	-2.855011800	-0.541069300	1.086961900
H	-2.187887300	-1.962032700	1.029121800

O	1.439506300	-0.490234200	-1.887391700
H	1.686967500	0.152036100	-2.584111200
H	1.813072800	-1.353985500	-2.158923000
O	-0.139131800	1.751943000	-1.663523400
H	0.396720800	2.571584400	-1.689719700
H	-0.736326600	1.782527600	-2.439534000
O	2.156330900	-0.741538900	0.809446700
H	2.387702800	-0.944293700	1.739431600
H	2.968667700	-0.885231800	0.281047400
O	1.340076400	1.886083600	0.705773400
H	1.026695700	2.780636500	0.952168300
H	2.302380400	1.867896900	0.887173600
O	-1.532645600	1.736472900	0.708397400
H	-1.842683700	1.925664700	1.617898800
H	-1.895319000	2.444335500	0.136841600
O	0.191813700	-2.414138300	-0.128961700
H	0.896268100	-2.940856100	0.301748000
H	-0.332167100	-3.039257900	-0.671168900

$[U(H_2O)_9]^{4+}$ E = -1164.066871 H (PBE)

28
UH18O9

U	-0.002662000	0.002635100	-0.000711400
O	-0.010173100	0.008094200	2.389081100
H	0.342244600	-0.678410100	2.992308800
H	-0.270634100	0.770406700	2.946494600
O	-2.286451000	0.687148400	-0.138110200
H	-3.028100300	0.219112500	0.298215800
H	-2.681082300	1.383975600	-0.702152800
O	-0.219481300	2.251517600	0.842473800
H	0.513761300	2.862385200	1.061435100
H	-1.052857100	2.724940000	1.043134800
O	-0.952477500	-1.168404100	-1.853461000
H	-0.424231800	-1.615966500	-2.546451000
H	-1.887826000	-1.201345100	-2.142545400
O	1.683358200	-0.508737400	-1.651022800
H	2.238835100	-1.314407800	-1.683462300
H	1.946936300	0.048576200	-2.411794200
O	1.250296100	-1.908399700	0.700114800
H	0.855040500	-2.784810200	0.887920300
H	2.193574700	-1.960223500	0.959048900
O	2.138126100	0.807909700	0.691505000
H	2.920075000	0.863084700	0.104104100
H	2.445550300	1.024001200	1.596101000
O	-0.142426200	1.571584100	-1.797360000
H	-0.051715800	2.540817500	-1.688053200
H	-0.400131500	1.412895500	-2.728924600
O	-1.469907200	-1.733786500	0.810700600
H	-1.764488400	-1.855540900	1.736516000
H	-1.883152700	-2.449052500	0.284894200

$[Np(H_2O)_9]^{4+}$ E = -1201.563178 H (PBE)

28
NpH18O9

Np	0.039667300	-0.009687900	-0.040915200
O	-0.018415800	0.017112400	2.340947600
H	0.280367000	-0.664380300	2.977528900
H	-0.200000300	0.830954100	2.855014100
O	-2.260666300	0.599096200	-0.136331900
H	-2.965927800	0.020954000	0.221060200
H	-2.701357000	1.312238900	-0.642596500
O	-0.293385500	2.224061900	0.834861700
H	0.409152500	2.861246500	1.076970900
H	-1.150425200	2.663442700	1.009197200
O	-0.950725400	-1.148182500	-1.863029700
H	-0.457679700	-1.666216500	-2.533390000
H	-1.889400100	-1.139580500	-2.143676000
O	1.711946700	-0.463901900	-1.657561600
H	2.293498100	-1.252175700	-1.684107400
H	1.957021500	0.097208900	-2.422696700
O	1.263302900	-1.927341400	0.633739900
H	0.830295000	-2.761131000	0.912561000
H	2.205963300	-1.995138000	0.893738000
O	2.106526700	0.835085900	0.742615700
H	2.889353700	1.013997800	0.180474500
H	2.376324100	1.004532900	1.669112500
O	-0.100742300	1.613288800	-1.764548900
H	-0.142059000	2.575798100	-1.584790200
H	-0.332173600	1.489318700	-2.708902500
O	-1.405392500	-1.761003800	0.807422700
H	-1.680589900	-1.885901900	1.738293000
H	-1.814478400	-2.483696200	0.289008600

$[Pu(H_2O)_9]^{4+}$ E = -1241.020176 H (PBE)

28
PuH18O9

Pu	-0.006918600	0.002358900	0.000567300
O	1.621871000	-0.003266000	1.709558000
H	1.740906500	-0.772773700	2.305180900
H	2.432582600	0.543248200	1.779731300
O	-1.728846100	1.310196200	-0.935937000
H	-2.580950900	1.575491000	-0.530486700
H	-1.796681300	1.498335800	-1.895687100
O	1.625648900	1.569042700	-0.667293800
H	2.479517000	1.404826200	-1.119559200
H	1.664589500	2.477529500	-0.300843400
O	-1.765476800	-0.047376500	1.571610400
H	-2.555480800	-0.627540500	1.584262400
H	-1.927678000	0.672838100	2.216695100
O	-1.567377200	-1.547400200	-0.850788000
H	-1.612104700	-2.475684600	-0.538702800
H	-2.376953000	-1.383082800	-1.378595000
O	0.071370100	-2.097008900	1.177318500
H	-0.467365100	-2.339276100	1.958154900
H	0.630252100	-2.871766300	0.962521600
O	1.786841700	-1.278096500	-0.840075000

H	1.924818400	-1.403400400	-1.802533700
H	2.612680100	-1.557700100	-0.392355100
O	0.103554900	0.053786200	-2.404535200
H	0.594936000	0.705562600	-2.945247400
H	-0.352061900	-0.549811600	-3.026458200
O	-0.158747300	2.053245200	1.247702500
H	0.310722900	2.250173100	2.084044800
H	-0.703650200	2.837550700	1.031749900

$[Th(H_2O)_9]^{4+}$ E = -1095.500413 H (TPSS)

28
ThH18O9

Th	-0.003951500	-0.003447400	-0.002584200
O	-0.003158100	0.016846500	2.444944300
H	0.529670600	0.581559300	3.040949800
H	-0.595808000	-0.508686100	3.020341300
O	-1.404592800	-0.711494600	-1.878071700
H	-1.122749300	-0.900995700	-2.796005600
H	-2.360860500	-0.916483200	-1.834225700
O	-2.085681400	-0.979577000	0.833759900
H	-2.893269400	-0.486826900	1.084895300
H	-2.278043200	-1.927062400	0.986664600
O	1.485610100	-0.541050200	-1.866022600
H	1.795775000	0.096234400	-2.541290900
H	1.841365000	-1.414657700	-2.127778400
O	-0.109338300	1.769615700	-1.689147000
H	0.415724400	2.595534500	-1.700688600
H	-0.673147300	1.785695900	-2.489130500
O	2.153782300	-0.789699300	0.852677400
H	2.375839100	-0.962626200	1.790101400
H	2.960238700	-0.986279500	0.334013200
O	1.363868500	1.900194900	0.695628500
H	1.061040200	2.798419800	0.939557700
H	2.330560000	1.883706900	0.849851600
O	-1.528737200	1.767878400	0.723785900
H	-1.831581700	1.965261800	1.633315500
H	-1.914827800	2.458704600	0.147499300
O	0.123009500	-2.442270100	-0.126688800
H	0.774287500	-2.994193100	0.352363300
H	-0.405024500	-3.054303100	-0.678715200

$[Pa(H_2O)_9]^{4+}$ E = -1129.175116 H (TPSS)

28
PaH18O9

Pa	-0.002783800	-0.003069200	-0.001969800
O	-0.006875100	0.011340300	2.417699500
H	0.527067100	0.579497100	3.008955800
H	-0.610992800	-0.501947500	2.991494200
O	-1.410243700	-0.746226200	-1.822818400
H	-1.139357900	-0.921789400	-2.746507500
H	-2.352292700	-1.001349700	-1.751985100
O	-2.027292500	-1.005892300	0.844587700
H	-2.854100100	-0.533812200	1.071805100

