SUPPLEMENTARY INFORMATION

Structural Characteristics, Population Analysis, and Binding Energies of $[An(NO_3)]^{2+}$ [with An = Ac - Lr]

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The Supporting Information contains interatomic distances and angles from structural analysis, partial charges and orbital occupancies from population analysis, and thermochemical data from Gibbs free energy calculations.

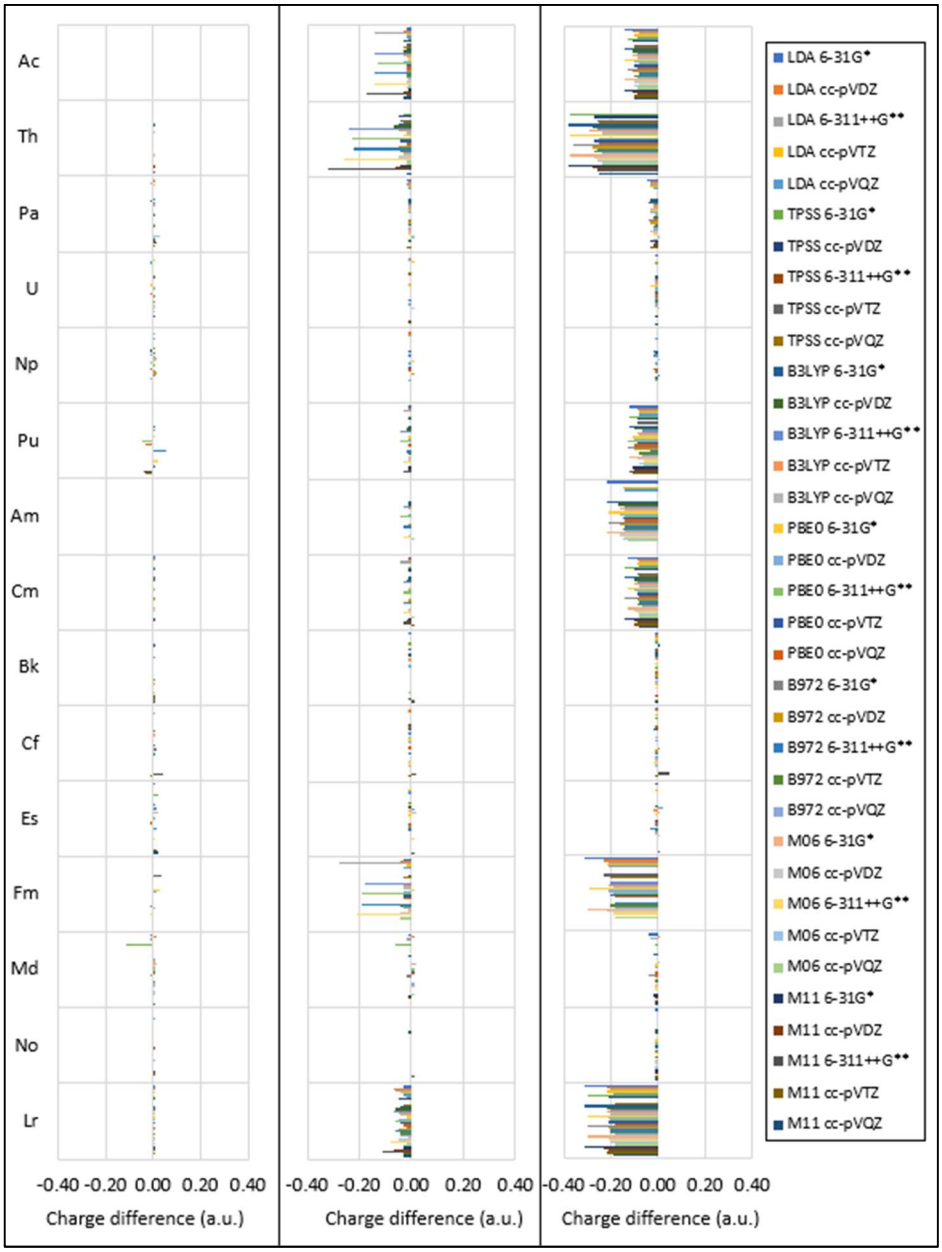

Figure S.1: Difference between charge of An in $[An(NO_3)]^{2+}$ including (ad) and excluding (nd) the most diffuse basis functions in the Stuttgart 1997 basis set calculated with NBO (left), Mulliken (middle), and Löwdin (right), as indicated in Eq. 8.a. [Difference equal = charge calculated with ad - charge calculated with nd].

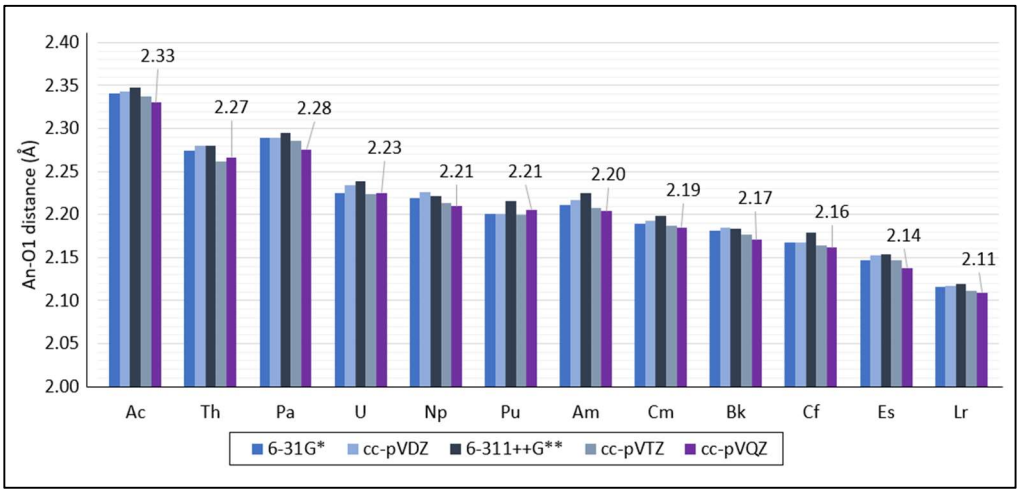

Figure S.2: Calculated An-O1 distance in $[An(NO_3)]^{+2}$ with the PBE0 functional, the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ basis set for N and O atoms, and the Stuttgart RSC 1997 ECP and basis set including all diffuse basis functions in the basis set (in Å). Fm, Md, and No are shown in Figure S.2. [Lines are included as a visual aid and do not represent function continuity].

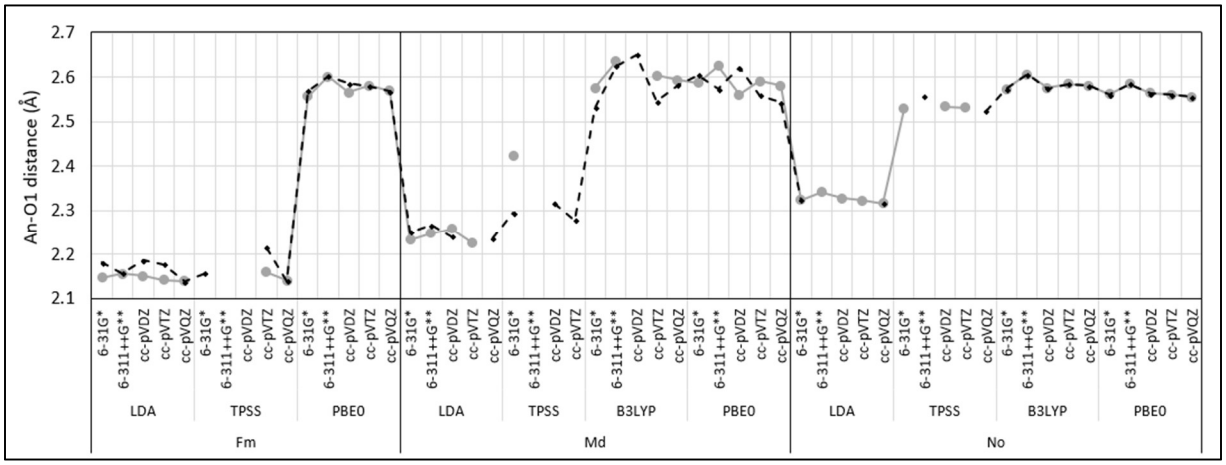

Figure S.3: Calculated An-O1 distance in $[An(NO_3)]^{+2}$ for Fm, Md, and No with LDA, TPSS, B3LYP, and PBE0 functionals, the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ basis set for N and O atoms, and the Stuttgart RSC 1997 ECP and basis set including all diffuse basis functions in the basis set (in Å). [Lines are included as a visual aid and do not represent function continuity].

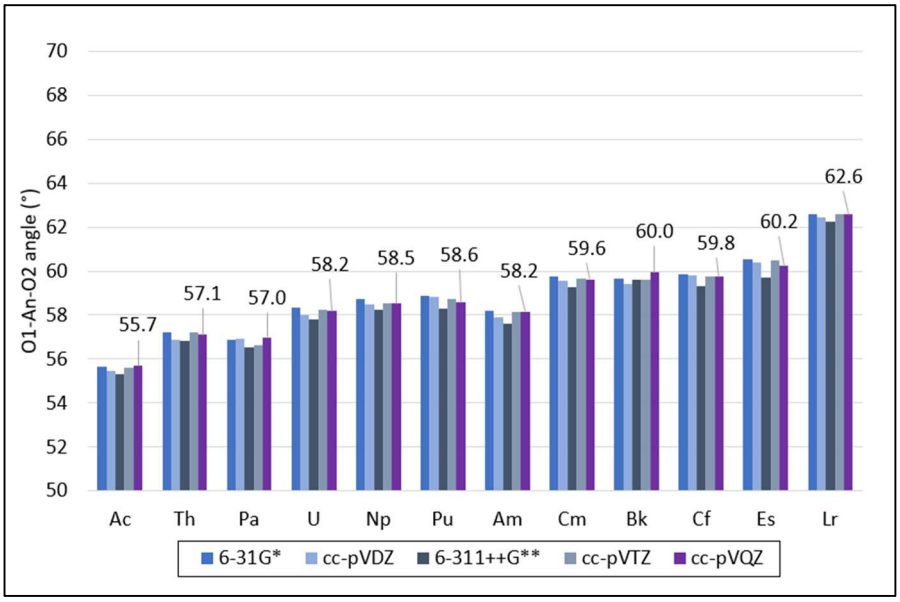

Figure S.4: Calculated O1-An-O2 angle in $[An(NO_3)]^{+2}$ with the PBE0 functional, the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ basis set for N and O atoms, and the Stuttgart RSC 1997 ECP and basis set including all diffuse basis functions in the basis set (in °). Fm, Md, and No are shown in Figure S.4. [Lines are included as a visual aid and do not represent function continuity].

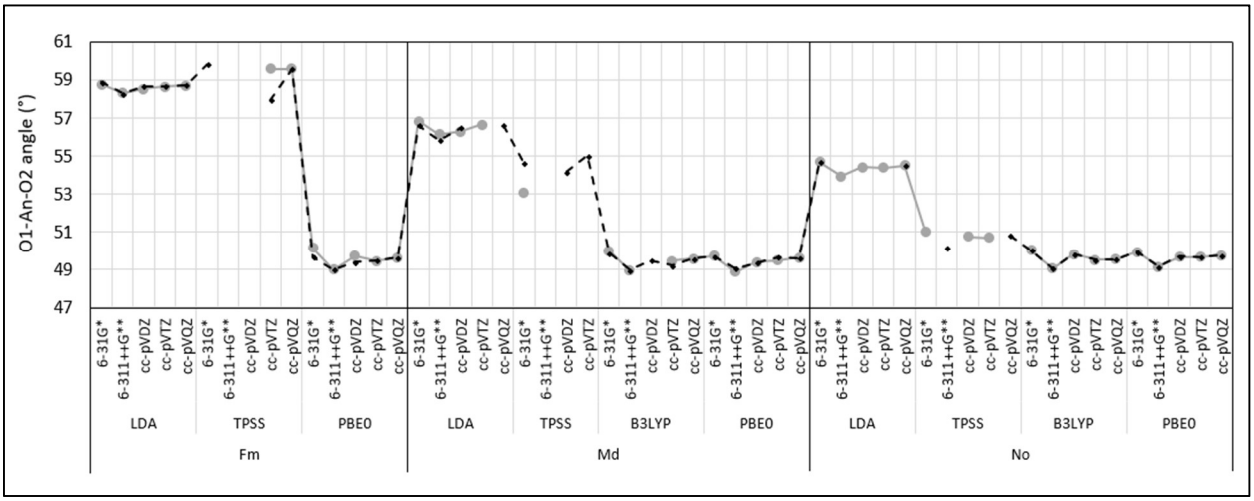

Figure S.5: Calculated O1-An-O2 angle in $[An(NO_3)]^{+2}$ for Fm, Md, and No with LDA, TPSS, B3LYP, and PBE0 functionals, the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ basis set for N and O atoms, and the Stuttgart RSC 1997 ECP and basis set including all diffuse basis functions in the basis set (in °). [Lines are included as a visual aid and do not represent function continuity].

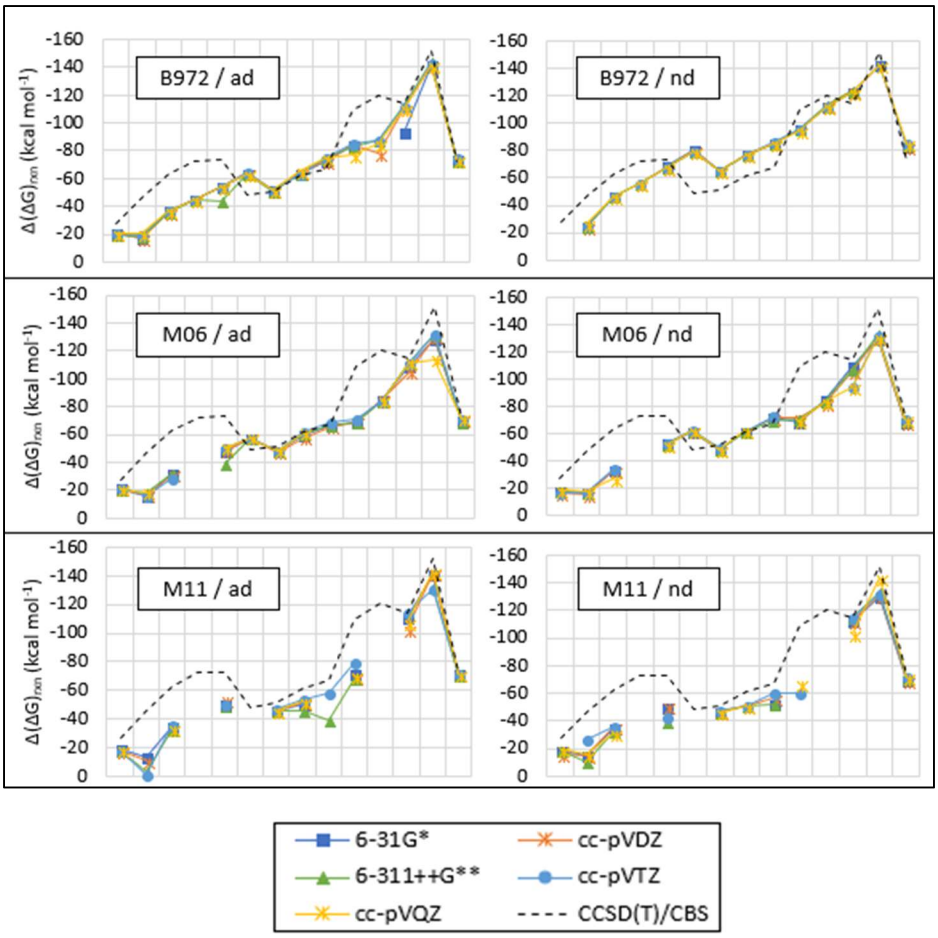

Figure S.6: $\Delta(\Delta G)_{rxn}$ for $[An(NO_3)]^{2+}$ normalized to Ac calculated with B972, M06, and M11 with the Stuttgart RSC 1997 ECP and basis set including (ad) and excluding (nd) the most diffuse basis functions for An, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O. $\Delta(\Delta G)_{rxn}$ calculated with CCSD(T)-FC1, the cc-pV∞Z-X2C basis set for An, and the cc-pV∞Z-DK basis set for N and O is included as reference (indicated as CCSD(T)/CBS in labels). Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn}$ = $\Delta G_{rxn,An}$ - $\Delta G_{rxn,Ac}$, as indicated in Eq. 5.a; with An = Th - Lr]. The lines are included as a visual aid, and do not represent function continuity.