H	-2.184607200	-1.954048800	1.029943200
O	1.431076000	-0.489634900	-1.886851600
H	1.686373200	0.150643500	-2.581416100
H	1.804838600	-1.354635600	-2.150975200
O	-0.152444100	1.748549500	-1.658293700
H	0.384411800	2.566245500	-1.692537500
H	-0.756272600	1.775614800	-2.428220700
O	2.149643800	-0.741566200	0.810355900
H	2.374886200	-0.953549000	1.738975600
H	2.962478900	-0.884574100	0.284090500
O	1.335142600	1.881450400	0.709842200
H	1.019637400	2.775393500	0.952297400
H	2.298086300	1.866282900	0.883713800
O	-1.515703400	1.741312200	0.722464200
H	-1.817786100	1.925966000	1.634616300
H	-1.888479300	2.445283600	0.154094500
O	0.193572400	-2.408157500	-0.141794300
H	0.885247500	-2.942666900	0.297944600
H	-0.333230700	-3.024659700	-0.689510600

$[U(H_2O)_9]^{4+}$ E = -1164.676900 H (TPSS)

28
UH18O9

U	-0.001078800	0.002376800	-0.001606300
O	0.006827700	0.006478500	2.384144000
H	0.357017300	-0.685602300	2.980779100
H	-0.275348900	0.755099200	2.947901100
O	-2.280255000	0.687561500	-0.154851100
H	-3.025544500	0.236778700	0.291409600
H	-2.665424200	1.381956800	-0.726729800
O	-0.219091700	2.245070100	0.835379100
H	0.513533600	2.850056400	1.068563300
H	-1.053139800	2.715727600	1.036058200
O	-0.961007600	-1.156368700	-1.851296400
H	-0.444735200	-1.624268400	-2.538617600
H	-1.897625300	-1.174102700	-2.134437800
O	1.678476900	-0.506945500	-1.647282700
H	2.235639700	-1.310523300	-1.678284300
H	1.936224800	0.048698100	-2.410189000
O	1.255838600	-1.897370200	0.707998800
H	0.871155100	-2.780429500	0.881519300
H	2.198990700	-1.939928400	0.965513500
O	2.135059900	0.795365000	0.704441200
H	2.916776800	0.875337000	0.121083000
H	2.432548500	1.002383100	1.613428900
O	-0.153575800	1.562068900	-1.799330300
H	-0.054836600	2.530423500	-1.697280500
H	-0.413020000	1.396029200	-2.728125100
O	-1.463043100	-1.726378000	0.810079400
H	-1.752665100	-1.850360500	1.736284200
H	-1.877698200	-2.439132900	0.283448400

$[Np(H_2O)_9]^{4+}$ E = -1202.163854 H (TPSS)

28
NpH18O9

Np	0.036083500	-0.006735800	-0.035232700
O	0.114009200	-0.034927400	2.362982800
H	0.510654600	-0.709657300	2.949291700
H	-0.181074600	0.704844400	2.930957500
O	-2.266840200	0.619267200	-0.282053700
H	-3.006205800	0.156766800	0.160841800
H	-2.658769100	1.281739400	-0.885277600
O	-0.390877300	2.166953100	0.871122500
H	0.263728300	2.811557500	1.206964200
H	-1.275765000	2.573941000	0.961921100
O	-0.962405400	-1.184975700	-1.824512900
H	-0.482363600	-1.716874100	-2.492036800
H	-1.900570700	-1.155110500	-2.101980000
O	1.702048500	-0.453336500	-1.645904600
H	2.296208200	-1.231291200	-1.666902200
H	1.929281500	0.099172500	-2.421642300
O	1.256611400	-1.932964100	0.582920700
H	0.822016700	-2.771423800	0.841547000
H	2.210477100	-2.033607800	0.779867700
O	2.085380900	0.875143200	0.750604900
H	2.870144500	1.068646400	0.197718900
H	2.350314400	1.012318200	1.682821000
O	-0.059174500	1.606062600	-1.758929500
H	-0.073356800	2.568858000	-1.581646500
H	-0.233402300	1.488573100	-2.715224400
O	-1.423487800	-1.666632800	0.885148600
H	-1.613218500	-1.774512600	1.838659600
H	-1.919447200	-2.361793500	0.407973300

$[Pu(H_2O)_9]^{4+}$ E = -1241.601367 H (TPSS)

28
PuH18O9

Pu	-0.001353200	0.000189600	-0.003815600
O	0.002601700	-0.005152900	2.347774600
H	0.089056100	-0.823868700	2.877858400
H	0.129069300	0.749516000	2.958698400
O	-1.203999700	0.787336700	-1.863952400
H	-2.107266200	0.540011600	-2.149715900
H	-0.788790400	1.273393900	-2.605777300
O	0.582306000	2.183616500	0.643371400
H	1.463817800	2.524132000	0.899258800
H	-0.077296300	2.855072400	0.913802000
O	-1.973378200	-1.276619700	-0.077707100
H	-2.154523000	-2.077417800	-0.611304200
H	-2.830947000	-0.966961400	0.279533000
O	0.325304700	-1.597408500	-1.696774600
H	0.611499100	-2.514874700	-1.508847000
H	0.046464700	-1.566367500	-2.634893800
O	0.461246100	-2.176231500	0.884120900
H	-0.196952300	-2.868899600	1.094050100
H	1.340112200	-2.528548100	1.130485300
O	2.259391500	-0.091416600	0.637451700

H	2.990421000	0.173814100	0.042419700
H	2.635097200	-0.177885400	1.537468800
O	1.438376600	0.941122500	-1.669005100
H	1.729067900	1.873951300	-1.718239000
H	1.839321100	0.472683600	-2.428552500
O	-1.894622100	1.236393900	0.779499500
H	-2.206500400	1.287636000	1.705325900
H	-2.507524000	1.772782100	0.237466100

$[Th(H_2O)_9]^{4+}$ E_{ECP} = -1094.844441 H, E_{AE} = -27103.083755 H (PBE0)

28
ThH18O9

Th	0.000000000	0.000000000	0.000000000
O	0.000000000	0.000000000	2.440486237
H	0.530123816	0.551706995	3.036350912
H	-0.594027612	-0.519541539	3.004413002
O	-1.410250357	-0.695981509	-1.864363846
H	-1.139428179	-0.880060907	-2.777130427
H	-2.355816890	-0.905143705	-1.810944323
O	-2.079189771	-0.973552194	0.835444487
H	-2.879636389	-0.486254578	1.084682446
H	-2.267984000	-1.912554533	0.986828978
O	1.481452449	-0.542503649	-1.861827648
H	1.782381938	0.088443966	-2.534124021
H	1.836637097	-1.407407911	-2.119392830
O	-0.083299532	1.765832878	-1.685964725
H	0.436190524	2.584605859	-1.687463671
H	-0.642189183	1.790167904	-2.478098140
O	2.160037189	-0.773864466	0.841286801
H	2.390212565	-0.946338460	1.767317779
H	2.954574566	-0.962446530	0.318305905
O	1.358247452	1.900093096	0.709368481
H	1.056516769	2.790381762	0.947575458
H	2.315441922	1.883969369	0.864943433
O	-1.532928456	1.763278306	0.708365332
H	-1.842727121	1.958517195	1.606347316
H	-1.906682641	2.450046100	0.134392227
O	0.123220077	-2.434262196	-0.101389081
H	0.778078011	-2.975346613	0.366677656
H	-0.400940807	-3.045549434	-0.641830055

$[Pa(H_2O)_9]^{4+}$ E_{ECP} = -1128.513287 H, E_{AE} = -27859.183331 H (PBE0)

28
PaH18O9

Pa	0.000000000	0.000000000	0.000000000
O	0.000000000	0.000000000	2.412735849
H	0.532263998	0.556034876	3.002155608
H	-0.599780041	-0.511260775	2.977448925
O	-1.415064873	-0.725370959	-1.812997985
H	-1.153937544	-0.893801951	-2.731295704
H	-2.348268171	-0.976962605	-1.736897621
O	-2.021558445	-1.001967467	0.847125675
H	-2.838611598	-0.533818680	1.078093883

H	-2.177769875	-1.942309702	1.024636750
O	1.432196586	-0.492528031	-1.878384621
H	1.687924803	0.141371990	-2.565890183
H	1.802112106	-1.350304314	-2.137957311
O	-0.132324846	1.747139400	-1.654857426
H	0.398844770	2.557923174	-1.677884613
H	-0.725768327	1.778562140	-2.421050596
O	2.153554509	-0.729445063	0.802218166
H	2.387108397	-0.932778180	1.721013876
H	2.953696523	-0.868108867	0.272402057
O	1.329142877	1.884026078	0.713339607
H	1.014509950	2.769730273	0.950723652
H	2.283286868	1.870964451	0.884696086
O	-1.522579312	1.733083964	0.711488518
H	-1.831862089	1.914312386	1.612244155
H	-1.889268883	2.429407951	0.145169717
O	0.191613727	-2.400526996	-0.123918982
H	0.881312577	-2.926094776	0.309723556
H	-0.335053518	-3.016218923	-0.656402804

$[U(H_2O)_9]^{4+}$ E_{ECP} = -1164.034512 H, E_{AE} = -28629.674785 H (PBE0)

28
UH18O9

U	0.000000000	0.000000000	0.000000000
O	0.000000000	0.000000000	2.379889836
H	0.344500631	-0.686530028	2.971386567
H	-0.274918364	0.744327676	2.937144766
O	-2.273560012	0.685359188	-0.144009319
H	-3.011477704	0.237801960	0.297838021
H	-2.654969188	1.380779741	-0.701641621
O	-0.217774977	2.235681812	0.838296076
H	0.508476047	2.835473131	1.066620554
H	-1.044374571	2.707264179	1.022358115
O	-0.957309034	-1.157045144	-1.844379787
H	-0.447878925	-1.624101154	-2.524425253
H	-1.887107157	-1.181956620	-2.118035680
O	1.674343563	-0.511042562	-1.639761085
H	2.229471246	-1.305197651	-1.662645495
H	1.927633224	0.034165060	-2.400100715
O	1.254344797	-1.897571268	0.701163571
H	0.875254895	-2.772810266	0.875325362
H	2.191313460	-1.939272923	0.946875171
O	2.128947555	0.798963746	0.699440861
H	2.903296770	0.881658585	0.121813213
H	2.419532403	1.017736702	1.598151462
O	-0.141416069	1.558254510	-1.791782422
H	-0.045690374	2.518015760	-1.692923947
H	-0.383624839	1.389879692	-2.715337564
O	-1.457104656	-1.725701505	0.806993085
H	-1.739675793	-1.852999106	1.725343556
H	-1.873593657	-2.425888414	0.281838156

$[Np(H_2O)_9]^{4+}$ E_{ECP} = -1201.515171 H, E_{AE} = -29415.337443 H (PBE0)

28
NpH18O9

Np	0.000000000	0.000000000	0.000000000
O	0.000000000	0.000000000	2.378381814
H	0.323011639	-0.687179881	2.980410635
H	-0.255425075	0.761299604	2.921090133
O	-2.291453447	0.616466475	-0.155047501
H	-3.011444683	0.110777823	0.251372147
H	-2.696727564	1.323898495	-0.678949652
O	-0.355165325	2.199769532	0.875282723
H	0.330723412	2.823056784	1.158313858
H	-1.211908392	2.628856787	1.019462429
O	-0.994924091	-1.145964916	-1.803813913
H	-0.518783927	-1.665698059	-2.470238122
H	-1.926348492	-1.122736411	-2.072347353
O	1.658908791	-0.454693240	-1.603748575
H	2.244607070	-1.227747537	-1.616225484
H	1.889431837	0.090897231	-2.371966902
O	1.236053484	-1.897445513	0.664475522
H	0.830870095	-2.748368876	0.893065083
H	2.174263141	-1.955006029	0.903863941
O	2.051936150	0.860559888	0.781703446
H	2.830171174	1.044094376	0.232511202
H	2.307920113	1.023616997	1.702573681
O	-0.130346699	1.600337066	-1.727058604
H	-0.127033699	2.557667376	-1.571943621
H	-0.332292812	1.464417642	-2.665947695
O	-1.433405530	-1.714787895	0.858511042
H	-1.673413481	-1.841108248	1.788704752
H	-1.878104578	-2.408949184	0.349051119

$[Pu(H_2O)_9]^{4+}$ E_{ECP} = -1240.964737 H, E_{AE} = -30215.893594 H (PBE0)

28
PuH18O9

Pu	0.000000000	0.000000000	0.000000000
O	0.000000000	0.000000000	2.348357592
H	0.009269899	-0.801272004	2.894853348
H	0.090697923	0.756568147	2.948736388
O	-1.237299004	0.761325259	-1.840044100
H	-2.129335273	0.488724705	-2.104136067
H	-0.859908665	1.269471310	-2.574467461
O	0.581083251	2.185608053	0.634347474
H	1.462684615	2.530488555	0.846183896
H	-0.055493140	2.891147740	0.829125019
O	-1.944223944	-1.308488426	-0.103292958
H	-2.098783878	-2.094526103	-0.649220907
H	-2.799112197	-1.039484231	0.266654520
O	0.325640353	-1.561885079	-1.719104433
H	0.626625237	-2.470857791	-1.567033184
H	0.034216008	-1.503788725	-2.641912953
O	0.426980534	-2.169198048	0.865264516

H	-0.239002352	-2.850748240	1.042478193
H	1.289952852	-2.537494763	1.108967653
O	2.259961706	-0.109545439	0.629806951
H	3.002067989	0.072301514	0.032269954
H	2.632978992	-0.246854836	1.514708812
O	1.410051087	0.918285379	-1.674385234
H	1.711705148	1.838018258	-1.727390684
H	1.786594545	0.446506131	-2.432788615
O	-1.899169436	1.210780547	0.745923651
H	-2.209458439	1.282535285	1.661497775
H	-2.506726083	1.725517584	0.193233821

$[Th(H_2O)_9]^{4+}$ E = -1095.431361 H (TPSSh)

28
ThH18O9

Th	-0.003848000	-0.002842200	-0.002265000
O	-0.003339800	0.017977100	2.442638300
H	0.527706000	0.581139600	3.035132800
H	-0.592790200	-0.505855100	3.016026300
O	-1.402522200	-0.710887300	-1.876171300
H	-1.120182700	-0.901085000	-2.789547500
H	-2.355018200	-0.914488800	-1.833460600
O	-2.082163900	-0.978355000	0.833110900
H	-2.886938400	-0.488627700	1.083533200
H	-2.272090300	-1.922559800	0.983599500
O	1.483777000	-0.540922100	-1.863968700
H	1.794207300	0.093621000	-2.535863600
H	1.838423400	-1.411341500	-2.122829900
O	-0.108390500	1.766493500	-1.687172200
H	0.412916600	2.590037600	-1.699067100
H	-0.672186700	1.781579600	-2.482280800
O	2.151062200	-0.786842200	0.851627300
H	2.371485500	-0.960438600	1.785127500
H	2.952961400	-0.985560600	0.334280600
O	1.362490700	1.898463300	0.696426200
H	1.061126200	2.793696900	0.937032400
H	2.325469400	1.881781000	0.848256200
O	-1.527075900	1.766050800	0.723397200
H	-1.826469400	1.962026800	1.630067500
H	-1.914052500	2.453040500	0.149975500
O	0.121872900	-2.439194200	-0.127295700
H	0.767723900	-2.991588600	0.350335400
H	-0.404153500	-3.045319000	-0.680644500

$[Pa(H_2O)_9]^{4+}$ E = -1129.099386 H (TPSSh)

28
PaH18O9

Pa	-0.002554900	-0.002762500	-0.001815200
O	-0.005971900	0.010855200	2.414569300
H	0.528638700	0.573784800	3.003612000
H	-0.607835800	-0.500742700	2.985419900
O	-1.408100000	-0.744652100	-1.820368400
H	-1.138970200	-0.915535600	-2.741216800