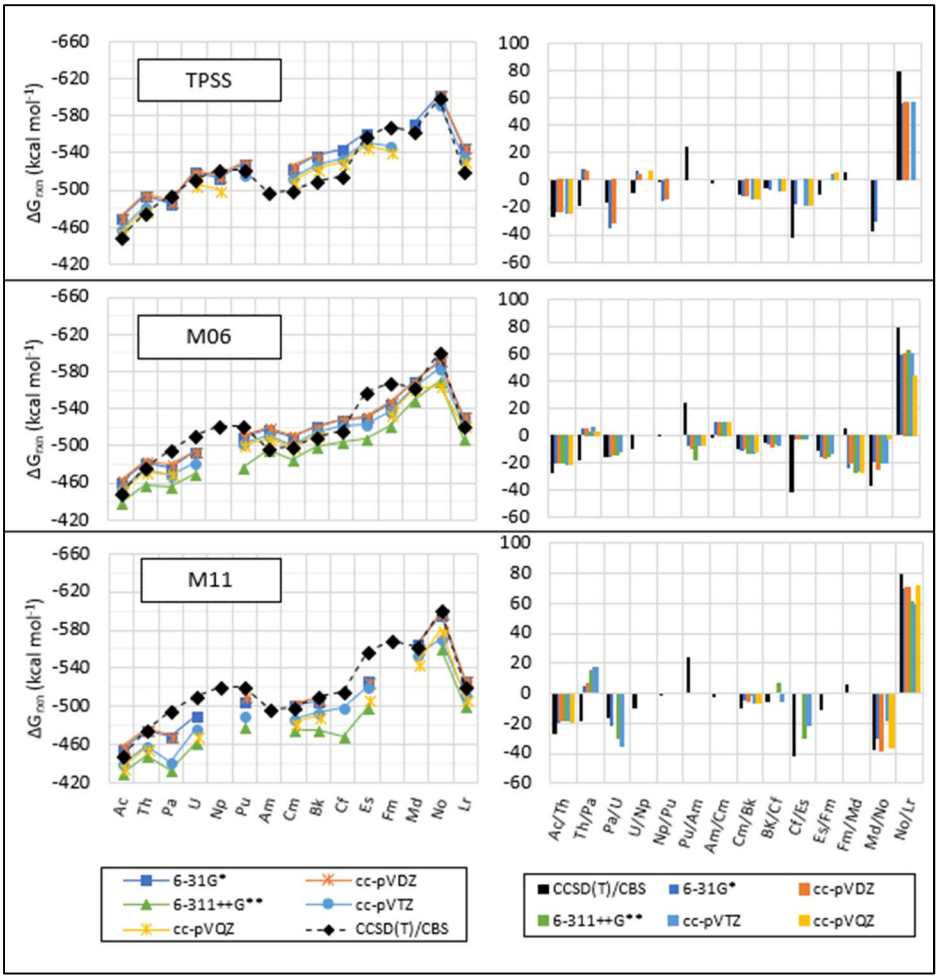

Figure S.7: Calculated ΔG_{rxn} (left) and difference between contiguous actinides (right) for $[An(NO_3)]^{2+}$ [with An = Ac to Lr] with B972, M06, and M11, the Stuttgart RSC 1997 ECP and associated basis set including all diffuse functions for An, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O; and CCSD(T)-FC1/cc-pV∞Z-X2C:cc-pV∞Z-DK (indicated as CCSD(T)/CBS in labels). Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn\ An1/An2} = \Delta G_{rxn,An2} - \Delta G_{rxn,An1}$; with An = Ac - Lr].

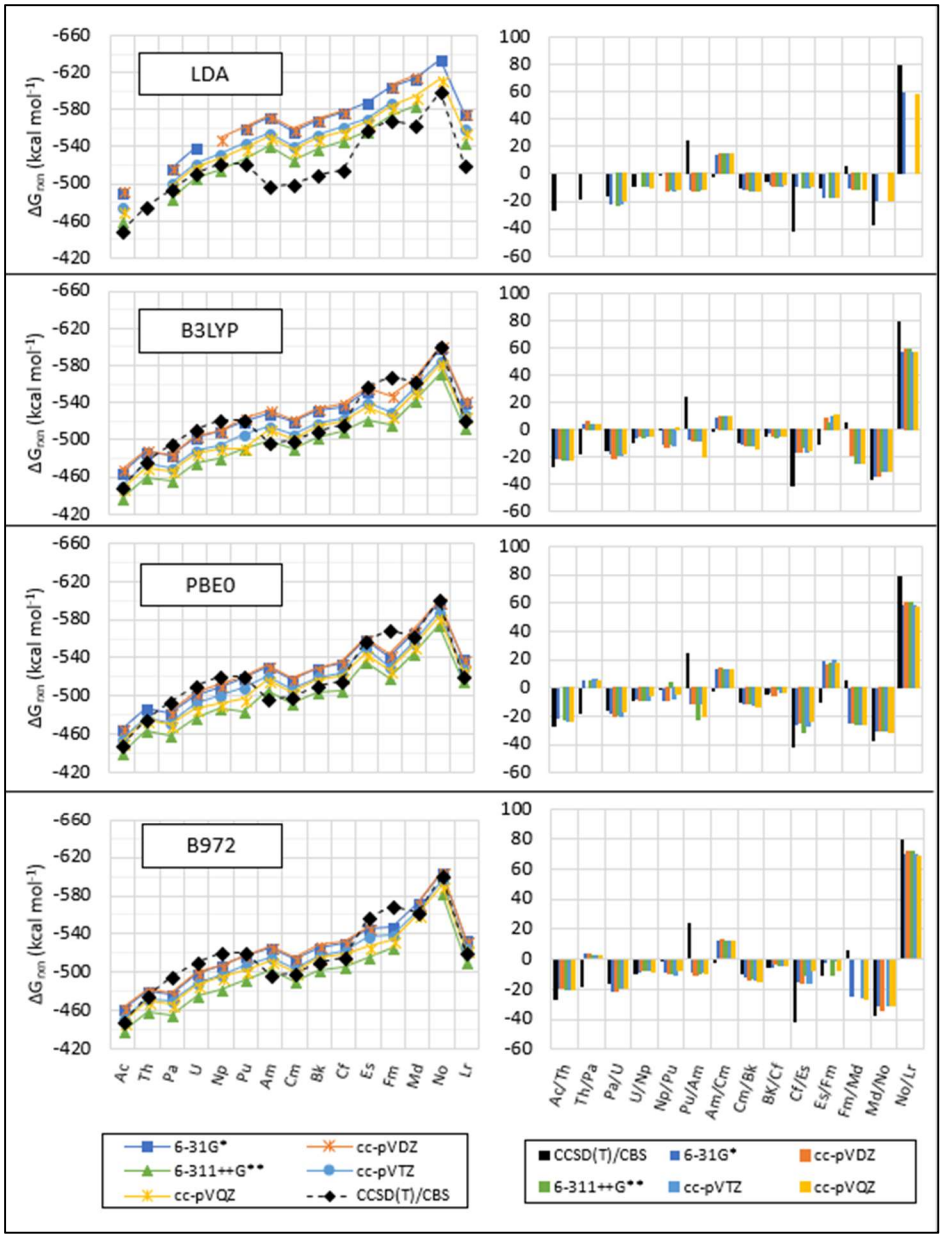

Figure S.8: Calculated ΔG_{rxn} (left) and difference between contiguous actinides (right) for $[An(NO_3)]^{2+}$ [with An = Ac to Lr] with LDA, B3LYP, PBE0, and B972, the Stuttgart RSC 1997 ECP and associated basis set excluding the most diffuse functions in the basis set for An, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O; and CCSD(T)-FC1/cc-pV∞Z-X2C:cc-pV∞Z-DK (indicated as CCSD(T)/CBS in labels). Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn\ An1/An2} = \Delta G_{rxn,An2} - \Delta G_{rxn,An1}$; with An = Ac - Lr].

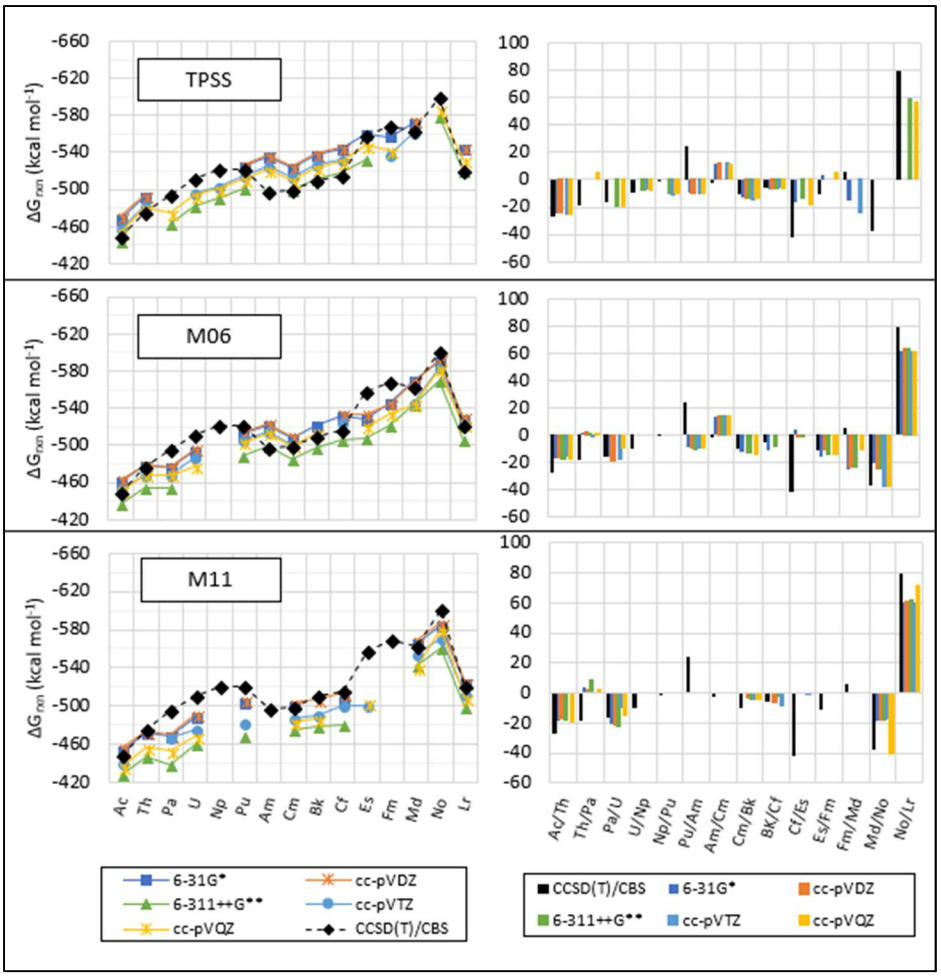

Figure S.9: Calculated ΔG_{rxn} (left) and difference between contiguous actinides (right) for $[An(NO_3)]^{2+}$ [with An = Ac to Lr] with TPSS, M06, and M11, the Stuttgart RSC 1997 ECP and associated basis set excluding most diffuse functions in the basis set for An, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O; and CCSD(T)-FC1/cc-pV∞Z-X2C:cc-pV∞Z-DK (indicated as CCSD(T)/CBS in labels). Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn\ An1/An2} = \Delta G_{rxn,An2} - \Delta G_{rxn,An1}$; with An = Ac - Lr].

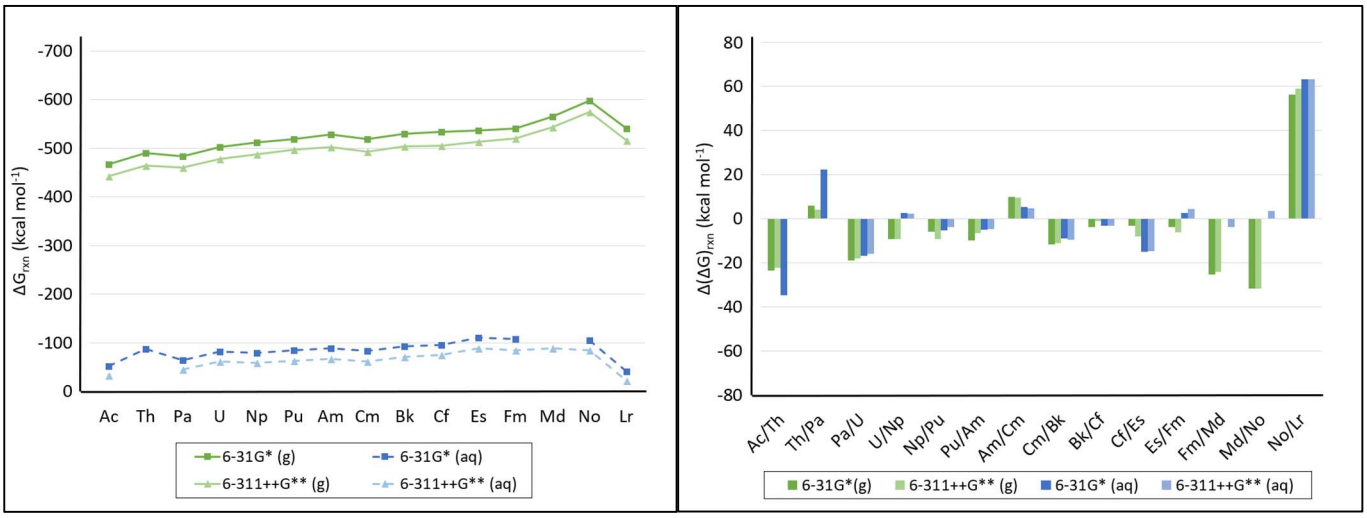

Figure S.10: ΔG_{rxn} for $[An(NO_3)]^{2+}$ calculated with the PBE0 functional, the Stuttgart RSC 1997 ECP including all diffuse basis functions in the An basis set, and the 6-31G* and 6-311++G** basis set for N and O (right), with differences between contiguous actinides (left). Values reported in kcal mol^{-1}. [Lines are included as a visual aid and do not represent function continuity.]