H	-2.346717700	-0.997143100	-1.749871300
O	-2.024584400	-1.004834900	0.844150400
H	-2.848466600	-0.535132400	1.069415200
H	-2.179667100	-1.949292000	1.028834200
O	1.430289400	-0.489201700	-1.883306800
H	1.685845200	0.147587700	-2.575345800
H	1.804962400	-1.350789900	-2.142436400
O	-0.152693100	1.746298600	-1.656448200
H	0.383339000	2.559639900	-1.691344900
H	-0.755105900	1.773136900	-2.422313800
O	2.147333400	-0.740066700	0.809536500
H	2.371520000	-0.952185300	1.734098800
H	2.956443500	-0.881268500	0.284609900
O	1.333250500	1.879958900	0.707463100
H	1.018513400	2.770892100	0.945840900
H	2.292208100	1.864929200	0.880813500
O	-1.514127200	1.738298700	0.721998100
H	-1.816794100	1.920480400	1.630152900
H	-1.886430700	2.438279300	0.155265500
O	0.192141700	-2.404708400	-0.142149600
H	0.877693400	-2.938842600	0.298761700
H	-0.334159200	-3.016983200	-0.687924500

$[U(H_2O)_9]^{4+}$ E = -1164.603227 H (TPSSh)

28
UH18O9

Np	0.032823600	-0.006091100	-0.032250200
O	0.108211900	-0.034096000	2.359671400
H	0.494211000	-0.713566100	2.940941600
H	-0.199737400	0.693281500	2.929925000
O	-2.264311100	0.619408800	-0.276031400
H	-3.003176500	0.171634200	0.173896500
H	-2.649343400	1.289570600	-0.868637500
O	-0.380325500	2.166648200	0.865669200
H	0.278452300	2.801237200	1.200864400
H	-1.257734000	2.579300000	0.960057500
O	-0.963375500	-1.180465600	-1.822481800
H	-0.485354300	-1.707372400	-2.489355200
H	-1.898782800	-1.153285000	-2.095055400
O	1.694871400	-0.453090600	-1.640464300
H	2.286021300	-1.228238600	-1.660325400
H	1.921641900	0.098395100	-2.411913900
O	1.262907000	-1.920442300	0.592200100
H	0.842793700	-2.767329900	0.830973200
H	2.212891400	-2.006481900	0.794362900
O	2.082624200	0.871141300	0.750879400
H	2.865258900	1.060639700	0.200933900
H	2.342858000	1.014760400	1.679128500
O	-0.069465100	1.595985300	-1.761337500
H	-0.064572700	2.557148700	-1.597816100
H	-0.249227500	1.468468300	-2.711114600
O	-1.417979000	-1.671419800	0.874862700
H	-1.611525400	-1.787818300	1.822435300
H	-1.910656300	-2.357922000	0.389981900

$[Np(H_2O)_9]^{4+}$ E = H (TPSSh)

28
NpH18O9

Np	0.032823600	-0.006091100	-0.032250200
O	0.108211900	-0.034096000	2.359671400
H	0.494211000	-0.713566100	2.940941600
H	-0.199737400	0.693281500	2.929925000
O	-2.264311100	0.619408800	-0.276031400
H	-3.003176500	0.171634200	0.173896500
H	-2.649343400	1.289570600	-0.868637500
O	-0.380325500	2.166648200	0.865669200
H	0.278452300	2.801237200	1.200864400
H	-1.257734000	2.579300000	0.960057500
O	-0.963375500	-1.180465600	-1.822481800
H	-0.485354300	-1.707372400	-2.489355200
H	-1.898782800	-1.153285000	-2.095055400
O	1.694871400	-0.453090600	-1.640464300
H	2.286021300	-1.228238600	-1.660325400
H	1.921641900	0.098395100	-2.411913900
O	1.262907000	-1.920442300	0.592200100
H	0.842793700	-2.767329900	0.830973200
H	2.212891400	-2.006481900	0.794362900
O	2.082624200	0.871141300	0.750879400
H	2.865258900	1.060639700	0.200933900
H	2.342858000	1.014760400	1.679128500
O	-0.069465100	1.595985300	-1.761337500
H	-0.064572700	2.557148700	-1.597816100
H	-0.249227500	1.468468300	-2.711114600
O	-1.417979000	-1.671419800	0.874862700
H	-1.611525400	-1.787818300	1.822435300
H	-1.910656300	-2.357922000	0.389981900

$[Pu(H_2O)_9]^{4+}$ E = -1241.520780 H (TPSSh)

28
PuH18O9

Pu	-0.003260800	-0.001665500	-0.005847200
O	0.006500400	0.003970500	2.340975100
H	0.071444200	-0.805510700	2.880337100
H	0.126397900	0.762724400	2.941241800
O	-1.205177100	0.779679500	-1.862763300
H	-2.109817800	0.539831000	-2.136127400
H	-0.806979500	1.287240800	-2.593863200
O	0.586601900	2.174183000	0.642430800
H	1.469196900	2.507478400	0.887678300
H	-0.061837500	2.857480900	0.893934600
O	-1.970560000	-1.276253600	-0.085920500
H	-2.142078600	-2.079911100	-0.610723700
H	-2.822816400	-0.989147600	0.290794500
O	0.318476600	-1.594675200	-1.697376800
H	0.629115300	-2.503089600	-1.527259900
H	0.047543800	-1.551063800	-2.632994900

O	0.460426900	-2.165701600	0.883459800
H	-0.192954800	-2.861203100	1.080263800
H	1.336247800	-2.513275100	1.131646100
O	2.249805400	-0.090529400	0.643848100
H	2.988419600	0.151596300	0.055434600
H	2.613917400	-0.184805200	1.543219900
O	1.432242200	0.938134200	-1.661475300
H	1.727888500	1.865548400	-1.703638900
H	1.829086000	0.472965000	-2.420088300
O	-1.887448400	1.228923900	0.778953500
H	-2.188867100	1.287009400	1.703633800
H	-2.501512800	1.760065800	0.240227600

$[Na(H_2O)_6]^+$ E_{ECP} = -620.520604 H, E_{AE} = -620. 984741 H (PBE0)

19
NaH12O6

Na	-0.0093991	-0.0099930	-0.0083496
O	-0.7388554	-0.1128787	2.2572579
H	-0.3499285	0.2234737	3.0734504
H	-0.4629065	-1.0460946	2.1965766
O	-2.2672425	0.7287581	0.1728325
H	-2.1506708	0.5058216	1.1150324
H	-3.1017270	0.3227563	-0.0908949
O	-0.2224759	2.2134729	-0.8341885
H	0.0642141	3.0639062	-0.4803089
H	-1.1545814	2.1149973	-0.5654541
O	0.7380602	0.1122254	-2.2672844
H	0.4672737	1.0461558	-2.1937922
H	0.3546022	-0.2071409	-3.0929338
O	2.2572480	-0.7250116	-0.1715363
H	3.0838980	-0.3116446	0.1048878
H	2.1518745	-0.5024789	-1.1149889
O	0.2271791	-2.2236885	0.8391401
H	1.1579362	-2.1116406	0.5706180
H	-0.0444988	-3.0809958	0.4899358

Th-Fe_{13} - $[\{Th(H_2O)_5\}\{Na(H_2O)_2\}_5Fe_{13}O_{28}H_{12}]^{4+}$ E_{ECP} = -20903.866774 H, E_{AE} = -47006.151069 H (PBE0)

104
ThNa5Fe13H42O43

Na	3.2203250	-3.5371084	-0.7576347
Na	3.5313728	3.2280491	-0.7310408
Na	0.0097126	0.0183980	-5.4645899
Fe	1.4149258	-1.9756387	1.5518607
Fe	3.4026605	-0.1514433	-1.1079504
Fe	1.8306014	-1.5420642	-3.1381039
Fe	-0.1547881	-3.3687900	-0.4495314
Fe	1.9620186	1.3889859	-3.1271297
Fe	-1.5918145	-1.8463337	1.5633521
Fe	0.0005047	0.0016582	-0.7815765
O	1.4946146	-0.0643580	-1.8011462
O	-0.0724216	-1.4936268	0.2770255
O	-0.0763045	-1.8172965	-3.9644962