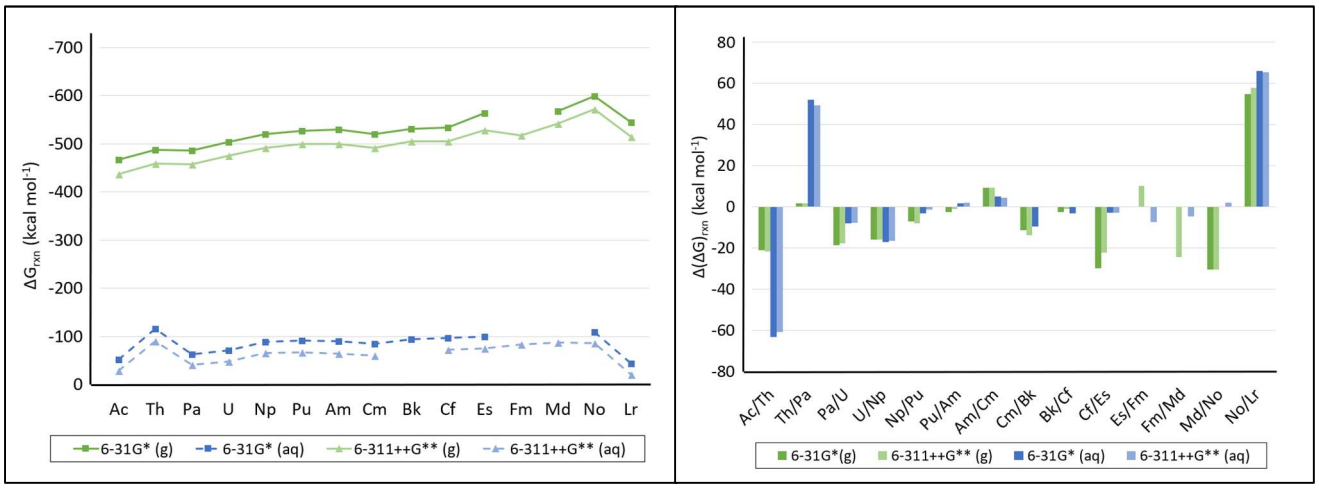

Figure S.11: ΔG_{rxn} for $[An(NO_3)]^{2+}$ calculated with the B3LYP functional, the Stuttgart RSC 1997 ECP including all diffuse basis functions in the An basis set, and the 6-31G* and 6-311++G** basis set for N and O (right), with differences between contiguous actinides (left). Values reported in kcal mol^{-1}. [Lines are included as a visual aid and do not represent function continuity.]

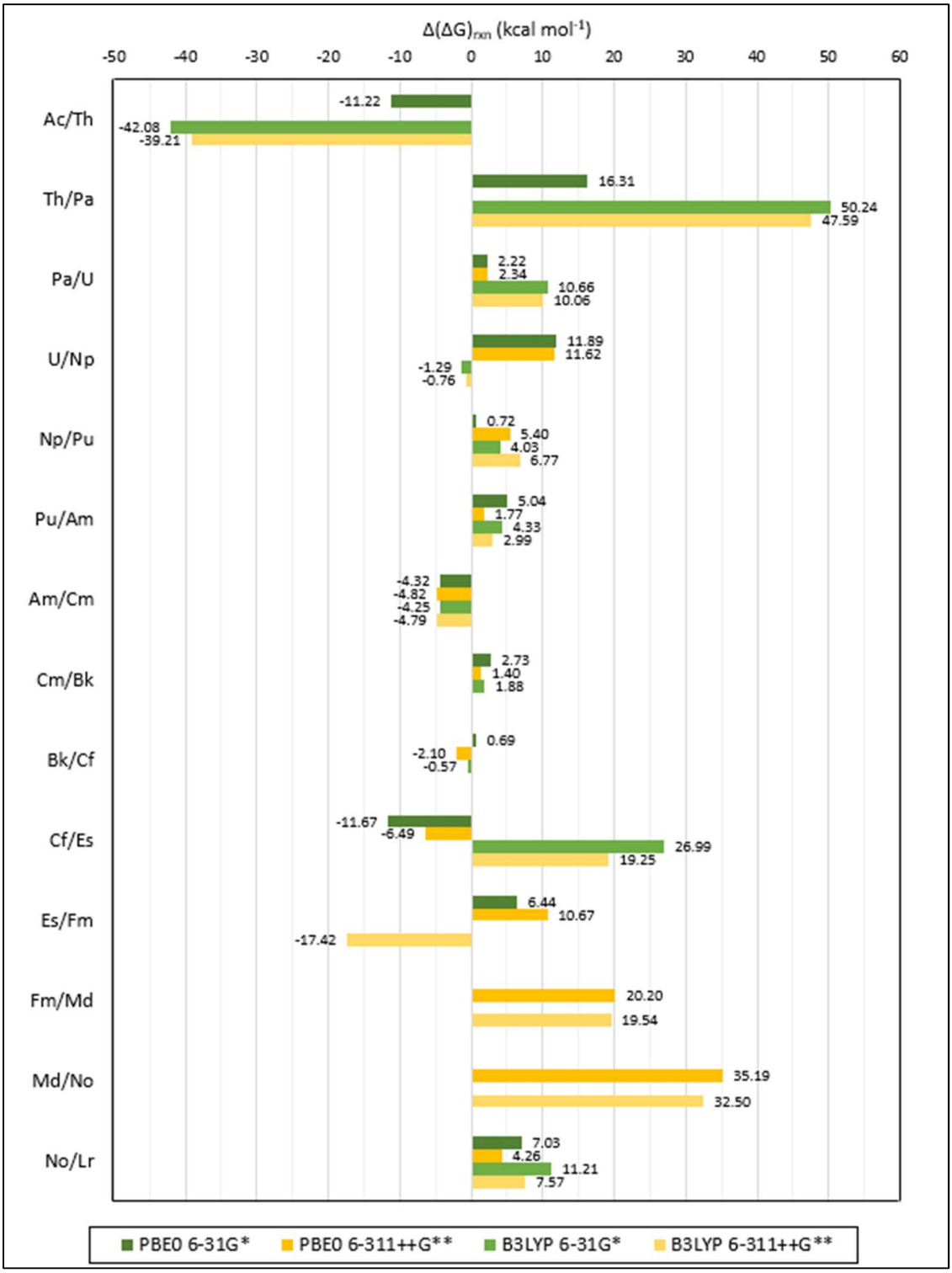

Figure S.12: Difference in predicted ΔG_{rxn} for $[An(NO_3)]^{2+}$ amongst contiguous actinides in gas and aqueous phase calculated with the B3LYP and PBE0 functional, the Stuttgart RSC 1997 ECP including all diffuse basis functions in the An basis set, and the 6-31G* and 6-311++G** basis set for N and O. Values reported in kcal mol^{-1}. [Calculated as Δ(ΔG)rxn between contiguous actinides (aq) - Δ(ΔG)rxn between contiguous actinides (g).]

Table S.1: Differences in An partial charge predicted with NBO including (ad) and excluding (nd) the most diffuse basis functions in the bob-An basis sets. [Calculated as ad - nd].

Functional	Non-An Basis Set	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr
LDA	6-31G*	0.00	-	-0.01	0.00	-	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	0.00	0.00
	cc-pVDZ	0.00	-	0.01	-	0.00	0.00	-	0.00	0.00	0.00	-	0.00	0.01	-	0.00
	6-311++G**	-0.01	-	-0.02	0.00	0.00	0.00	-	0.00	0.00	0.00	0.00	0.00	-0.01	-	0.00
	cc-pVTZ	0.00	-	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-	-	0.00
	cc-pVQZ	0.00	0.00	0.00	-0.01	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-	0.00	0.00
TPSS	6-31G*	0.00	0.00	-	-	-	0.00	-	0.00	0.00	0.00	0.02	-	-0.11		0.00
	cc-pVDZ	0.00	0.00	-	-	-	0.00	-	0.00	0.01	-	-	-	-	-	0.00
	6-311++G**	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	cc-pVTZ	0.00	0.00	-	-	-	0.00	-	0.00	-0.01	0.00	-	0.03	-	-	-
	cc-pVQZ	0.00	0.00	-	0.00	0.00	-	-	0.00	0.00	0.00	0.00	0.00	-	-	0.00
B3LYP	6-31G*	0.00	0.00	0.00	0.00	-0.01	0.01	0.00	0.00	0.00	-0.01	0.01	-	-0.01	0.00	0.00
	cc-pVDZ	0.00	0.00	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.01	-0.01	-	-	0.00	0.00
	6-311++G**	0.00	-0.01	0.00	0.00	-0.01	0.00	0.00	0.00	0.00	-0.01	0.01	0.00	0.00	0.00	0.00
	cc-pVTZ	0.00	-0.01	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	-0.01	0.00	0.01	0.00	0.00
	cc-pVQZ	0.00	0.01	-0.01	0.00	0.02	-0.01	0.00	0.00	0.00	0.00	0.02	0.00	0.01	0.00	0.00
PBE0	6-31G*	0.00	0.00	0.00	-0.01	0.01	0.01	0.00	0.00	0.00	0.00	-0.01	0.03	0.00	0.00	0.00
	cc-pVDZ	0.00		0.00	0.00	-0.01	-0.01	0.00	0.00	0.00	0.00	0.01	0.02	0.00	0.00	0.00
	6-311++G**	-0.01	0.00	0.00	0.00	0.00	-0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00
	cc-pVTZ	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00
	cc-pVQZ	0.00	0.00	0.00	-0.01	-0.01	-0.03	0.00	0.00	0.00	0.00	-0.01	0.00	0.00	0.00	0.00
B972	6-31G*	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	-	-0.01	0.00	0.00
	cc-pVDZ	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	-	-0.01	0.00	0.00
	6-311++G**	-0.01	0.00	0.00	0.00	0.01	0.05	0.00	0.00	0.00	0.00	0.02	0.00	-	0.00	0.00
	cc-pVTZ	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	-0.01	0.00	0.00	0.00
	cc-pVQZ	0.00	0.00	0.00	0.00	-0.01	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00
M06	6-31G*	0.00	0.00	0.00	0.00	-	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	cc-pVDZ	0.00	0.00	0.00	0.01	-	-0.01	0.00	0.00	-	0.00	0.00	-0.01	0.00	0.00	0.00
	6-311++G**	-0.01	0.00	0.00	-	-	0.02	0.00	0.00	0.00	0.00	0.00	-0.01	0.00	0.00	0.00
	cc-pVTZ	0.00	0.00	0.03	0.00	-	0.00	0.00	0.00	-	0.00	-	-	0.01	0.00	0.00
	cc-pVQZ	0.00	0.00	0.00	-	-	0.01	0.00	0.00	0.00	-	-	0.00	0.01	0.00	0.00
M11	6-31G*	0.00	0.00	0.01	0.00	-	0.00	-	0.00	-	-	-	-	0.00	0.00	0.00
	cc-pVDZ	0.00	0.01	0.01	-	-	0.00	-	0.00	0.00	-	-	-	0.00	0.00	0.00
	6-311++G**	0.00	0.00	0.00	0.00	-	-0.04	-	0.00	0.00	0.04	-	-	-	0.00	0.00
	cc-pVTZ	0.00	-	0.00	0.00	-	-0.03	-	0.00	0.00	-0.01	0.01	-	0.00	0.00	0.00
	cc-pVQZ	0.00	0.00	-	0.00	-	-	-	0.00	0.00	-	0.02	-	0.00	0.00	0.00

Table S.2: Differences in An partial charge predicted with Mulliken including (ad) and excluding (nd) the most diffuse basis functions in the bob-An basis sets. [Calculated as ad - nd].

Functional	Non-An Basis Set	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr
LDA	6-31G*	-0.02	-	-0.02	0.00		-0.01	0.00	-0.01	-0.01	0.00	0.00	-0.03	-0.01	0.00	-0.03
	cc-pVDZ	-0.03	-	-0.01		-0.01	-0.01	-	-0.01	0.00	-0.01	-	-0.04	0.01	-	-0.07
	6-311++G**	-0.14	-	-0.02	-0.01	-0.01	-0.03	-	-0.04	0.00	0.00	0.00	-0.28	-0.02	-	-0.06
	cc-pVTZ	-0.02	-	-0.01	0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	-0.02	-	-	-0.04
	cc-pVQZ	-0.02	-	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	-0.03	-	0.00	-0.03
TPSS	6-31G*	-0.01	-0.03	-	-	-	-0.01	-	-0.01	-0.01	0.00	0.00	-	-0.06	-	-0.02
	cc-pVDZ	-0.03	-0.05	-	-	-	-0.01	-	-0.01	0.00	-	-	-	-	-	-0.05
	6-311++G**	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	cc-pVTZ	-0.02	-0.04	-	-	-	0.00	-	0.00	-0.01	-0.01	-	-0.01	-	-	-
	cc-pVQZ	-0.03	-0.03	-	-0.01	0.00	-	-	0.00	0.00	-0.01	-0.01	-0.03	-	-	-0.03
B3LYP	6-31G*	-0.02	-0.05	-0.01	0.00	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	0.00	-	-0.01	-0.01	-0.04
	cc-pVDZ	-0.03	-0.07	-0.01	0.00	0.00	-0.02	-0.01	-0.02	-0.01	0.00	-0.01	-	-	-0.01	-0.06
	6-311++G**	-0.14	-0.24	-0.01	0.00	-0.01	-0.04	-0.03	-0.03	0.00	-0.01	0.01	-0.18	0.00	0.00	-0.07
	cc-pVTZ	-0.03	-0.05	-0.01	0.00	0.00	0.00	-0.01	0.00	-0.01	0.00	-0.01	-0.03	0.00	0.00	-0.05
	cc-pVQZ	-0.02	-0.02	-0.01	-0.01	0.00	0.00	0.00	0.00	0.00	-0.01	0.02	-0.03	0.02	0.00	-0.04
PBE0	6-31G*	-0.01	-0.03	-0.01	-0.01	0.01	-0.01	0.00	-0.01	0.00	-0.01	-0.01	0.01	0.00	0.00	-0.02
	cc-pVDZ	-0.03	-	-0.01	0.00	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	0.00	-0.03	0.00	0.00	-0.05
	6-311++G**	-0.13	-0.23	0.00	0.00	0.00	-0.04	-0.04	-0.03	0.00	0.00	0.00	-0.19	0.01	0.00	-0.06
	cc-pVTZ	-0.02	-0.04	0.00	0.00	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.03	0.01	0.00	-0.04
	cc-pVQZ	-0.02	-0.02	0.00	0.00	-0.01	-0.01	0.00	0.00	0.00	-0.01	-0.01	-0.03	0.01	0.00	-0.03
B972	6-31G*	-0.02	-0.02	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	-	-0.02	0.00	-0.02
	cc-pVDZ	-0.02	-0.05	-0.01	0.00	0.01	0.00	-0.01	-0.01	0.00	0.00	-0.01	-	0.00	0.00	-0.05
	6-311++G**	-0.14	-0.22	-0.01	-0.01	0.00	-0.02	-0.03	-0.03	0.00	-0.01	-0.01	-0.19	-	0.00	-0.06
	cc-pVTZ	-0.02	-0.04	0.00	0.00	0.00	-0.01	0.00	0.00	0.00	0.00	0.00	-0.04	0.00	0.00	-0.04
	cc-pVQZ	-0.02	-0.03	-0.01	-0.01	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.03	0.01	0.00	-0.04
M06	6-31G*	-0.01	-0.03	-0.01	0.00	-	-0.01	0.00	-0.01	0.00	0.00	0.00	-0.01	0.01	0.00	-0.01
	cc-pVDZ	-0.02	-0.05	-0.01	0.01	-	-0.01	-0.01	-0.01	-	-0.01	0.00	-0.04	0.00	0.00	-0.05
	6-311++G**	-0.14	-0.26	-0.01	-	-	-0.03	-0.03	-0.03	0.00	-0.01	0.01	-0.21	0.00	0.00	-0.08
	cc-pVTZ	-0.01	-0.02	0.01	0.00	-	0.00	0.01	0.00	-	-0.01	-	-	0.00	0.00	-0.02
	cc-pVQZ	-0.02	-0.02	-0.01	-	-	0.00	0.00	0.01	-0.01	-	-	-0.04	0.01	0.00	-0.03
M11	6-31G*	-0.01	-0.04	-0.01	0.00	-	-0.01	-	-0.01	-	-	-	-	-0.01	0.00	-0.03
	cc-pVDZ	-0.03	-0.06	0.00	-	-	-0.01	-	-0.02	0.00	-	-	-	-0.01	0.00	-0.07
	6-311++G**	-0.17	-0.32	0.00	-0.01	-	-0.03	-	-0.03	-0.01	0.02	-	-	-	0.01	-0.11
	cc-pVTZ	-0.02	-	-0.02	-0.01	-	0.00	-	0.01	0.01	-0.01	0.00	-	0.00	0.00	-0.03
	cc-pVQZ	-0.03	-0.02	-	0.00	-	-	-	0.00	0.01	-	0.01	-	0.00	0.00	-0.03

Table S.3: Differences in An partial charge predicted with Lowdin including (ad) and excluding (nd) the most diffuse basis functions in the bob-An basis sets. [Calculated as ad - nd].