O	-1.5293984	-3.1252508	-1.9612477
O	1.2448271	-3.2443613	-1.9590077
O	3.5261917	-1.6091174	-2.2894402
O	3.1029730	-1.5397172	0.5234204
O	1.8811167	-0.1089942	2.5794341
O	3.6566155	1.2992243	-2.2717138
O	-0.0780860	-1.8529583	2.8409291
O	2.1313381	-0.0801806	-4.3000228
O	1.2506459	-3.6589966	0.8435146
O	-1.5761945	-3.5344965	0.8462813
Na	-3.2210680	3.5362176	-0.7430560
Na	-3.5291343	-3.2296218	-0.7673617
Fe	-1.4213903	1.9626383	1.5619243
Fe	-3.4000525	0.1513832	-1.1192633
Fe	-1.8214023	1.5593900	-3.1328055
Fe	0.1547402	3.3695625	-0.4256883
Fe	-1.9512947	-1.3719994	-3.1459601
Fe	1.5855984	1.8343938	1.5807612
O	-1.4895281	0.0709034	-1.8065610
O	0.0709178	1.4896710	0.2876550
O	0.0894590	1.8403875	-3.9480262
O	1.5355598	3.1339043	-1.9327056
O	-1.2399729	3.2557329	-1.9396290
O	-3.2273400	-1.2559004	0.5171805
O	-3.5193225	1.6194011	-2.2888059
O	-3.1045162	1.5324574	0.5247078
O	-1.8795142	0.0739346	2.5545670
O	-3.6493923	-1.2887156	-2.2981346
O	0.0745356	1.8367195	2.8520349
O	-2.1167094	0.1071711	-4.3075856
O	-1.2538927	3.6504284	0.8662122
O	1.5735135	3.5270382	0.8756887
O	3.2199881	1.2456299	0.5431596
H	3.9432275	1.0465670	1.1549300
H	2.7854224	-0.1309669	2.9234071
H	1.4033741	3.8525218	-2.5679280
H	0.1277305	2.7301505	-4.3269422
H	1.0387704	-3.9422775	-2.5976395
H	-0.1130328	-2.7028466	-4.3533191
H	-2.7803593	0.1188991	2.9052664
H	-3.8414816	1.3917733	1.1363068
H	-3.9512214	-1.0618777	1.1298660
H	-1.3964302	-3.8404596	-2.6001624
H	3.8380289	-1.4071093	1.1391484
H	-1.0364316	3.9604235	-2.5716565
H	-2.3199565	5.0325208	0.8431876
O	-3.0708533	5.5246153	0.4419199
H	-3.6593096	5.7273805	1.1790808
H	2.2446164	-0.1978517	-5.9993527
O	1.8436927	-0.2973782	-6.8959320
H	2.1806307	0.4509437	-7.4031464
O	3.0576533	-5.5388970	0.4033114
H	2.3092037	-5.0463745	0.8086571
H	3.6430727	-5.7541941	1.1393252
H	-4.8812382	-2.4493572	-2.4751641

O -5.2956160 -3.3258868 -2.2857550
H -6.1976249 -3.1218376 -2.0112192
H 2.8634090 4.6968596 0.9909091
O 3.7715521 4.9919658 0.7557529
H 3.6733419 5.9113458 0.4802071
O 5.2187373 -3.5800033 -1.9554427
H 4.7634374 -2.7761232 -2.3057078
H 5.3246533 -4.1599501 -2.7190454
O -3.7722410 -4.9999614 0.7106743
H -3.6703994 -5.9179306 0.4317899
H -2.8657626 -4.7035420 0.9512070
O -1.8189028 0.3437031 -6.9004385
H -2.1542120 -0.4005074 -7.4147176
H -2.2232500 0.2374527 -6.0061596
H -4.7565716 2.7866558 -2.3036760
O 5.3060548 3.3335956 -2.2396511
H 4.8919881 2.4587761 -2.4370810
H 6.2068273 3.1270701 -1.9629257
O -5.2129757 3.5889339 -1.9511712
H -5.3145673 4.1732201 -2.7120493
Th -0.0052354 -0.0126145 4.1539666
H -1.5555447 2.6178846 4.8142782
O -1.6920318 1.6841515 5.0312191
H -2.1514734 1.6827535 5.8830413
O -1.8743804 -1.4454961 5.1070899
H -2.6907112 -1.2337958 5.5821923
H -1.8274536 -2.4126527 5.0886541
H 1.7957466 2.4454893 5.0361109
O 1.8224387 1.4800726 5.0982357
H 2.6188854 1.2696185 5.6065062
O 1.6999219 -1.5830783 5.1735966
H 1.9461212 -2.4658886 4.8644919
H 1.8024965 -1.6054998 6.1362549
O 0.0203853 -0.0066319 6.6990223
H 0.3906054 0.6992623 7.2472430
H -0.5213495 -0.5521604 7.2860182

Pa-Fe_{13} - $[\{Pa(H_2O)_5\}\{Na(H_2O)_2\}_5Fe_{13}O_{28}H_{12}]^{4+}$ E_{ECP} = -20937.543881 H, E_{AE} = -47762.260532 H (PBE0)

104
PaNa5Fe13H42O43
Na 3.2215678 -3.5392149 -0.7553169
Na 3.5499412 3.2531753 -0.7575986
Na 0.0093938 0.0178341 -5.4655386
Fe 1.4260701 -1.9597402 1.5596343
Fe 3.4092438 -0.1492916 -1.1120731
Fe 1.8295595 -1.5424991 -3.1368713
Fe -0.1524095 -3.3599054 -0.4348236
Fe 1.9605432 1.3899680 -3.1273317
Fe -1.5987227 -1.8279691 1.5652858
Fe 0.0026141 0.0051068 -0.7828131
O 1.4977820 -0.0633613 -1.8003836
O -0.0691688 -1.4834735 0.2852921
O -0.0768427 -1.8197638 -3.9629647

O	-1.5273548	-3.1233902	-1.9508447
O	1.2436399	-3.2426708	-1.9493260
O	3.5273227	-1.6084034	-2.2935187
O	3.1094186	-1.5373999	0.5161029
O	1.9126134	-0.1406959	2.6091926
O	3.6574375	1.3030262	-2.2765986
O	-0.0843781	-1.8156485	2.8412027
O	2.1283681	-0.0798748	-4.2994848
O	1.2514394	-3.6469889	0.8627591
O	-1.5747632	-3.5212819	0.8625866
Na	-3.2263109	3.5369284	-0.7404773
Na	-3.5299947	-3.2295676	-0.7651645
Fe	-1.4322930	1.9448516	1.5651569
Fe	-3.4034921	0.1508808	-1.1223836
Fe	-1.8224457	1.5576495	-3.1345497
Fe	0.1436347	3.3677471	-0.4208771
Fe	-1.9499358	-1.3743990	-3.1438513
Fe	1.6042817	1.8356196	1.5889742
O	-1.4894847	0.0700670	-1.8059182
O	0.0691354	1.4888434	0.2902579
O	0.0882596	1.8429176	-3.9450533
O	1.5314103	3.1353743	-1.9220651
O	-1.2441577	3.2606566	-1.9357570
O	-3.2317712	-1.2525334	0.5137099
O	-3.5192851	1.6200056	-2.2900911
O	-3.1119838	1.5317342	0.5229226
O	-1.9091997	0.0779714	2.5700677
O	-3.6496870	-1.2901872	-2.3006113
O	0.0740768	1.7875141	2.8493526
O	-2.1163207	0.1048265	-4.3073729
O	-1.2582596	3.6379320	0.8823629
O	1.5651329	3.5266921	0.8920176
O	3.2348411	1.2506495	0.5455759
H	3.9572861	1.0418934	1.1557172
H	2.8137919	-0.1573715	2.9580284
H	1.4013457	3.8526859	-2.5597486
H	0.1264672	2.7305719	-4.3289873
H	1.0355061	-3.9401743	-2.5879983
H	-0.1132903	-2.7038270	-4.3552132
H	-2.8053038	0.1189950	2.9289407
H	-3.8474691	1.3889072	1.1358403
H	-3.9532190	-1.0583424	1.1293498
H	-1.3929020	-3.8387423	-2.5895002
H	3.8364526	-1.4031169	1.1408181
H	-1.0380644	3.9625424	-2.5704553
H	-2.3193054	5.0301704	0.8499368
O	-3.0669913	5.5218207	0.4431132
H	-3.6587963	5.7278485	1.1768221
H	2.2451159	-0.1979738	-5.9999813
O	1.8435629	-0.2972476	-6.8961054
H	2.1807479	0.4507185	-7.4036590
O	3.0526955	-5.5359878	0.4046527
H	2.3076149	-5.0436382	0.8154763
H	3.6420745	-5.7538503	1.1370076
H	-4.8818952	-2.4501003	-2.4758180

O -5.2960707 -3.3264057 -2.2852153
H -6.1982929 -3.1222237 -2.0114192
H 2.8631983 4.6956555 0.9939142
O 3.7692383 4.9897518 0.7514160
H 3.6717265 5.9115280 0.4823712
O 5.2190840 -3.5803625 -1.9552621
H 4.7639079 -2.7767327 -2.3058615
H 5.3250302 -4.1604554 -2.7188776
O -3.7687022 -4.9971811 0.7088192
H -3.6677887 -5.9161910 0.4329247
H -2.8643396 -4.7027897 0.9578955
O -1.8187188 0.3436489 -6.9006984
H -2.1539986 -0.4002408 -7.4154936
H -2.2242003 0.2372966 -6.0071106
H -4.7577604 2.7870245 -2.3046864
O 5.3152846 3.3319958 -2.2522690
H 4.8994492 2.4567290 -2.4443844
H 6.2128387 3.1260504 -1.9652035
O -5.2141668 3.5888697 -1.9515288
H -5.3152970 4.1739232 -2.7119329
Pa -0.0050408 -0.0209777 4.1284095
H -1.5478333 2.5834641 4.7974589
O -1.6574384 1.6499182 5.0288407
H -2.0962607 1.6473851 5.8914026
O -1.8479717 -1.4379364 5.0802130
H -2.6379891 -1.2075263 5.5894863
H -1.8205956 -2.4056363 5.0624233
H 1.7822646 2.4338657 5.0132081
O 1.7933050 1.4680707 5.0704045
H 2.5648688 1.2416673 5.6092597
O 1.6521321 -1.5568388 5.1671962
H 1.9285481 -2.4285345 4.8537295
H 1.7478204 -1.5704417 6.1312912
O 0.0342731 -0.0224805 6.6775779
H 0.4146425 0.6813145 7.2207587
H -0.5574112 -0.5229310 7.2559913

U-Fe$_{13}$ - [{U(H$_2$O)$_5$}{Na(H$_2$O)$_2$}$_5$Fe$_{13}$O$_{28}$H$_{12}$]$^{4+}$ E_{ECP} = -20973.067953 H, E_{AE} = -48532.755009 H (PBE0)

104
UNa5Fe13H42O43
Na -3.3841114 -3.4098373 -0.7015322
Na 3.4047156 -3.3753910 -0.6949898
Na -0.0034110 0.0085580 -5.5129915
Fe -1.8552312 -1.5297510 1.5649899
Fe 0.0148827 -3.4141613 -1.1200216
Fe -1.4747879 -1.9207869 -3.1521119
Fe -3.3592017 -0.0243082 -0.4077204
Fe 1.4836960 -1.9070813 -3.1542040
Fe -1.8732396 1.5064128 1.5620763
Fe -0.0005716 -0.0107795 -0.8065975
O 0.0040564 -1.5097167 -1.8164097
O -1.4739641 -0.0141809 0.2847158

O	-1.8635104	-0.0241912	-3.9794874
O	-3.1808865	1.3537550	-1.9242104
O	-3.2019662	-1.4148352	-1.9095896
O	-1.4452299	-3.6100059	-2.2979246
O	-1.3671505	-3.1917091	0.5162746
O	0.0004531	-1.9059454	2.6020171
O	1.4724116	-3.5993657	-2.2953701
O	-1.7624546	-0.0111421	2.8405591
O	0.0070984	-2.1325818	-4.3131248
O	-3.5585347	-1.4378315	0.9026055
O	-3.5731620	1.3927490	0.8954711
Na	3.3252892	3.4092758	-0.8114899
Na	-3.3517990	3.3756277	-0.7933094
Fe	1.8586364	1.5449318	1.5358665
Fe	-0.0169490	3.3801060	-1.1674656
Fe	1.4565538	1.8749753	-3.1671662
Fe	3.3587431	0.0104375	-0.4141144
Fe	-1.4784512	1.8618779	-3.1641427
Fe	1.8762203	-1.4886595	1.5839854
O	-0.0078183	1.4797350	-1.8334643
O	1.4751169	0.0074339	0.2815146
O	1.8559232	-0.0125425	-3.9825790
O	3.2097506	-1.3863348	-1.9060290
O	3.1634427	1.3820716	-1.9394141
O	-1.3831699	3.1444569	0.4648574
O	1.4365266	3.5798579	-2.3481990
O	1.3581851	3.1861568	0.4548410
O	0.0055775	1.9370682	2.5665226
O	-1.4737158	3.5684164	-2.3399955
O	1.7679710	0.0491249	2.8378297
O	-0.0149764	2.0828284	-4.3363530
O	3.5601284	1.4458392	0.8694458
O	3.5759548	-1.3840805	0.9150711
O	1.3935191	-3.1466729	0.5193789
H	1.2094699	-3.8622586	1.1449054
H	0.0304524	-2.7917172	2.9917640
H	3.9270617	-1.2309079	-2.5376453
H	2.7407271	-0.0082947	-4.3752122
H	-3.9192119	-1.2517087	-2.5392785
H	-2.7480655	-0.0312659	-4.3726163
H	-0.0236494	2.8276796	2.9450864
H	1.1760046	3.9121623	1.0688449
H	-1.2045185	3.8708661	1.0794172
H	-3.8875963	1.1661809	-2.5588234
H	-1.1829934	-3.9099263	1.1388815
H	3.8692109	1.1898968	-2.5736108
H	4.8654961	2.5898274	0.7895175
O	5.3171327	3.3563649	0.3692512
H	5.4990511	3.9671863	1.0935164
H	-0.1073442	-2.2469082	-6.0079863
O	-0.2258776	-1.8565737	-6.9074531
H	0.5332636	-2.1672932	-7.4152496
O	-5.3649250	-3.3285401	0.4966389
H	-4.9009139	-2.5580014	0.8938312
H	-5.5472686	-3.9195629	1.2370473

H -2.6920827 4.7525599 -2.5268934
O -3.5855955 5.1221145 -2.3277944
H -3.4251151 6.0368444 -2.0665902
H 4.8122065 -2.6075093 1.0400523
O 5.1626322 -3.4966525 0.8075118
H 6.0772240 -3.3442312 0.5409367
O -3.3254151 -5.3905623 -1.9164377
H -2.5534098 -4.8926639 -2.2814851
H -3.9105827 -5.5330683 -2.6700063
O -5.0991507 3.5422178 0.7160215
H -6.0150786 3.3993575 0.4487153
H -4.7643863 2.6518022 0.9685184
O 0.2126556 1.8598597 -6.9393858
H -0.5495928 2.1681335 -7.4441294
H 0.0944376 2.2462973 -6.0392966
H 2.5205300 4.8994828 -2.3821981
O 3.5986597 -5.1245185 -2.2132378
H 2.7065606 -4.7551959 -2.4248369
H 3.4357266 -6.0389358 -1.9527251
O 3.2880497 5.4066460 -2.0237380
H 3.8761649 5.5355038 -2.7775305
U 0.0016379 0.0291600 4.1166603
H 2.3034630 2.0393619 4.8285333
O 1.4311657 1.7642412 5.1414758
H 1.4614515 1.8255241 6.1069320
O -1.5332912 1.7659655 5.0529003
H -1.3163875 2.5867237 5.5161618
H -2.4970930 1.6894403 5.0789051
H 2.5187424 -1.5761526 5.0920983
O 1.5554278 -1.6613977 5.0973311
H 1.3588956 -2.4669821 5.5952224
O -1.4130286 -1.6937040 5.1774422
H -2.2712360 -2.0127859 4.8674977
H -1.4497365 -1.7192270 6.1443447
O -0.0088655 0.0578548 6.6689278
H 0.5999941 -0.4129533 7.2540116
H -0.6104447 0.5569185 7.2380129