Functional	Non-An Basis Set	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr
LDA	6-31G*	-0.14	-	-0.05	-0.01	-	-0.12	-0.22	-0.13	-0.01	-0.01	-0.01	-0.31	-0.04	-0.01	-0.31
	cc-pVDZ	-0.11	-	-0.03	-	0.00	-0.09	-	-0.09	-0.01	-0.01	-	-0.23	0.01	-	-0.22
	6-311++G**	-0.08	-	-0.03	0.00	0.00	-0.08	-	-0.08	-0.01	0.00	0.00	-0.21	-0.03	-	-0.20
	cc-pVTZ	-0.10	-	-0.03	-0.01	0.00	-0.08	-0.15	-0.09	-0.01	-0.01	-0.01	-0.22	-	-	-0.22
	cc-pVQZ	-0.09	-	-0.02	-0.01	-0.01	-0.08	-0.14	-0.08	-0.01	0.00	0.00	-0.21	-	0.00	-0.19
TPSS	6-31G*	-0.13	-0.37	-	-	-	-0.12	-	-0.14	-0.01	-0.01	0.00	-	-0.01	-	-0.30
	cc-pVDZ	-0.11	-0.27	-	-	-	-0.09	-	-0.10	0.01	-	-	-	-	-	-0.21
	6-311++G**	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	cc-pVTZ	-0.10	-0.26	-	-	-	-0.09	-	-0.09	-0.01	0.00	-	-0.23	-	-	-
	cc-pVQZ	-0.10	-0.25	-	0.00	0.00	-	-	-0.08	-0.01	-0.01	0.00	-0.20	-	-	-0.18
B3LYP	6-31G*	-0.14	-0.38	-0.03	-0.01	-0.02	-0.12	-0.22	-0.14	-0.01	-0.02	0.00	-	-0.02	-0.01	-0.31
	cc-pVDZ	-0.11	-0.28	-0.04	-0.01	-0.01	-0.10	-0.17	-0.10	-0.01	0.00	-0.01	-	-	-0.01	-0.22
	6-311++G**	-0.09	-0.26	-0.02	-0.01	-0.02	-0.07	-0.14	-0.09	-0.01	0.00	0.02	-0.20	0.00	0.00	-0.20
	cc-pVTZ	-0.11	-0.29	-0.02	-0.01	0.00	-0.09	-0.16	-0.10	-0.01	0.00	-0.02	-0.21	0.00	0.00	-0.22
	cc-pVQZ	-0.09	-0.24	-0.03	-0.01	0.01	-0.08	-0.14	-0.08	0.00	-0.01	0.01	-0.19	0.01	0.00	-0.20
PBE0	6-31G*	-0.14	-0.37	-0.03	-0.03	0.00	-0.11	-0.21	-0.13	-0.01	0.00	-0.01	-0.29	-0.01	-0.01	-0.30
	cc-pVDZ	-0.10		-0.03	-0.01	-0.01	-0.10	-0.16	-0.10	0.00	-0.01	0.00	-0.21	-0.01	-0.01	-0.21
	6-311++G**	-0.08	-0.25	-0.01	-0.01	0.00	-0.13	-0.14	-0.09	-0.01	0.00	0.00	-0.19	0.00	-0.01	-0.20
	cc-pVTZ	-0.10	-0.27	-0.02	-0.01	0.00	-0.09	-0.15	-0.09	0.00	0.00	-0.01	-0.20	0.00	-0.01	-0.21
	cc-pVQZ	-0.09	-0.25	-0.02	-0.01	-0.02	-0.10	-0.14	-0.08	-0.01	0.00	-0.01	-0.18	-0.01	0.00	-0.18
B972	6-31G*	-0.13	-0.36	-0.04	-0.01	-0.01	-0.13	-0.21	-0.14	-0.01	0.01	-0.01	-	-0.04	-0.01	-0.30
	cc-pVDZ	-0.11	-0.28	-0.03	-0.01	0.00	-0.10	-0.16	-0.09	-0.01	-0.01	0.00	-	-0.01	-0.01	-0.21
	6-311++G**	-0.08	-0.26	-0.01	-0.01	0.01	-0.03	-0.14	-0.08	-0.01	-0.01	-0.03	-0.18	-	0.00	-0.20
	cc-pVTZ	-0.10	-0.27	-0.02	-0.01	-0.01	-0.08	-0.15	-0.09	0.00	0.00	-0.01	-0.20	-0.01	-0.01	-0.20
	cc-pVQZ	-0.09	-0.24	-0.02	-0.01	-0.01	-0.07	-0.14	-0.07	-0.01	-0.01	-0.01	-0.18	0.00	0.00	-0.18
M06	6-31G*	-0.14	-0.37	-0.03	-0.01	-	-0.12	-0.22	-0.13	-0.01	-0.01	-0.01	-0.30	-0.01	-0.01	-0.30
	cc-pVDZ	-0.10	-0.27	-0.02	0.01	-	-0.09	-0.16	-0.10	-	-0.01	0.01	-0.22	-0.01	-0.01	-0.21
	6-311++G**	-0.08	-0.26	-0.02	-	-	-0.06	-0.14	-0.09	-0.01	-0.02	0.00	-0.19	-0.01	0.00	-0.20
	cc-pVTZ	-0.10	-0.26	0.01	0.00	-	-0.08	-0.15	-0.08	-	-0.01	-	-	0.00	-0.01	-0.20
	cc-pVQZ	-0.09	-0.24	-0.02			-0.06	-0.13	-0.08	0.00	-	-	-0.18	0.00	-0.01	-0.18
M11	6-31G*	-0.14	-0.38	-0.03	-0.01	-	-0.11	-	-0.14	-	-	-	-	-0.02	-0.01	-0.31
	cc-pVDZ	-0.11	-0.28	-0.01	-	-	-0.10	-	-0.10	-0.01	-	-	-	-0.01	-0.01	-0.23
	6-311++G**	-0.09	-0.26	-0.02	0.00	-	-0.12	-	-0.09	0.00	0.05	-	-	-	0.00	-0.21
	cc-pVTZ	-0.10	-	-0.03	0.00	-	-0.11	-	-0.10	-0.01	-0.01	0.00	-	-0.01	-0.01	-0.22
	cc-pVQZ	-0.10	-0.25	-	-0.01	-	-	-	-0.08	-0.01	-	0.01	-	-0.01	-0.01	-0.19

Table S.4 Predicted 5f orbital occupancies with NBO for An in proposed model for $[An(NO_3)]^{2+}$ structure, with LDA, TPSS, B3LYP, and PBE0.

Functional	An basis set (ad/nd)	Non-An basis set	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr
LDA	ad	6-31G*	0.1		2.3	3.2	4.3	5.4	6.4	7.2	8.3	9.3	10.4	11.6	12.7	13.8	14.0
		cc-pVDZ	0.1		2.3		4.3	5.3		7.2	8.3	9.4		11.6	12.7	13.8	14.0
		6-311++G**	0.1		2.3	3.2	4.3	5.3		7.2	8.3	9.4	10.4	11.6	12.7	13.8	14.0
		cc-pVTZ	0.1		2.3	3.2	4.3	5.4	6.4	7.2	8.3	9.3	10.4	11.6	12.7		14.0
		cc-pVQZ	0.1		2.3	3.2	4.3	5.4	6.4	7.2	8.3	9.3	10.4	11.6		13.8	14.0
	nd	6-31G*	0.1		2.3	3.2		5.3	6.4	7.2	8.3	9.3	10.4	11.6	12.7	13.8	14.0
		cc-pVDZ	0.1		2.3		4.3	5.3	6.4	7.2	8.3	9.4		11.6	12.7		14.0
		6-311++G**	0.1		2.3	3.2	4.3	5.3	6.4	7.2	8.3	9.4	10.4	11.6	12.7		14.0
		cc-pVTZ	0.1		2.3	3.2	4.3	5.4	6.4	7.2	8.3	9.3	10.4	11.6			14.0
		cc-pVQZ	0.1		2.3	3.2	4.3	5.3	6.4	7.2	8.3	9.3	10.4	11.6	12.7	13.8	14.0
TPSS	ad	6-31G*	0.1	0.7	2.2	3.1	4.2	5.2		7.1	8.2	9.3	10.2		12.8	14.0	14.0
		cc-pVDZ	0.1	0.7	2.2	3.1	4.2	5.2		7.1	8.2		10.4			14.0	14.0
		cc-pVTZ	0.1	0.7				5.2		7.1	8.2	9.3	10.2	11.4		14.0	14.0
		cc-pVQZ	0.1	0.7		3.1	4.2			7.1	8.2	9.3	10.2	11.4			14.0
	nd	6-31G*	0.1	0.7				5.2	6.4	7.1	8.2	9.3	10.3	11.4	12.7		14.0
		cc-pVDZ	0.1	0.7				5.2	6.4	7.1	8.2	9.3			12.7		14.0
		6-311++G**	0.1		2.2	3.1	4.2	5.2		7.1	8.2	9.3	10.3			14.0	14.0
		cc-pVTZ	0.1	0.7		3.1	4.2	5.2	6.4	7.1	8.2	9.3		11.4	12.7		
		cc-pVQZ	0.1	0.7	2.2	3.1	4.2	5.2	6.4	7.1	8.2	9.3	10.2	11.4		14.0	14.0
B3LYP	ad	6-31G*	0.1	0.7	2.2	3.1	4.2	5.2	6.3	7.1	8.2	9.2	10.2		13.0	14.0	14.0
		cc-pVDZ	0.1	0.7	2.2	3.1	4.1	5.2	6.3	7.1	8.2	9.2	10.2			14.0	14.0
		6-311++G**	0.1	0.7	2.2	3.1	4.2	5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVTZ	0.1	0.7	2.2	3.1	4.2	5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVQZ	0.1	0.7	2.2	3.1	4.1	5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
	nd	6-31G*	0.1	0.7	2.2	3.1	4.1	5.2	6.3	7.1	8.2	9.2	10.2		13.0	14.0	14.0
		cc-pVDZ	0.1	0.7	2.2	3.1	4.2	5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		6-311++G**	0.1	0.7	2.2	3.1	4.1	5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVTZ	0.1	0.7	2.2	3.1	4.2	5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVQZ	0.1	0.7	2.2	3.1	4.2	5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
PBE0	ad	6-31G*	0.1	0.7	2.2	3.1	4.1	5.2	6.3	7.1	8.1	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVDZ	0.1	0.7	2.2	3.1	4.2	5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		6-311++G**	0.1	0.7	2.2	3.1	4.1	5.2	6.3	7.1	8.2	9.1	10.2	12.0	13.0	14.0	14.0
		cc-pVTZ	0.1	0.7	2.2	3.1	4.1	5.2	6.3	7.1	8.1	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVQZ	0.1	0.6	2.2	3.1	4.1	5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
	nd	6-31G*	0.1	0.7	2.2	3.1	4.2	5.2	6.3	7.1	8.1	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVDZ	0.1		2.2	3.1	4.1	5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		6-311++G**	0.1	0.7	2.2	3.1	4.1	5.1	6.3	7.1	8.2	9.1	10.2	12.0	13.0	14.0	14.0
		cc-pVTZ	0.1	0.7	2.2	3.1	4.1	5.2	6.3	7.1	8.1	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVQZ	0.1	0.7	2.2	3.1	4.1	5.1	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0

Table S.5: Predicted 5f orbital occupancies with NBO for An in proposed model for $[An(NO_3)]^{2+}$ structure, with B972, M06, and M11.

Functional	An basis set (ad/nd)	Non-An basis set	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr
B972	ad	6-31G*	0.1	0.9	2.2	3.1	4.2	5.2	6.4	7.1	8.2	9.2	10.3		13.0	14.0	14.0
		cc-pVDZ	0.1	0.9	2.2	3.1	4.2	5.2	6.4	7.1	8.2	9.2	10.3	12.0	13.0	14.0	14.0
		6-311++G**	0.1	0.9	2.2	3.1	4.2	5.1	6.4	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVTZ	0.1	0.9	2.2	3.1	4.2	5.2	6.4	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVQZ	0.1	0.9	2.2	3.1	4.2	5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
	nd	6-31G*	0.1	0.9	2.2	3.1	4.2	5.2	6.4	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVDZ	0.1	0.9	2.2	3.1	4.2	5.2	6.4	7.1	8.2	9.2	10.2		13.0	14.0	14.0
		6-311++G**	0.1	0.9	2.2	3.1	4.2	5.2	6.4	7.1	8.2	9.2	10.3	12.0		14.0	14.0
		cc-pVTZ	0.1	0.9	2.2	3.1	4.2	5.2	6.4	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVQZ	0.1	0.9	2.2	3.1	4.2	5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
M06	ad	6-31G*	0.1	1.0	2.2	3.1		5.2	6.3	7.1	8.1	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVDZ	0.1	1.0	2.2	3.1		5.2	6.3	7.1	8.1	9.2	10.2	12.0	13.0	14.0	14.0
		6-311++G**	0.1	1.0	2.2	3.1		5.1	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVTZ	0.1	1.0	2.2	3.1		5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVQZ	0.1	1.0	2.2			5.2	6.3	7.1	8.2			12.0	13.0	14.0	14.0
	nd	6-31G*	0.1	1.0	2.2	3.1		5.2	6.3	7.1	8.1	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVDZ	0.1	1.0	2.2	3.1		5.2	6.3	7.1		9.2	10.2	12.0	13.0	14.0	14.0
		6-311++G**	0.1	1.0	2.2			5.2	6.3	7.1	8.2	9.2	10.2	12.0	13.0	14.0	14.0
		cc-pVTZ	0.1	1.1	2.2	3.1		5.2	6.3	7.1		9.2			13.0	14.0	14.0
		cc-pVQZ	0.1	1.0	2.2	3.1		5.2	6.3	7.1	8.2		10.2	12.0	13.0	14.0	14.0
M11	ad	6-31G*	0.1	0.7	2.1	3.1		5.1		7.1	8.1		10.1		13.0	14.0	14.0
		cc-pVDZ	0.1	0.8	2.1			5.1		7.1	8.1		10.1		13.0	14.0	14.0
		6-311++G**	0.1	0.7	2.1	3.1		5.2		7.1	8.1	9.0	10.1			14.0	14.0
		cc-pVTZ	0.1	0.7	2.1	3.1		5.2		7.1	8.1	9.1	10.1		13.0	14.0	14.0
		cc-pVQZ	0.1	0.7		3.1				7.1	8.1		10.1		13.0	14.0	14.0
	nd	6-31G*	0.1	0.7	2.1	3.1		5.2		7.1		9.1			13.0	14.0	14.0
		cc-pVDZ	0.1	0.7	2.1	3.1		5.1		7.1	8.1	9.1			13.0	14.0	14.0
		6-311++G**	0.1	0.7	2.1	3.1		5.1		7.1	8.1	9.1			13.0	14.0	14.0
		cc-pVTZ	0.1		2.1	3.1		5.1		7.1	8.1	9.1	10.1		13.0	14.0	14.0
		cc-pVQZ	0.1	0.7	2.1	3.1				7.1	8.1		10.1		13.0	14.0	14.0

Table S.6: An-O1 distance (in Å) in for $[An(NO_3)]^{2+}_{(g)}$ [with An = Ac to Lr] calculated with LDA, the TPSS, B3LYP, and PBE0 functionals, the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ basis set for non-An atoms, and the Stuttgart RSC 1997/ECP for the actinide atoms (including and excluding the most diffuse basis functions in the basis set).

Functional	An basis set (ad/nd)	Non-An basis set	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr
LDA	ad	6-31G*	2.30	-	2.24	2.17	2.16	2.16	2.19	2.15	2.15	2.13	2.14	2.15	2.23	2.32	2.07
		cc-pVDZ	2.31	-	2.25	-	2.17	2.18	-	2.15	2.15	2.13	-	2.15	2.26	2.33	2.08
		6-311++G**	2.31	-	2.25	2.18	2.15	2.19	-	2.16	2.16	2.16	2.16	2.16	2.25	2.34	2.08
		cc-pVTZ	2.30	-	2.24	2.17	2.14	2.16	2.19	2.15	2.14	2.13	2.13	2.14	2.23	2.32	2.07
		cc-pVQZ	2.29	-	2.22	2.17	2.14	2.16	2.18	2.15	2.14	2.12	2.13	2.14	-	2.32	2.07
	nd	6-31G*	2.30	-	2.24	2.18		2.17	2.19	2.15	2.15	2.13	2.14	2.18	2.25	2.32	2.07
		cc-pVDZ	2.30	-	2.25	-	2.17	2.18	2.20	2.15	2.15	2.13	-	2.19	2.24	-	2.07
		6-311++G**	2.31	-	2.28	2.18	2.18	2.18	2.21	2.16	2.16	2.16	2.16	2.16	2.27	-	2.08
		cc-pVTZ	2.30	-	2.23	2.17	2.14	2.16	2.19	2.15	2.14	2.13	2.14	2.18	-	-	2.07
		cc-pVQZ	2.29	-	2.22	2.18	2.16	2.16	2.18	2.15	2.14	2.12	2.14	2.14	2.24	2.32	2.07
TPSS	ad	6-31G*	2.33	2.27	2.29	2.21	2.21	2.19	-	2.18	2.18	2.17	2.15	-	2.42	2.53	2.11
		cc-pVDZ	2.33	2.27	2.29	2.22	2.19	2.19	-	2.18	2.18	-	2.21	-	-	2.53	2.11
		6-311++G**	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
		cc-pVTZ	2.32	2.26	-	-	-	2.18	-	2.18	2.17	2.17	2.13	2.16	-	2.53	2.10
		cc-pVQZ	2.32	2.26	-	2.20	2.20	-	-	2.17	2.17	2.16	2.14	2.14	-	-	2.10
	nd	6-31G*	2.33	2.27	-	-	-	2.19	2.21	2.18	2.17	2.17	2.16	2.16	2.29	-	2.10
		cc-pVDZ	2.33	2.27	-	-	-	2.19	2.22	2.18	2.18	2.18	-	-	2.32	-	2.11
		6-311++G**	2.34	-	2.29	2.22	2.21	2.20	-	2.19	2.18	2.18	2.16	-	-	2.56	2.11
		cc-pVTZ	2.32	2.26	-	2.20	2.20	2.18	2.21	2.18	2.17	2.16	-	2.21	2.28	-	-
		cc-pVQZ	2.32	2.26	2.26	2.20	2.19	2.18	2.20	2.17	2.16	2.16	2.13	2.14	-	2.52	2.10
B3LYP	ad	6-31G*	2.34	2.27	2.29	2.22	2.22	2.20	2.21	2.19	2.18	2.17	2.15	-	2.58	2.57	2.12
		cc-pVDZ	2.34	2.28	2.29	2.23	2.23	2.20	2.22	2.19	2.18	2.17	2.15	-	-	2.58	2.12
		6-311++G**	2.35	2.28	2.29	2.24	2.22	2.22	2.23	2.20	2.18	2.18	2.15	-	2.64	2.61	2.12
		cc-pVTZ	2.34	2.26	2.29	2.22	2.21	2.20	2.21	2.19	2.18	2.16	2.15	-	2.60	2.58	2.11
		cc-pVQZ	2.33	2.27	2.28	2.23	2.21	2.21	2.20	2.19	2.17	2.16	2.14	-	2.59	2.58	2.11
	nd	6-31G*	2.34	2.27	2.28	2.23	2.21	2.20	2.21	2.19	2.18	2.15	2.15	-	2.53	2.57	2.11
		cc-pVDZ	2.34	2.28	2.31	2.23	2.22	2.20	2.22	2.19	2.18	2.17	2.15	-	2.65	2.57	2.11
		6-311++G**	2.35	2.29	2.30	2.25	2.23	2.21	2.23	2.20	2.19	2.17	2.16	-	2.63	2.61	2.12
		cc-pVTZ	2.34	2.27	2.28	2.23	2.20	2.20	2.21	2.19	2.18	2.16	2.15	-	2.55	2.58	2.11
		cc-pVQZ	2.33	2.26	2.27	2.22	2.21	2.24	2.20	2.19	2.17	2.16	2.15	-	2.58	2.58	2.11
PBE0	ad	6-31G*	2.33	2.26	2.27	2.21	2.20	2.19	2.19	2.17	2.16	2.15	2.14	2.56	2.59	2.56	2.10
		cc-pVDZ	2.33	2.26	2.28	2.21	2.20	2.18	2.20	2.18	2.17	2.15	2.13	2.57	2.56	2.56	2.10
		6-311++G**	2.33	2.27	2.28	2.23	2.21	2.19	2.20	2.18	2.17	2.13	2.14	2.60	2.62	2.59	2.10
		cc-pVTZ	2.32	2.25	2.26	2.21	2.19	2.17	2.18	2.17	2.15	2.13	2.12	2.58	2.59	2.56	2.09
		cc-pVQZ	2.31	2.24	2.25	2.20	2.19	2.17	2.18	2.17	2.15	2.14	2.13	2.57	2.58	2.56	2.09
	nd	6-31G*	2.32	2.26	2.27	2.22	2.19	2.17	2.19	2.17	2.16	2.14	2.13	2.57	2.60	2.56	2.10
		cc-pVDZ	2.33	-	2.28	2.22	2.20	2.18	2.19	2.18	2.17	2.15	2.13	2.59	2.62	2.56	2.10
		6-311++G**	2.33	2.27	2.27	2.23	2.21	2.17	2.20	2.18	2.17	2.13	2.14	2.60	2.57	2.59	2.10
		cc-pVTZ	2.32	2.25	2.27	2.21	2.19	2.17	2.18	2.17	2.15	2.13	2.12	2.58	2.56	2.56	2.09
		cc-pVQZ	2.31	2.24	2.25	2.21	2.21	2.15	2.18	2.17	2.15	2.14	2.12	2.57	2.54	2.56	2.09

Table S.7: An-O1 distance (in Å) in for $[An(NO_3)]^{2+}_{(g)}$ [with An = Ac to Lr] calculated with the B972, M06, and M11 functionals, the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ basis set for non-An atoms, and the Stuttgart RSC 1997/ECP for the actinide atoms (including and excluding the most diffuse basis functions in the basis set).

Functional	An basis set (ad/nd)	Non-An basis set	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr
B972	ad	6-31G*	2.33	2.27	2.29	2.22	2.20	2.19	2.22	2.18	2.17	2.14	2.14	-	2.58	2.59	2.11
		cc-pVDZ	2.33	2.27	2.30	2.22	2.21	2.19	2.23	2.18	2.17	2.16	2.15	2.60	2.60	2.59	2.11
		6-311++G**	2.33	2.28	2.29	2.23	2.21	2.21	2.23	2.18	2.18	2.16	2.15	2.59	2.63	2.62	2.11
		cc-pVTZ	2.32	2.26	2.27	2.21	2.18	2.19	2.21	2.17	2.17	2.15	2.14	2.67	2.64	2.59	2.10
		cc-pVQZ	2.31	2.26	2.26	2.21	2.18	2.17	2.20	2.17	2.15	2.15	2.13	2.57	2.61	2.59	2.10
	nd	6-31G*	2.33	2.27	2.30	2.22	2.20	2.19	2.22	2.18	2.17	2.16	2.15	-	2.59	2.59	2.10
		cc-pVDZ	2.33	2.27	2.30	2.22	2.21	2.19	2.23	2.18	2.17	2.16	2.15	-	2.60	2.59	2.11
		6-311++G**	2.33	2.28	2.29	2.22	2.20	2.20	2.23	2.19	2.18	2.15	2.16	2.60	-	2.62	2.11
		cc-pVTZ	2.32	2.26	2.26	2.21	2.18	2.18	2.21	2.17	2.16	2.15	2.14	2.59	2.57	2.59	2.10
		cc-pVQZ	2.31	2.26	2.26	2.20	2.19	2.17	2.20	2.17	2.15	2.15	2.13	2.59	2.59	2.59	2.09
M06	ad	6-31G*	2.34	2.28	2.31	2.24	-	2.21	2.21	2.19	2.16	2.17	2.15	2.60	2.60	2.58	2.12
		cc-pVDZ	2.34	2.29	2.28	2.26	-	2.19	2.22	2.19	2.17	2.18	2.15	2.63	2.60	2.59	2.12
		6-311++G**	2.34	2.29	2.32	2.29	-	2.25	2.22	2.20	2.18	2.18	2.12	2.66	2.63	2.61	2.12
		cc-pVTZ	2.32	2.27	2.28	2.25	-	2.18	2.20	2.18	2.17	2.16	2.15	2.63	2.61	2.58	2.11
		cc-pVQZ	2.31	2.27	2.23	-	-	2.19	2.19	2.18	2.15	-	-	2.59	2.59	2.57	2.10
	nd	6-31G*	2.33	2.28	2.32	2.23	-	2.21	2.21	2.19	2.18	2.14	2.15	2.59	2.59	2.58	2.12
		cc-pVDZ	2.34	2.28	2.28	2.24	-	2.20	2.22	2.19	-	2.14	2.15	2.55	2.60	2.59	2.12
		6-311++G**	2.34	2.29	2.32	-	-	2.22	2.23	2.20	2.18	2.21	2.14	2.62	2.62	2.61	2.12
		cc-pVTZ	2.32	2.27	2.30	2.22	-	2.20	2.20	2.18	-	2.13	-	-	2.56	2.58	2.11
		cc-pVQZ	2.31	2.27	2.25	2.19	-	2.19	2.19	2.18	2.16	-	2.19	2.59	2.53	2.57	2.10
M11	ad	6-31G*	2.35	2.28	2.29	2.27	-	2.17	-	2.20	2.19	-	2.14	-	2.55	2.57	2.12
		cc-pVDZ	2.35	2.29	2.30	-	-	2.17	-	2.20	2.18	-	2.16	-	2.70	2.57	2.12
		6-311++G**	2.36	2.30	2.34	2.23	-	2.24	-	2.21	2.19	2.19	2.10	-	-	2.61	2.13
		cc-pVTZ	2.35	2.28	2.27	2.26	-	2.21	-	2.19	2.17	2.15	2.11	-	2.55	2.58	2.12
		cc-pVQZ	2.34	2.28	-	2.26	-	-	-	2.19	2.17	-	2.11	-	2.58	2.58	2.11
	nd	6-31G*	2.34	2.28	2.31	2.25	-	2.21	-	2.19	-	2.12	-	-	2.55	2.57	2.12
		cc-pVDZ	2.35	2.28	2.31	2.25	-	2.17	-	2.20	2.18	2.14	-	-	2.55	2.57	2.12
		6-311++G**	2.36	2.30	2.26	2.26	-	2.20	-	2.21	2.18	2.14	-	-	2.56	2.61	2.12
		cc-pVTZ	2.34	-	2.28	2.23	-	2.22	-	2.19	2.17	2.15	2.10	-	2.55	2.58	2.12
		cc-pVQZ	2.34	2.28	2.31	2.24	-	-	-	2.19	2.17	-	2.14	-	2.56	2.58	2.11

Table S.8: An-O1 distance (in Å), O1-An-O2 angle (in °), and O1-N-O2 (in °) calculated with the B3LYP functionals, the 6-311++G** basis set for non-An atoms, and the Stuttgart RSC 1997/ECP for the actinide atoms (including all basis functions in the basis set), for $[An(NO_3)]^{2+}$ and $[An(NO_3)]^{+}$ [with An = Fm, Md, and No].

An	An-O1 distance (Å)		O1-An-O2 angle (°)		O1-N-O2 angle (°)	
	III	IV	III	IV	III	IV
Fm	2.28	2.60	57.00	48.93	112.93	122.85
Md	2.31	2.64	56.99	48.95	112.93	122.62
No	2.28	2.61	57.33	49.06	113.12	122.91

Table S.9: An-O1 distance (in Å) calculated with the B3LYP and PBE0 functionals, the 6-31G* and 6-311++G** basis set for non-An atoms, and the Stuttgart RSC 1997/ECP for the actinide atoms (including all basis functions in the basis set).

An	B3LYP				PBE0			
	6-31G*		6-311++G**		6-31G*		6-311++G**	
	(g)	(aq)	(g)	(aq)	(g)	(aq)	(g)	(aq)
Ac	2.34	2.48	2.35	2.51	2.33	2.47	2.33	2.49
Pa	2.29	2.41	2.29	2.44	2.27	2.40	2.28	2.42
U	2.22	2.35	2.24	2.38	2.21	2.34	2.23	2.36
Np	2.22	2.33	2.22	2.36	2.20	2.32	2.21	2.34
Pu	2.20	2.31	2.22	2.33	2.19	2.30	2.19	2.31
Am	2.21	2.30	2.23	2.33	2.19	2.29	2.20	2.31
Cm	2.19	2.30	2.20	2.32	2.17	2.29	2.18	2.31
Bk	2.18	2.28	2.18	-	2.16	2.27	2.17	2.29
Cf	2.17	2.26	2.18	2.29	2.15	2.25	2.13	2.27
Es	2.15	2.25	-	2.28	2.14	2.24	-	2.26
Fm	-	-	2.60	2.26	2.56	2.23	2.60	2.24
Md	2.58	-	2.64	2.31	2.59	-	2.62	2.26
Lr	2.12	2.34	2.12	2.37	2.10	2.33	2.10	2.36

Table S.10: O1-An-O2 angle (in °) calculated with the B3LYP and PBE0 functionals, the 6-31G* and 6-311++G** basis set for non-An atoms, and the Stuttgart RSC 1997/ECP for the actinide atoms (including all basis functions in the basis set).