Np-Fe_{13} - $[\{Np(H_2O)_5\}\{Na(H_2O)_2\}_5Fe_{13}O_{28}H_{12}]^{4+}$ E_{ECP} = -21010.551418 H, E_{AE} = -49318.421189 H (PBE0)

104
NpNa5Fe13H42O43
Na -3.3847094 -3.4109755 -0.6972064
Na 3.4042851 -3.3781848 -0.6947707
Na -0.0075335 0.0184864 -5.5003913
Fe -1.8809897 -1.5287548 1.5692792
Fe 0.0135933 -3.4082814 -1.0973094
Fe -1.4764578 -1.9204828 -3.1363679
Fe -3.3706720 -0.0239900 -0.4131487
Fe 1.4794316 -1.9085845 -3.1415931
Fe -1.8973989 1.4909590 1.5770021
Fe -0.0026064 -0.0103868 -0.7848367
O 0.0026312 -1.5066660 -1.7995964

O	-1.4872074	-0.0174143	0.2920596
O	-1.8541691	-0.0175058	-3.9619469
O	-3.1859991	1.3611013	-1.9198415
O	-3.2124524	-1.4144782	-1.9151140
O	-1.4471834	-3.6059827	-2.2728103
O	-1.3695800	-3.1913672	0.5402900
O	0.0070959	-1.8690179	2.5978829
O	1.4678492	-3.5978103	-2.2746105
O	-1.8069159	-0.0276865	2.8566632
O	0.0025141	-2.1378353	-4.2988509
O	-3.5777202	-1.4414294	0.8890075
O	-3.5908129	1.3881123	0.8953431
Na	3.3324936	3.4042900	-0.7966884
Na	-3.3470148	3.3792364	-0.7705557
Fe	1.8814070	1.5299720	1.5442733
Fe	-0.0112068	3.3738085	-1.1395349
Fe	1.4575190	1.8781046	-3.1525656
Fe	3.3696435	0.0100217	-0.4270363
Fe	-1.4747346	1.8682479	-3.1452745
Fe	1.9069660	-1.4863755	1.5845552
O	-0.0036788	1.4766658	-1.8172262
O	1.4876521	0.0039661	0.2850991
O	1.8439658	-0.0095212	-3.9712922
O	3.2142580	-1.3895672	-1.9166040
O	3.1779095	1.3844732	-1.9462697
O	-1.3812952	3.1266070	0.4907409
O	1.4408449	3.5770625	-2.3205126
O	1.3672769	3.1812746	0.4834509
O	0.0091907	1.8840841	2.5554867
O	-1.4674773	3.5701921	-2.3097472
O	1.8148563	0.0359560	2.8520375
O	-0.0143814	2.0967993	-4.3192215
O	3.5759875	1.4410757	0.8599984
O	3.5982500	-1.3870193	0.8938958
O	1.3979877	-3.1447318	0.5385180
H	1.2168429	-3.8604679	1.1647565
H	0.0330455	-2.7504069	2.9986934
H	3.9293418	-1.2368926	-2.5513945
H	2.7304275	-0.0068206	-4.3600123
H	-3.9280126	-1.2522708	-2.5469569
H	-2.7426361	-0.0234080	-4.3460512
H	-0.0283046	2.7564031	2.9748440
H	1.1937519	3.9089699	1.0979339
H	-1.1785085	3.8403074	1.1121228
H	-3.8959909	1.1876420	-2.5548869
H	-1.1911516	-3.9121681	1.1616739
H	3.8831895	1.1990896	-2.5830662
H	4.8858321	2.5870615	0.7910008
O	5.3323938	3.3575279	0.3732818
H	5.5149018	3.9650788	1.1001527
H	-0.1088186	-2.2396379	-5.9934350
O	-0.2247638	-1.8466316	-6.8922284
H	0.5350877	-2.1568536	-7.3992827
O	-5.3763819	-3.3414808	0.4868232
H	-4.9203451	-2.5657750	0.8825837

H	-5.5571404	-3.9304926	1.2292592
H	-2.6846186	4.7543107	-2.5035393
O	-3.5796045	5.1231111	-2.3098593
H	-3.4212039	6.0375886	-2.0464905
H	4.8353067	-2.6144813	1.0227335
O	5.1755615	-3.5080891	0.7935056
H	6.0892588	-3.3658404	0.5183155
O	-3.3245356	-5.3949247	-1.9101121
H	-2.5505824	-4.8968352	-2.2699475
H	-3.9065618	-5.5336384	-2.6668237
O	-5.1093768	3.5442922	0.7228197
H	-6.0242009	3.4109832	0.4469804
H	-4.7834318	2.6493889	0.9697156
O	0.2018234	1.8724768	-6.9237978
H	-0.5624882	2.1809434	-7.4252981
H	0.0876900	2.2589232	-6.0231735
H	2.5244460	4.8995511	-2.3606883
O	3.5917579	-5.1277264	-2.2147439
H	2.6979937	-4.7587705	-2.4189380
H	3.4310418	-6.0417337	-1.9514071
O	3.2946865	5.4045293	-2.0054356
H	3.8799355	5.5318910	-2.7617208
Np	0.0028468	0.0120927	4.0851092
H	2.3497471	1.9116764	4.7615009
O	1.4311927	1.7836070	5.0361619
H	1.4304202	1.8687237	6.0006776
O	-1.5523234	1.7691212	4.9184621
H	-1.3861963	2.5254764	5.4980739
H	-2.5089192	1.6239067	4.9332747
H	2.4895153	-1.5507301	5.0348496
O	1.5274921	-1.6145972	5.1143027
H	1.3480201	-2.3921481	5.6611556
O	-1.3542938	-1.7296532	5.1609656
H	-2.2268923	-2.0214460	4.8635769
H	-1.3903461	-1.7275179	6.1284599
O	-0.0371255	0.1339266	6.6117453
H	0.6053757	-0.2777614	7.2058250
H	-0.6843847	0.5806306	7.1740616

Pu-Fe_{13} - $[\{Pu(H_2O)_5\}\{Na(H_2O)_2\}_5Fe_{13}O_{28}H_{12}]^{4+}$ E_{ECP} = -21050.00420 H, E_{AE} = -50118.986237 H (PBE0)

104
PuNa5Fe13H42O43

Na	4.666501439	-0.001577305	-0.004122424
Na	0.065905752	4.646452281	0.064412986
Na	-0.013388248	0.024671624	-4.575512565
Fe	2.430935276	-0.300372375	2.316134258
Fe	2.413716260	2.371680859	-0.329568302
Fe	2.374431280	0.306239124	-2.363718367
Fe	2.382208066	-2.393863746	0.308396410
Fe	0.325992640	2.382980832	-2.337471842
Fe	0.301522256	-2.421932935	2.326958089
Fe	0.000000000	0.000000000	0.000000000
O	1.063485935	1.045935695	-1.017906777