An	B3LYP				PBE0			
	6-31G*		6-311++G**		6-31G*		6-311++G**	
	(g)	(aq)	(g)	(aq)	(g)	(aq)	(g)	(aq)
Ac	55.66	51.35	55.30	50.37	55.41	51.21	55.13	50.42
Th	57.19	81.47	56.84	81.91	56.90	63.64	56.51	-
Pa	56.89	53.21	56.56	52.22	56.41	53.02	56.08	52.19
U	58.36	54.75	57.81	53.72	58.09	54.63	57.60	53.81
Np	58.75	55.47	58.27	54.42	58.45	55.32	57.99	54.44
Pu	58.90	56.16	58.31	55.31	58.72	56.01	58.48	55.31
Am	58.21	56.25	57.63	55.42	58.21	56.13	57.73	55.44
Cm	59.76	56.47	59.29	55.59	59.51	56.23	59.12	55.51
Bk	59.65	57.07	59.61	-	59.91	56.84	59.14	56.15
Cf	59.86	57.65	59.31	56.59	59.86	57.43	60.17	56.71
Es	60.54	57.88	-	57.10	60.28	57.61	-	56.98
Fm	-	-	48.93	57.29	50.16	57.92	49.01	57.35
Md	50.00	-	48.95	55.71	49.73	-	48.90	56.55
No	50.04	53.19	49.06	52.26	49.93	52.78	49.15	51.86
Lr	62.58	55.20	62.25	54.34	62.19	54.89	61.91	54.15

Table S.11: Predicted Gibbs free energy of reaction for the $[An(NO_3)]^{2+}$ compounds (from Ac to Lr) with CCSD(T), the cc-pVQZ-DK, and the cc-pVTZ-DK basis set for N and O atoms, and the cc-pVTZ-X2C basis set for the actinides.

	CBS	**cc-pVQZ-DK**	**cc-pVTZ-DK**
Ac	-448.34	-450.57	-456.16
Th	-475.73	-478.04	-483.55
Pa	-494.73	-496.62	-501.52
U	-511.24	-512.84	-517.27
Np	-520.96	-522.59	-527.09
Pu	-521.40	-522.95	-527.39
Am	-496.93	-498.60	-503.15
Cm	-499.42	-501.00	-505.33
Bk	-509.96	-511.56	-516.02
Cf	-515.31	-516.88	-521.33
Es	-557.51	-558.59	-562.58
Fm	-568.52	-568.62	-571.23
Md	-562.86	-563.40	-566.43
No	-600.32	-600.90	-604.07
Lr	-520.91	-522.16	-525.88

Table S.12: $\Delta(\Delta G)_{rxn}$ for $[An(NO_3)]^{2+}$ including and excluding the most diffuse functions in the An basis set with LDA, TPSS, B3LYP, PBE0, B972, M06, and M11 with the Stuttgart RSC 1997 ECP and associated basis set, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O. Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn} = \Delta G_{rxn,ad} - \Delta G_{rxn,nd}$, as indicated in Eq. 6; with An = Th - Lr].

Functional	Non-An basis set	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr
LDA	6-31G*	-1.15	-	-0.93	-0.01	-	-0.52	-1.06	-0.67	-0.49	-0.09	0.54	-2.99	-0.99	-4.76	-2.72
	cc-pVDZ	-1.49	-	-1.42	-	-0.27	-0.73	-	-0.85	-0.37	-0.06	-	-3.57	0.93	-	-3.59
	6-311++G**	-1.37	-	-1.54	0.09	-0.27	-0.23	-	-0.33	-0.30	-0.23	0.47	-1.85	-1.07	-	-0.84
	cc-pVTZ	-0.45	-	-0.17	0.40	-0.24	-0.14	-0.26	-0.19	-0.05	-0.02	0.23	-1.73	-	-	-0.93
	cc-pVQZ	-0.34	-	-0.08	-0.50	-0.12	-0.03	-0.09	-0.08	-0.02	-0.03	0.18	-1.49	-	-4.66	-0.33
TPSS	6-31G*	-1.01	-0.46	-	-	-	-2.63	-	-0.59	-0.08	-0.55	-1.69	-	0.52	-	-2.37
	cc-pVDZ	-1.30	-1.09	-	-	-	-2.77	-	-0.75	2.06	-	-	-	-	-	-3.08
	6-311++G**	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	cc-pVTZ	-0.41	0.84	-	-	-	-2.25	-	-0.16	0.78	-1.19	-	-9.98	-	-	-
	cc-pVQZ	-0.30	1.25	-	-13.20	0.05	-	-	-0.07	-0.02	-0.47	0.20	-0.09	-	-	-0.29
B3LYP	6-31G*	-1.22	-0.07	-1.74	-1.37	-10.49	-6.73	-1.13	-0.72	-0.12	0.75	-11.24	-	-4.66	-0.11	-2.76
	cc-pVDZ	-1.57	-3.35	-4.58	-1.29	-13.52	-10.59	-1.45	-0.91	-0.16	2.30	-9.46	-	-	-0.17	-3.63
	6-311++G**	-1.14	-0.38	-2.34	-1.19	-10.70	-8.20	-0.34	-0.29	-1.90	3.07	-5.77	-0.54	0.03	0.04	-0.82
	cc-pVTZ	-0.47	-0.48	-1.60	-1.29	-13.64	-9.10	-0.25	-0.17	-0.01	2.48	-9.25	-0.48	-0.12	-0.04	-0.93
	cc-pVQZ	-0.31	-0.95	-1.48	-1.14	-13.35	-20.31	-0.10	-0.07	-0.02	3.08	-2.94	-0.13	0.11	-0.01	-0.33
PBE0	6-31G*	-1.01	-2.18	-1.96	-2.29	-2.80	0.80	2.55	-0.59	-0.09	0.36	23.40	1.11	0.79	-0.06	-2.36
	cc-pVDZ	-1.33	-	-2.11	-0.34	1.11	-1.65	2.25	-0.76	-0.15	-0.14	21.65	0.74	0.89	-0.15	-3.16
	6-311++G**	-1.24	-1.03	-1.23	-0.18	-0.69	-12.90	3.13	-0.34	-0.04	-0.06	22.90	-0.54	1.00	-0.03	-0.87
	cc-pVTZ	-0.41	-0.92	-1.95	-0.22	-0.80	-1.80	3.28	-0.15	-0.04	-0.59	23.50	-0.54	0.94	-0.04	-0.82
	cc-pVQZ	-0.28	-0.50	-1.18	-2.18	-6.39	-10.57	3.44	-0.06	-0.01	-0.02	20.30	-0.21	0.85	-0.01	-0.29
B972	6-31G*	-0.98	-2.14	-2.37	-0.11	-0.08	-0.40	-0.95	-0.57	-0.10	-3.91	-0.29	-	16.77	-0.07	-2.26
	cc-pVDZ	-1.29	-2.82	-2.51	-0.15	-1.18	-1.32	-1.22	-0.74	-0.13	-5.42	-0.43	-	-3.54	-0.11	-3.03
	6-311++G**	-1.28	-0.97	-2.11	-0.05	-1.18	10.13	-0.38	-0.33	-0.04	-7.78	-6.28	-0.51	-	-0.05	-0.85
	cc-pVTZ	-0.41	-0.86	-1.52	-0.04	-0.14	0.89	-0.24	-0.15	2.08	-5.33	-0.04	-0.88	0.07	-0.05	-0.79
	cc-pVQZ	-0.30	-0.45	-1.97	0.01	0.95	-0.36	-0.09	-0.06	-0.02	-5.30	-0.03	-0.21	0.44	-0.01	-0.27
M06	6-31G*	-1.02	-4.92	-0.35	-0.19	-	2.65	3.80	-0.59	0.35	4.23	-1.74	-1.14	-0.52	-0.01	-2.33
	cc-pVDZ	-1.28	-5.42	-3.06	0.72	-	2.60	3.54	-0.74	-	4.20	1.85	-3.32	-0.18	-0.06	-3.03
	6-311++G**	-1.10	-3.58	-2.66	-	-	12.50	4.36	-0.33	-0.82	1.52	-0.35	-1.55	-4.39	-0.01	-0.82
	cc-pVTZ	-0.36	-5.85	0.11	5.69	-	2.56	4.55	-0.15	-	4.29	-	-	-17.27	0.02	-0.71
	cc-pVQZ	-0.29	-2.80	-1.45	-	-	1.85	4.64	-0.08	1.66	-	-	-0.20	-17.54	17.12	-0.26
M11	6-31G*	-1.19	-2.37	-0.45	-1.60	-	-1.11	-	-0.66	-	-	-	-	0.01	-12.33	-2.61
	cc-pVDZ	-1.58	-3.11	1.60	-	-	-4.96	-	-0.85	-2.48	-	-	-	7.69	-12.24	-3.55
	6-311++G**	-1.17	-1.20	5.13	-2.39	-	-10.99	-	-0.40	3.41	12.26	-	-	-	-0.06	-0.98
	cc-pVTZ	-0.44	-	24.51	-0.67	-	-8.59	-	-0.17	-2.68	1.44	-19.77	-	0.03	-0.19	-0.76
	cc-pVQZ	-0.30	-0.55	-	-2.20	-	-	-	-0.08	-2.66	-	-3.85	-	-4.84	-0.06	-0.25

Table S.13: $\Delta(\Delta G)_{rxn}$ for $[An(NO_3)]^{2+}$ normalized to Ac calculated with LDA and TPSS with the Stuttgart RSC 1997 ECP and basis set including (ad) and excluding (nd) the most diffuse basis functions for An, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O. $\Delta(\Delta G)_{rxn}$ calculated with CCSD(T)-FC1 includes the cc-pV∞Z-X2C basis set for An, and the cc-pV∞Z-DK basis set for N and O is included as reference (indicated as CCSD(T)/CBS). Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn} = \Delta G_{rxn,An} - \Delta G_{rxn,Ac}$, as indicated in Eq. 5.a; with An = Th - Lr].

			6-31G*	cc-pVDZ	6-311++G**	cc-pVTZ	cc-pVQZ	CCSD(T)/CBS
LDA	ad	Th	-	-	-	-	-	-27.39
		Pa	-24.19	-23.58	-24.30	-25.06	-26.41	-46.38
		U	-45.70	-	-45.08	-46.05	-46.41	-62.90
		Np	-54.97	-54.41	-54.33	-55.70	-55.61	-72.61
		Pu	-67.80	-66.88	-66.08	-67.75	-67.29	-73.06
		Am	-79.79	-	-	-79.49	-78.81	-48.59
		Cm	-65.04	-64.84	-64.08	-65.10	-64.65	-51.08
		Bk	-76.19	-75.75	-75.84	-77.04	-76.88	-61.61
		Cf	-84.49	-84.15	-84.32	-85.36	-85.14	-66.97
		Es	-93.57	-	-93.82	-94.55	-94.17	-109.17
		Fm	-114.37	-115.00	-113.30	-113.62	-112.68	-120.18
		Md	-122.44	-121.16	-123.42	-123.62	-	-114.51
		No	-146.03	-146.01	-147.26	-147.23	-146.74	-151.98
		Lr	-84.80	-84.65	-83.59	-85.19	-84.81	-72.56
	nd	Th	-	-	-	-	-	-27.39
		Pa	-24.40	-23.64	-24.13	-25.34	-26.66	-46.38
		U	-46.84	-	-46.54	-46.89	-46.25	-62.90
		Np	-	-55.62	-55.43	-55.91	-55.83	-72.61
		Pu	-68.42	-67.64	-67.21	-68.06	-67.59	-73.06
		Am	-79.88	-80.29	-79.79	-79.67	-79.06	-48.59
		Cm	-65.52	-65.48	-65.11	-65.36	-64.90	-51.08
		Bk	-76.85	-76.86	-76.90	-77.44	-77.19	-61.61
		Cf	-85.54	-85.57	-85.45	-85.78	-85.45	-66.97
		Es	-95.26	-	-95.66	-95.23	-94.69	-109.17
		Fm	-112.52	-112.92	-112.82	-112.34	-111.53	-120.18
		Md	-122.60	-123.58	-123.72	-	-122.97	-114.51
		No	-142.42	-	-	-	-142.42	-151.98
		Lr	-83.23	-82.55	-84.11	-84.70	-84.81	-72.56
TPSS	ad	Th	-23.65	-23.33	-	-23.61	-23.52	-27.39
		Pa	-15.24	-17.40	-	-	-	-46.38
		U	-50.83	-48.29	-	-	-51.74	-62.90
		Np	-43.53	-44.58	-	-	-45.93	-72.61
		Pu	-58.60	-58.35	-	-58.91	-	-73.06
		Am	-	-	-	-	-	-48.59
		Cm	-54.47	-54.24	-	-54.90	-54.48	-51.08
		Bk	-66.76	-65.05	-	-68.10	-67.91	-61.61
		Cf	-74.11	-	-	-75.42	-75.19	-66.97
		Es	-92.17	-82.04	-	-93.56	-92.95	-109.17
		Fm	-	-	-	-89.77	-87.53	-120.18
		Md	-102.31	-	-	-	-	-114.51
		No	-132.34	-132.60	-	-133.81	-	-151.98
		Lr	-76.42	-76.25	-	-77.22	-76.85	-72.56
	nd	Th	-24.20	-23.55	-	-24.86	-25.08	-27.39
		Pa	-	-	-19.13	-	-19.84	-46.38
		U	-	-	-38.49	-38.96	-38.84	-62.90
		Np	-	-	-46.01	-45.66	-46.28	-72.61
		Pu	-56.99	-56.88	-56.59	-57.06	-56.63	-73.06
		Am	-66.61	-66.97	-	-66.83	-66.24	-48.59
		Cm	-54.90	-54.80	-54.85	-55.15	-54.72	-51.08
		Bk	-67.69	-68.42	-68.36	-69.29	-68.20	-61.61
		Cf	-74.57	-74.57	-75.12	-74.63	-75.02	-66.97
		Es	-91.49	-	-89.16	-	-93.45	-109.17
		Fm	-88.35	-	-	-80.19	-87.74	-120.18
		Md	-103.84	-104.36	-	-104.64	-	-114.51
		No	-	-	-135.42	-	-133.28	-151.98
		Lr	-75.07	-74.47	-76.08	-	-76.87	-72.56

Table S.14: $\Delta(\Delta G)_{rxn}$ for $[An(NO_3)]^{2+}$ normalized to Ac calculated with B3LYP and PBE0 with the Stuttgart RSC 1997 ECP and basis set including (ad) and excluding (nd) the most diffuse basis functions for An, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O. $\Delta(\Delta G)_{rxn}$ calculated with CCSD(T)-FC1 includes the cc-pV∞Z-X2C basis set for An, and the cc-pV∞Z-DK basis set for N and O is included as reference (indicated as CCSD(T)/CBS). Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn} = \Delta G_{rxn,An} - \Delta G_{rxn,Ac}$, as indicated in Eq. 5.a; with An = Th - Lr].

			6-31G*	cc-pVDZ	6-311++G**	cc-pVTZ	cc-pVQZ	CCSD(T)/CBS
B3LYP	ad	Th	-21.00	-23.26	-21.58	-22.78	-23.26	-27.39
		Pa	-19.14	-19.07	-19.99	-20.12	-20.66	-46.38
		U	-37.77	-37.09	-37.72	-38.73	-38.75	-62.90
		Np	-53.54	-54.43	-53.57	-56.10	-56.11	-72.61
		Pu	-60.65	-64.11	-61.57	-63.58	-62.25	-73.06
		Am	-63.27	-63.59	-62.52	-63.17	-62.67	-48.59
		Cm	-53.86	-53.64	-53.28	-54.09	-53.78	-51.08
		Bk	-65.18	-64.71	-66.88	-66.40	-67.53	-61.61
		Cf	-67.64	-67.53	-67.82	-68.83	-68.80	-66.97
		Es	-97.33	-95.79	-90.00	-97.24	-90.70	-109.17
		Fm	-	-	-79.94	-79.10	-77.85	-120.18
		Md	-101.82	-	-104.14	-103.52	-102.71	-114.51
		No	-132.27	-132.45	-134.54	-134.02	-133.41	-151.98
		Lr	-77.62	-77.48	-76.62	-78.04	-77.72	-72.56
	nd	Th	-22.15	-21.47	-22.34	-22.77	-22.62	-27.39
		Pa	-18.62	-16.06	-18.79	-18.99	-19.49	-46.38
		U	-37.62	-37.36	-37.67	-37.90	-37.91	-62.90
		Np	-44.27	-42.47	-44.01	-42.93	-43.07	-72.61
		Pu	-55.14	-55.09	-54.52	-54.95	-42.25	-73.06
		Am	-63.36	-63.72	-63.33	-63.38	-62.88	-48.59
		Cm	-54.36	-54.30	-54.13	-54.38	-54.02	-51.08
		Bk	-66.28	-66.11	-66.12	-66.86	-67.82	-61.61
		Cf	-69.60	-71.40	-72.03	-71.78	-72.18	-66.97
		Es	-87.30	-87.90	-85.37	-88.45	-88.07	-109.17
		Fm	-	-80.09	-80.54	-79.09	-78.03	-120.18
		Md	-98.38	-99.01	-105.32	-103.87	-103.13	-114.51
		No	-133.37	-133.85	-135.73	-134.44	-133.70	-151.98
		Lr	-76.08	-75.42	-76.95	-77.58	-77.70	-72.56
PBE0	ad	Th	-23.54	-23.19	-22.24	-23.57	-23.50	-27.39
		Pa	-17.54	-16.95	-17.98	-18.60	-19.64	-46.38
		U	-36.41	-35.75	-36.09	-37.35	-37.38	-62.90
		Np	-45.70	-43.10	-45.25	-46.69	-46.58	-72.61
		Pu	-51.60	-54.35	-54.40	-55.66	-55.24	-73.06
		Am	-61.45	-61.72	-60.75	-61.60	-61.02	-48.59
		Cm	-51.63	-51.36	-51.05	-52.08	-51.65	-51.08
		Bk	-63.31	-61.56	-62.03	-64.67	-64.88	-61.61
		Cf	-67.04	-67.00	-62.94	-68.91	-68.31	-66.97
		Es	-70.17	-70.44	-71.07	-71.60	-71.92	-109.17
		Fm	-73.86	-74.84	-77.23	-76.55	-74.79	-120.18
		Md	-99.13	-99.34	-101.15	-100.48	-99.23	-114.51
		No	-130.69	-131.00	-132.73	-132.11	-131.02	-151.98
		Lr	-74.53	-74.33	-73.75	-75.31	-74.87	-72.56
	nd	Th	-22.36	-	-22.45	-23.07	-23.29	-27.39
		Pa	-16.59	-16.17	-18.00	-17.06	-18.75	-46.38
		U	-35.12	-36.75	-37.15	-37.54	-35.48	-62.90
		Np	-43.90	-45.54	-45.80	-46.30	-40.48	-72.61
		Pu	-53.41	-54.03	-42.75	-54.26	-44.96	-73.06
		Am	-65.01	-65.31	-65.12	-65.29	-64.74	-48.59
		Cm	-52.05	-51.93	-51.96	-52.33	-51.88	-51.08
		Bk	-64.22	-62.75	-63.23	-65.04	-65.15	-61.61
		Cf	-68.41	-68.19	-64.12	-68.74	-68.57	-66.97
		Es	-94.58	-93.43	-95.22	-95.52	-92.51	-109.17
		Fm	-75.98	-76.92	-77.92	-76.42	-74.86	-120.18
		Md	-100.93	-101.56	-103.40	-101.83	-100.36	-114.51
		No	-131.64	-132.19	-133.94	-132.48	-131.30	-151.98
		Lr	-73.18	-72.50	-74.12	-74.90	-74.87	-72.56

Table S.15: $\Delta(\Delta G)_{rxn}$ for $[An(NO_3)]^{2+}$ normalized to Ac calculated with B972 and M06 with the Stuttgart RSC 1997 ECP and basis set including (ad) and excluding (nd) the most diffuse basis functions for An, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O. $\Delta(\Delta G)_{rxn}$ calculated with CCSD(T)-FC1 includes the cc-pV∞Z-X2C basis set for An, and the cc-pV∞Z-DK basis set for N and O is included as reference (indicated as CCSD(T)/CBS). Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn} = \Delta G_{rxn,An} - \Delta G_{rxn,Ac}$, as indicated in Eq. 5.a; with An = Th - Lr].

			6-31G*	cc-pVDZ	6-311++G**	cc-pVTZ	cc-pVQZ	CCSD(T)/CBS
B972	ad	Th	-20.93	-20.65	-19.74	-20.84	-20.64	-27.39
		Pa	-17.27	-16.69	-18.58	-19.32	-20.16	-46.38
		U	-36.26	-35.63	-35.93	-37.24	-37.17	-62.90
		Np	-44.90	-44.26	-44.57	-44.85	-44.84	-72.61
		Pu	-54.43	-54.16	-43.23	-53.86	-53.52	-73.06
		Am	-63.75	-64.11	-63.23	-63.84	-63.16	-48.59
		Cm	-51.48	-51.20	-50.89	-51.93	-51.50	-51.08
		Bk	-62.89	-64.16	-62.82	-63.42	-65.75	-61.61
		Cf	-71.93	-72.95	-74.49	-74.77	-75.19	-66.97
		Es	-83.99	-83.76	-83.13	-85.34	-77.44	-109.17
		Fm	-	-78.70	-87.44	-86.96	-84.93	-120.18
		Md	-93.78	-111.05	-112.72	-111.96	-110.71	-114.51
		No	-141.57	-141.92	-143.61	-143.00	-141.96	-151.98
		Lr	-73.51	-73.28	-72.72	-74.32	-73.92	-72.56
	nd	Th	-19.77	-19.12	-20.06	-20.39	-20.49	-27.39
		Pa	-15.88	-15.47	-17.75	-18.21	-18.49	-46.38
		U	-37.13	-36.77	-37.16	-37.61	-37.48	-62.90
		Np	-45.80	-44.38	-44.67	-45.13	-46.09	-72.61
		Pu	-55.01	-54.13	-54.64	-55.16	-53.46	-73.06
		Am	-63.79	-64.18	-64.14	-64.01	-63.37	-48.59
		Cm	-51.89	-51.76	-51.84	-52.19	-51.74	-51.08
		Bk	-63.77	-65.32	-64.06	-65.91	-66.03	-61.61
		Cf	-69.00	-68.82	-68.00	-69.85	-70.19	-66.97
		Es	-84.68	-84.62	-78.13	-85.71	-77.71	-109.17
		Fm	-86.68	-	-88.20	-86.49	-85.02	-120.18
		Md	-111.53	-108.80	-	-112.43	-111.45	-114.51
		No	-142.48	-143.11	-144.84	-143.36	-142.25	-151.98
		Lr	-72.22	-71.55	-73.15	-73.94	-73.94	-72.56
M06	ad	Th	-21.06	-20.77	-20.25	-21.18	-20.87	-27.39
		Pa	-15.82	-16.21	-18.33	-15.72	-18.46	-46.38
		U	-32.26	-31.08	-32.10	-28.14	-	-62.90
		Np	-	-	-	-	-	-72.61
		Pu	-48.54	-48.00	-38.33	-49.88	-49.90	-73.06
		Am	-56.81	-57.15	-56.75	-57.56	-57.18	-48.59
		Cm	-47.49	-47.31	-47.44	-48.46	-48.14	-51.08
		Bk	-59.10	-57.22	-60.92	-61.45	-60.58	-61.61
		Cf	-66.17	-65.75	-66.63	-68.17	-	-66.97
		Es	-68.71	-68.56	-69.18	-70.70	-	-109.17
		Fm	-84.41	-84.67	-84.16	-83.95	-83.26	-120.18
		Md	-108.88	-104.72	-110.90	-110.63	-110.80	-114.51
		No	-128.89	-129.59	-131.39	-131.38	-113.84	-151.98
		Lr	-69.45	-69.31	-69.02	-70.73	-70.42	-72.56
	nd	Th	-17.15	-16.62	-17.77	-15.70	-18.36	-27.39
		Pa	-16.49	-14.42	-16.76	-16.20	-17.30	-46.38
		U	-33.09	-33.07	-	-34.19	-26.46	-62.90
		Np	-	-	-	-	-	-72.61
		Pu	-52.21	-51.88	-51.92	-52.81	-52.04	-73.06
		Am	-61.62	-61.97	-62.20	-62.47	-62.11	-48.59
		Cm	-47.92	-47.85	-48.20	-48.68	-48.35	-51.08
		Bk	-60.46	-	-61.20	-	-62.52	-61.61
		Cf	-71.41	-71.22	-69.25	-72.82	-	-66.97
		Es	-67.99	-71.68	-69.92	-	-69.00	-109.17
		Fm	-84.29	-82.63	-83.70	-	-83.36	-120.18
		Md	-109.38	-105.82	-107.61	-93.73	-93.55	-114.51
		No	-129.90	-130.80	-132.47	-131.76	-131.25	-151.98
		Lr	-68.13	-67.56	-69.29	-70.39	-70.45	-72.56

Table S.16: $\Delta(\Delta G)_{rxn}$ for $[An(NO_3)]^{2+}$ normalized to Ac calculated with M11 with the Stuttgart RSC 1997 ECP and basis set including (ad) and excluding (nd) the most diffuse basis functions for An, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O. $\Delta(\Delta G)_{rxn}$ calculated with CCSD(T)-FC1 includes the cc-pV∞Z-X2C basis set for An, and the cc-pV∞Z-DK basis set for N and O is included as reference (indicated as CCSD(T)/CBS). Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn} = \Delta G_{rxn,An} - \Delta G_{rxn,Ac}$, as indicated in Eq. 5.a; with An = Th - Lr].

			6-31G*	cc-pVDZ	6-311++G**	cc-pVTZ	cc-pVQZ	CCSD(T)/CBS
M11	ad	Th	-19.27	-18.15	-18.25	-18.14	-19.26	-27.39
		Pa	-13.82	-11.20	-3.69	-1.35	-	-46.38
		U	-35.19	-	-33.03	-36.20	-33.12	-62.90
		Np	-	-	-	-	-	-72.61
		Pu	-49.93	-53.91	-49.62	-50.72	-	-73.06
		Am	-	-	-	-	-	-48.59
		Cm	-46.09	-46.22	-45.70	-46.60	-46.29	-51.08
		Bk	-50.23	-51.36	-45.96	-53.23	-52.98	-61.61
		Cf	-	-	-38.89	-58.00	-	-66.97
		Es	-72.10	-68.82	-68.42	-79.50	-70.88	-109.17
		Fm	-	-	-	-	-	-120.18
		Md	-110.73	-103.39	-	-113.67	-107.36	-114.51
		No	-141.13	-141.42	-131.51	-131.23	-143.06	-151.98
		Lr	-71.24	-71.24	-70.20	-71.90	-71.52	-72.56
	nd	Th	-18.09	-16.62	-18.22	-	-19.01	-27.39
		Pa	-14.56	-14.38	-9.98	-26.30	-16.53	-46.38
		U	-34.78	-35.54	-31.81	-35.98	-31.23	-62.90
		Np	-	-	-	-	-	-72.61
		Pu	-50.01	-50.54	-39.80	-42.57	-	-73.06
		Am	-	-	-	-	-	-48.59
		Cm	-46.62	-46.94	-46.47	-46.87	-46.51	-51.08
		Bk	-	-50.46	-50.54	-51.00	-50.63	-61.61
		Cf	-52.10	-56.30	-52.32	-59.89	-	-66.97
		Es	-	-	-	-60.17	-67.33	-109.17
		Fm	-	-	-	-	-	-120.18
		Md	-111.93	-112.66	-114.62	-114.14	-102.81	-114.51
		No	-129.99	-130.76	-132.61	-131.49	-143.31	-151.98
		Lr	-69.82	-69.26	-70.39	-71.59	-71.57	-72.56

Table S.17: Difference between predicted Gibbs free energy of reaction for $[An(NO_3)]^{2+}$ [with An = Ac to Lr] amongst contiguous actinides calculated with LDA, TPSS, B3LYP, PBE0, B972, M06 and M11, the Stuttgart RSC 1997 ECP and associated basis set including all diffuse functions (ad) and excluding the most diffuse functions in the An basis set, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O with respect to the predicted energy with CCSD(T)-FC1/cc-pV∞Z-X2C:cc-pV∞Z-DK [CCSD(T)/CBS]. Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn\ An1/An2} = \Delta G_{rxn,An2} - \Delta G_{rxn,An1}$, as indicated in Eq. 5.a; with An = Ac - Lr.]

	An basis set	Functional	6-31G*	cc-pVDZ	6-311++G**	cc-pVTZ	cc-pVQZ
Ac/Th	ad	LDA	-	-	-	-	-
		TPSS	3.74	4.05	-	3.78	3.86
		B3LYP	6.39	4.13	5.81	4.61	4.13
		PBE0	3.85	4.20	5.15	3.81	3.89
		B972	6.46	6.74	7.64	6.55	6.75
		M06	6.33	6.62	7.13	6.21	6.52
		M11	8.12	9.24	9.14	9.25	8.13
	nd	LDA	-	-	-	-	-
		TPSS	3.19	3.84	-	2.53	2.31
		B3LYP	5.23	5.92	5.04	4.62	4.77
		PBE0	5.02	-	4.94	4.32	4.10
		B972	7.62	8.27	7.33	7.00	6.90
		M06	10.23	10.77	9.62	11.69	9.03
		M11	9.30	10.77	9.16	-	8.38
Th/Pa	ad	LDA	-	-	-	-	-
		TPSS	27.40	24.93	-	-	-
		B3LYP	20.86	23.19	20.58	21.65	21.59
		PBE0	24.99	25.23	23.25	23.97	22.85
		B972	22.65	22.95	20.16	20.52	19.48
		M06	24.24	23.55	20.92	24.46	21.40
		M11	24.45	25.95	33.56	35.78	-
	nd	LDA	-	-	-	-	-
		TPSS	-	-	-	-	24.24
		B3LYP	22.53	24.41	22.55	22.78	22.12
		PBE0	24.77		23.45	25.00	23.53
		B972	22.88	22.64	21.31	21.17	20.99
		M06	19.66	21.19	20.00	18.50	20.06
		M11	22.52	21.24	27.24	-	21.48
Pa/U	ad	LDA	-5.00	-	-4.26	-4.47	-3.49
		TPSS	-19.07	-14.37	-	-	-
		B3LYP	-2.12	-1.51	-1.22	-2.09	-1.57
		PBE0	-2.35	-2.29	-1.59	-2.24	-1.22
		B972	-2.47	-2.42	-0.84	-1.41	-0.49
		M06	0.07	1.65	2.74	4.10	-
		M11	-4.86	-	-12.83	-18.33	-
	nd	LDA	-5.92	-	-5.89	-5.04	-3.08
		TPSS	-	-	-2.85	-	-2.48
		B3LYP	-2.49	-4.79	-2.37	-2.40	-1.91
		PBE0	-2.02	-4.06	-2.64	-3.97	-0.22
		B972	-4.73	-4.78	-2.90	-2.89	-2.47
		M06	-0.09	-2.13	-	-1.48	7.35
		M11	-3.71	-4.65	-5.31	6.84	1.82

Table S.18: Difference between predicted Gibbs free energy of reaction for $[An(NO_3)]^{2+}$ [with An = Ac to Lr] amongst contiguous actinides calculated with LDA, TPSS, B3LYP, PBE0, B972, M06 and M11, the Stuttgart RSC 1997 ECP and associated basis set including all diffuse functions (ad) and excluding the most diffuse functions in the An basis set, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O with respect to the predicted energy with CCSD(T)-FC1/cc-pV∞Z-X2C:cc-pV∞Z-DK [CCSD(T)/CBS]. Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn\ An1/An2} = \Delta G_{rxn,An2} - \Delta G_{rxn,An1}$, as indicated in Eq. 5.a; with An = Ac - Lr.]