O	1.066208948	-1.060848141	1.042552685
O	1.289028692	-1.294754163	-3.185607954
O	1.277800457	-3.243342898	-1.215863889
O	3.241522189	-1.310435352	-1.219873004
O	3.586363776	1.464075081	-1.481672624
O	3.235376267	1.267174378	1.314790413
O	1.348956972	1.281539466	3.376836497
O	1.507000122	3.573069866	-1.448033778
O	1.313579491	-1.305729694	3.599548379
O	1.482359040	1.493657753	-3.531750062
O	3.567365479	-1.521219101	1.561402361
O	1.508203275	-3.570080886	1.565110472
Na	-4.666466981	-0.026281439	0.027684998
Na	-0.043188324	-4.652800957	0.006456388
Fe	-2.419664750	0.256694357	2.335333525
Fe	-2.399741305	-2.387561477	-0.340502092
Fe	-2.380513392	-0.295545982	-2.348567193
Fe	-2.373611800	2.375797042	0.358087911
Fe	-0.326299788	-2.366274368	-2.364724965
Fe	-0.280404266	2.395113994	2.363418830
O	-1.058585097	-1.048188064	-1.020941550
O	-1.051303362	1.039636527	1.085944980
O	-1.300892040	1.322819724	-3.148561099
O	-1.270726569	3.239364219	-1.147447752
O	-3.239992179	1.307996990	-1.169170163
O	-1.270913198	-3.191985799	1.306678410
O	-3.583903668	-1.464346634	-1.469016101
O	-3.223836526	-1.291532231	1.325903232
O	-1.292088867	-1.307790050	3.335263541
O	-1.501092754	-3.569208127	-1.486730614
O	-1.304676862	1.264173182	3.659727280
O	-1.495974768	-1.461366283	-3.538673741
O	-3.553863178	1.489754701	1.607605827
O	-1.496533173	3.540962292	1.635524805
O	1.275841932	3.155254853	1.341671205
H	1.953541895	3.418382363	1.978566018
H	1.957415551	1.916791161	3.781076672
H	-1.888584564	3.625631167	-1.783813308
H	-1.937885916	1.968681472	-3.482975189
H	3.587859542	-1.936005534	-1.870989961
H	1.921168876	-1.934915760	-3.539559141
H	-1.870583707	-1.934268600	3.794840807
H	-3.558320261	-1.943189205	1.957223467
H	-1.918878417	-3.520732657	1.944471926
H	1.897467130	-3.607381053	-1.863160257
H	3.659301402	1.877653661	1.934294744
H	-3.596328002	1.939137129	-1.809727314
Pu	0.000000000	0.000000000	4.828496210
O	2.225424559	-0.166922195	5.849440348
H	2.782651848	-0.870592181	5.487247327
H	2.399563296	-0.145091508	6.800596184
H	0.637767258	-2.891525258	5.682392264
O	-0.137012510	-2.313477488	5.700042990
H	-0.778097096	-2.733057551	6.289750806
H	1.101813514	2.568308425	6.373613785

O	0.344712210	2.215673722	5.885214749
H	-0.317364339	2.921423390	5.875315768
O	-2.251511459	-0.327003670	5.787024451
H	-2.970767062	0.274649832	5.548494015
H	-2.215019745	-0.317212009	6.756100286
O	-0.119860819	-0.063639740	7.341599080
H	-0.072272567	0.732888806	7.887908509
H	0.126059267	-0.805386873	7.911005589

Fe_{13} - $[\{Na(H_2O)_2\}_6Fe_{13}O_{28}H_{12}]^+$ E_{ECP} = -20429.461081 H, E_{AE} = -20523.919177 H (PBE0)

95
Na6Fe13H36O40

Na	-0.0006833	0.0052355	4.6975815
Na	0.0011793	-4.7028978	0.0034663
Na	4.7015965	-0.0007253	-0.0019053
Na	0.0001915	-0.0108544	-4.7047707
Fe	-0.3101178	-2.3727278	2.3750710
Fe	2.3759235	-2.3764949	-0.3106171
Fe	0.3128983	-2.3806093	-2.3739161
Fe	-2.3735817	-2.3761687	0.3124146
Fe	2.3753440	-0.3168945	-2.3773010
Fe	-2.3737145	-0.3095630	2.3718724
Fe	0.0008076	-0.0026053	-0.0033995
O	1.0514323	-1.0539725	-1.0521016
O	-1.0494498	-1.0507724	1.0489591
O	-1.2747436	-1.2833995	-3.2397525
O	-3.2385799	-1.2807451	-1.2755498
O	-1.2727665	-3.2414775	-1.2727203
O	1.5203362	-3.5640951	-1.5174328
O	1.2753585	-3.2378462	1.2770599
O	1.2764008	-1.2712706	3.2377178
O	3.5609343	-1.5234895	-1.5188927
O	-1.5183428	-1.5143734	3.5588554
O	1.5211045	-1.5260859	-3.5607049
O	-1.5174017	-3.5595779	1.5226569
O	-3.5588745	-1.5196489	1.5182226
Na	-0.0010228	4.6984196	-0.0124260
Na	-4.7008200	-0.0028098	-0.0020454
Fe	0.3105089	2.3758070	2.3671338
Fe	-2.3739733	2.3710953	-0.3196477
Fe	-0.3107790	2.3666761	-2.3821857
Fe	2.3740348	2.3735008	0.3048472
Fe	-2.3742182	0.3035246	-2.3785864
Fe	2.3745265	0.3135366	2.3711754
O	-1.0500311	1.0450186	-1.0556527
O	1.0502369	1.0498131	1.0455332
O	1.2762081	1.2671069	-3.2448018
O	3.2382134	1.2712850	-1.2790967
O	1.2745953	3.2329983	-1.2838597
O	-3.2372091	1.2749128	1.2681522
O	-1.5177297	3.5539379	-1.5303585
O	-1.2748522	3.2381550	1.2656967
O	-1.2760599	1.2782852	3.2330441
O	-3.5592438	1.5141733	-1.5250132

O	1.5190518	1.5223775	3.5541515
O	-1.5193819	1.5083077	-3.5657137
O	1.5172015	3.5604877	1.5107491
O	3.5594989	1.5210914	1.5134184
O	3.2386028	-1.2748667	1.2736699
H	3.5722543	-1.9127384	1.9188096
H	1.9142660	-1.9140603	3.5758983
H	3.5745877	1.9084283	-1.9235591
H	1.9136713	1.9095604	-3.5843944
H	-1.9176053	-3.5734523	-1.9116803
H	-1.9127034	-1.9275515	-3.5751796
H	-1.9145606	1.9224548	3.5673525
H	-1.9182312	3.5760322	1.9030881
H	-3.5710783	1.9148138	1.9111446
H	-3.5773054	-1.9196313	-1.9170330
H	1.9186966	-3.5736044	1.9156283
H	1.9186345	3.5630416	-1.9246749
O	1.2234235	1.4243258	6.1325934
H	1.5272156	1.6163536	5.2064634
H	1.9946131	1.0459893	6.5715330
H	1.5873429	5.2138372	1.5371553
O	1.3805703	6.1405280	1.2449675
H	0.9894972	6.5619256	2.0195775
H	1.5297040	-1.6203327	-5.2127540
O	1.2272957	-1.4280557	-6.1393235
H	1.9995696	-1.0505353	-6.5770537
O	-1.4206400	-6.1370883	1.2250649
H	-1.6118345	-5.2108170	1.5292273
H	-1.0459713	-6.5776661	1.9970944
H	-5.2111437	1.5229266	-1.6152065
O	-6.1373576	1.2207913	-1.4206169
H	-6.5747104	1.9939640	-1.0444918
H	5.2121523	1.5264647	1.6098132
O	6.1375783	1.2194723	1.4198173
H	6.5812939	1.9900634	1.0458686
O	1.4175677	-6.1414852	-1.2190144
H	1.6118853	-5.2159295	-1.5231984
H	1.0424245	-6.5809167	-1.9914855
O	-1.2210144	-1.4134781	6.1363197
H	-1.5253932	-1.6072966	5.2107595
H	-1.9943887	-1.0421312	6.5773585
O	-6.1367338	-1.2257644	1.4174533
H	-6.5761416	-1.9992330	1.0443310
H	-5.2107192	-1.5288267	1.6112629
O	-1.2215094	1.4062787	-6.1432138
H	-1.9945628	1.0343565	-6.5843311
H	-1.5262830	1.6001038	-5.2177985
H	-1.5905215	5.2084439	-1.5630591
O	6.1374492	-1.2117390	-1.4302398
H	5.2127125	-1.5219117	-1.6187690
H	6.5850781	-1.9817325	-1.0597841
O	-1.3857365	6.1357748	-1.2719640
H	-0.9916233	6.5551519	-2.0461372