	An basis set	Functional	6-31G*	cc-pVDZ	6-311++G**	cc-pVTZ	cc-pVQZ
U/Np	ad	LDA	0.44	-	0.46	0.07	0.51
		TPSS	17.01	13.43	-	-	15.52
		B3LYP	-6.05	-7.62	-6.14	-7.65	-7.65
		PBE0	0.43	2.37	0.56	0.38	0.51
		B972	1.07	1.09	1.07	2.10	2.05
		M06	-	-	-	-	9.72
		M11	-	-	-	-	-
	nd	LDA	-	-	0.82	0.70	0.14
		TPSS	-	-	2.20	3.02	2.27
		B3LYP	3.07	4.61	3.38	4.69	4.55
		PBE0	0.93	0.92	1.06	0.96	4.72
		B972	1.04	2.11	2.20	2.20	1.10
		M06	-	-	-	-	-
		M11	-	-	-	-	-
Np/Pu	ad	LDA	-12.38	-12.02	-11.30	-11.61	-11.23
		TPSS	-14.63	-13.33	-	-	-
		B3LYP	-6.67	-9.24	-7.56	-7.04	-5.69
		PBE0	-5.46	-10.81	-8.71	-8.52	-8.22
		B972	-9.08	-9.46	1.79	-8.56	-8.24
		M06	-	-	-	-	-
		M11	-	-	-	-	-
	nd	LDA	-	-11.57	-11.33	-11.71	-11.32
		TPSS	-	-	-10.13	-10.96	-9.90
		B3LYP	-10.43	-12.18	-10.06	-11.58	1.27
		PBE0	-9.07	-8.05	3.50	-7.52	-4.03
		B972	-8.76	-9.31	-9.52	-9.59	-6.93
		M06	-	-	-	-	-
		M11	-	-	-	-	-
Pu/Am	ad	LDA	-36.46	-	-	-36.21	-35.99
		TPSS	-	-	-	-	-24.47
		B3LYP	-27.09	-23.95	-25.42	-24.07	-24.89
		PBE0	-34.32	-31.84	-30.82	-30.41	-30.25
		B972	-33.80	-34.42	-44.48	-34.45	-34.11
		M06	-32.74	-33.63	-42.89	-32.15	-31.75
		M11	-	-	-	-	-
	nd	LDA	-35.93	-37.13	-37.05	-36.09	-35.94
		TPSS	-34.09	-34.56	-	-34.24	-34.08
		B3LYP	-32.69	-33.09	-33.28	-32.91	-45.10
		PBE0	-36.07	-35.75	-46.84	-35.50	-44.25
		B972	-33.25	-34.52	-33.97	-33.33	-34.38
		M06	-33.89	-34.56	-34.75	-34.14	-34.54
		M11	-	-	-	-	-

Table S.19: Difference between predicted Gibbs free energy of reaction for $[An(NO_3)]^{2+}$ [with An = Ac to Lr] amongst contiguous actinides calculated with LDA, TPSS, B3LYP, PBE0, B972, M06 and M11, the Stuttgart RSC 1997 ECP and associated basis set including all diffuse functions (ad) and excluding the most diffuse functions in the An basis set, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O with respect to the predicted energy with CCSD(T)-FC1/cc-pV∞Z-X2C:cc-pV∞Z-DK [CCSD(T)/CBS]. Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn\ An1/An2} = \Delta G_{rxn,An2} - \Delta G_{rxn,An1}$, as indicated in Eq. 5.a; with An = Ac - Lr.]

	An basis set	Functional	6-31G*	cc-pVDZ	6-311++G**	cc-pVTZ	cc-pVQZ
Am/Cm	ad	LDA	17.24	-	-	16.88	16.65
		TPSS	-	-	-	-	-
		B3LYP	11.90	12.45	11.73	11.58	11.38
		PBE0	12.31	12.85	12.18	12.02	11.86
		B972	14.76	15.40	14.83	14.40	14.15
		M06	11.81	12.34	11.80	11.59	11.54
		M11	-	-	-	-	-
	nd	LDA	16.85	17.31	17.17	16.80	16.65
		TPSS	14.20	14.67		14.17	14.01
		B3LYP	11.49	11.91	11.69	11.49	11.35
		PBE0	15.45	15.87	15.65	15.45	15.36
		B972	14.39	14.92	14.79	14.31	14.12
		M06	16.20	16.61	16.49	16.28	16.26
		M11	-	-	-	-	-
Cm/Bk	ad	LDA	-0.61	-0.37	-1.22	-1.41	-1.70
		TPSS	-1.76	-0.28	-	-2.67	-2.90
		B3LYP	-0.79	-0.54	-3.07	-1.78	-3.21
		PBE0	-1.14	0.33	-0.45	-2.06	-2.70
		B972	-0.88	-2.42	-1.39	-0.96	-3.71
		M06	-1.07	0.62	-2.95	-2.45	-1.91
		M11	6.39	5.39	10.27	3.90	3.84
	nd	LDA	-0.80	-0.85	-1.26	-1.54	-1.76
		TPSS	-2.26	-3.09	-2.98	-3.61	-2.95
		B3LYP	-1.39	-1.28	-1.46	-1.95	-3.27
		PBE0	-1.64	-0.29	-0.74	-2.17	-2.74
		B972	-1.35	-3.03	-1.69	-3.19	-3.75
		M06	-2.01	-	-2.46	-	-3.64
		M11	-	7.02	6.46	6.40	6.42
Bk/Cf	ad	LDA	-2.95	-3.04	-3.12	-2.96	-2.91
		TPSS	-1.99	-	-	-1.96	-1.92
		B3LYP	2.90	2.53	4.42	2.93	4.08
		PBE0	1.62	-0.09	4.44	1.11	1.92
		B972	-3.68	-3.43	-6.32	-5.99	-4.09
		M06	-1.72	-3.17	-0.36	-1.36	-
		M11	-	-	12.42	0.59	-
	nd	LDA	-3.34	-3.35	-3.20	-2.99	-2.90
		TPSS	-1.53	-0.80	-1.40	0.02	-1.47
		B3LYP	2.03	0.07	-0.55	0.44	0.99
		PBE0	1.17	-0.09	4.46	1.66	1.93
		B972	0.13	1.85	1.42	1.42	1.19
		M06	-5.59	-	-2.69	-	-
		M11	-	-0.49	3.58	-3.53	-

Table S.20: Difference between predicted Gibbs free energy of reaction for $[An(NO_3)]^{2+}$ [with An = Ac to Lr] amongst contiguous actinides calculated with LDA, TPSS, B3LYP, PBE0, B972, M06 and M11, the Stuttgart RSC 1997 ECP and associated basis set including all diffuse functions (ad) and excluding the most diffuse functions in the An basis set, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O with respect to the predicted energy with CCSD(T)-FC1/cc-pV∞Z-X2C:cc-pV∞Z-DK [CCSD(T)/CBS]. Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn\ An1/An2} = \Delta G_{rxn,An2} - \Delta G_{rxn,An1}$, as indicated in Eq. 5.a; with An = Ac - Lr.]

	An basis set	Functional	6-31G*	cc-pVDZ	6-311++G**	cc-pVTZ	cc-pVQZ
Cf/Es	ad	LDA	33.12	-	32.69	33.01	33.17
		TPSS	24.15	-	-	24.05	24.44
		B3LYP	12.51	13.94	20.02	13.79	20.30
		PBE0	39.07	38.76	34.07	39.51	38.59
		B972	30.14	31.39	33.56	31.62	39.95
		M06	39.66	39.39	39.66	39.67	42.20
		M11	-	-	12.67	20.71	-
	nd	LDA	32.48	-	31.99	32.76	32.96
		TPSS	25.28	-	28.16	-	23.77
		B3LYP	24.50	25.70	28.86	25.52	26.32
		PBE0	16.03	16.96	11.10	15.42	18.27
		B972	26.52	26.40	32.07	26.34	34.68
		M06	45.63	41.73	41.53	-	-
		M11	-	-	-	41.92	-
Es/Fm	ad	LDA	-9.79	-	-8.47	-8.06	-7.50
		TPSS	-	-		14.81	16.44
		B3LYP	-	-	21.07	29.15	23.86
		PBE0	7.32	6.61	4.86	6.07	8.15
		B972	-	16.07	6.71	9.39	3.52
		M06	-4.68	-5.10	-3.97	-2.23	-
		M11	-	-	-	-	-
	nd	LDA	-6.25	-	-6.14	-6.10	-5.82
		TPSS	14.15	-	-	-	16.72
		B3LYP	-	18.82	15.84	20.38	21.05
		PBE0	29.61	27.53	28.31	30.11	28.66
		B972	9.01	-	0.94	10.23	3.70
		M06	-5.29	0.07	-2.77	-	-3.34
		M11	-	-	-	-	-
Fm/Md	ad	LDA	-13.74	-11.83	-15.79	-15.66	-
		TPSS	-	-5.67	-	-	-
		B3LYP	-	-	-29.87	-30.09	-30.52
		PBE0	-30.94	-30.16	-29.60	-29.60	-30.11
		B972	-	-38.01	-30.95	-30.66	-31.45
		M06	-30.14	-25.71	-32.41	-32.36	-33.21
		M11	-	-	-	-	-
	nd	LDA	-15.75	-16.33	-16.57	-	-17.11
		TPSS	-21.15	-	-	-30.13	-
		B3LYP	-	-24.59	-30.45	-30.45	-30.77
		PBE0	-30.62	-30.31	-31.14	-31.08	-31.17
		B972	-30.52	-	-	-31.61	-32.10
		M06	-30.76	-28.86	-29.57	-	-15.87
		M11	-	-	-	-	-

Table S.21: Difference between predicted Gibbs free energy of reaction for $[An(NO_3)]^{2+}$ [with An = Ac to Lr] amongst contiguous actinides calculated with LDA, TPSS, B3LYP, PBE0, B972, M06 and M11, the Stuttgart RSC 1997 ECP and associated basis set including all diffuse functions (ad) and excluding the most diffuse functions in the An basis set, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ for N and O with respect to the predicted energy with CCSD(T)-FC1/cc-pV∞Z-X2C:cc-pV∞Z-DK [CCSD(T)/CBS]. Values reported in kcal mol^{-1}. [$\Delta(\Delta G)_{rxn\ An1/An2} = \Delta G_{rxn,An2} - \Delta G_{rxn,An1}$, as indicated in Eq. 5.a; with An = Ac - Lr.]

	An basis set	**Functional**	**6-31G***	**cc-pVDZ**	**6-311++G****	**cc-pVTZ**	**cc-pVQZ**
No/Lr	**ad**	LDA	-18.18	-18.06	-15.74	-17.37	-17.48
		TPSS	-23.50	-23.07	-	-22.83	-
		B3LYP	-24.77	-24.45	-21.50	-23.45	-23.73
		PBE0	-23.25	-22.74	-20.44	-22.62	-23.27
		B972	-11.36	-10.78	-8.53	-10.74	-11.37
		M06	-19.97	-19.14	-17.05	-18.77	-36.01
		M11	-9.53	-9.23	-18.11	-20.09	-7.88
	nd	LDA	-20.22	-	-	-	-21.81
		TPSS	-	-	-20.08	-	-23.01
		B3LYP	-22.12	-20.98	-20.63	-22.55	-23.41
		PBE0	-20.96	-19.73	-19.60	-21.84	-22.99
		B972	-9.16	-7.86	-7.73	-9.99	-11.11
		M06	-17.65	-16.17	-16.24	-18.04	-18.63
		M11	-19.24	-17.92	-17.19	-19.52	-7.68

Table S.22: Percentage contribution of enthalpy and entropy terms to the Gibbs free energy of reaction in $[An(NO_3)^{2+}$ with An = Ac to Np] calculated with LDA, TPSS, B3LYP, PBE0, B972, M06, and M11, the Stuttgart RSC 1997 ECP and associated basis set including all basis functions for the actinides, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ basis set for the non-actinide atoms. [Calculated as ΔH/ΔG x 100 and T.ΔS x 100]

Functional	Non-An basis set	Ac		Th		Pa		U		Np	
		ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$
LDA	6-31G*	99.12%	0.88%	-	-	99.25%	0.75%	99.28%	0.72%	99.30%	0.70%
	cc-pVDZ	99.12%	0.88%	-	-	99.25%	0.75%	-	-	99.30%	0.70%
	6-311++G**	99.05%	0.95%	-	-	99.20%	0.80%	99.23%	0.77%	99.25%	0.75%
	cc-pVTZ	99.08%	0.92%	-	-	99.22%	0.78%	99.26%	0.74%	99.28%	0.72%
	cc-pVQZ	99.07%	0.93%	-	-	99.22%	0.78%	99.25%	0.75%	99.27%	0.73%
TPSS	6-31G*	99.07%	0.93%	99.12%	0.88%	99.10%	0.90%	99.17%	0.83%	99.17%	0.83%
	cc-pVDZ	99.07%	0.93%	99.11%	0.89%	99.12%	0.88%	99.17%	0.83%	99.17%	0.83%
	cc-pVTZ	99.04%	0.96%	99.09%	0.91%	-	-	-	-	-	-
	cc-pVQZ	99.04%	0.96%	99.09%	0.91%	-	-	99.15%	0.85%	99.17%	0.83%
B3LYP	6-31G*	99.07%	0.93%	99.11%	0.89%	99.13%	0.87%	99.15%	0.85%	99.20%	0.80%
	cc-pVDZ	99.07%	0.93%	99.11%	0.89%	99.13%	0.87%	99.15%	0.85%	99.19%	0.81%
	6-311++G**	99.00%	1.00%	99.05%	0.95%	99.07%	0.93%	99.09%	0.91%	99.13%	0.87%
	cc-pVTZ	99.03%	0.97%	99.09%	0.91%	99.09%	0.91%	99.13%	0.87%	99.17%	0.83%
	cc-pVQZ	99.03%	0.97%	99.09%	0.91%	99.08%	0.92%	99.13%	0.87%	99.17%	0.83%
PBE0	6-31G*	99.08%	0.92%	99.13%	0.87%	99.12%	0.88%	99.17%	0.83%	99.20%	0.80%
	cc-pVDZ	99.08%	0.92%	99.13%	0.87%	99.12%	0.88%	99.16%	0.84%	99.18%	0.82%
	6-311++G**	99.03%	0.97%	99.08%	0.92%	99.08%	0.92%	99.12%	0.88%	99.16%	0.84%
	cc-pVTZ	99.06%	0.94%	99.12%	0.88%	99.11%	0.89%	99.15%	0.85%	99.18%	0.82%
	cc-pVQZ	99.05%	0.95%	99.12%	0.88%	99.10%	0.90%	99.15%	0.85%	99.18%	0.82%
B972	6-31G*	99.07%	0.93%	99.11%	0.89%	99.12%	0.88%	99.16%	0.84%	99.19%	0.81%
	cc-pVDZ	99.07%	0.93%	99.10%	0.90%	99.12%	0.88%	99.15%	0.85%	99.18%	0.82%
	6-311++G**	99.02%	0.98%	99.05%	0.95%	99.06%	0.94%	99.10%	0.90%	99.14%	0.86%
	cc-pVTZ	99.04%	0.96%	99.09%	0.91%	99.10%	0.90%	99.14%	0.86%	99.17%	0.83%
	cc-pVQZ	99.04%	0.96%	99.09%	0.91%	99.08%	0.92%	99.13%	0.87%	99.17%	0.83%
M06	6-31G*	99.06%	0.94%	99.10%	0.90%	99.10%	0.90%	99.14%	0.86%	-	-
	cc-pVDZ	99.06%	0.94%	99.10%	0.90%	99.10%	0.90%	99.13%	0.87%	-	-
	6-311++G**	99.00%	1.00%	99.05%	0.95%	99.07%	0.93%	99.08%	0.92%	-	-
	cc-pVTZ	99.04%	0.96%	99.09%	0.91%	99.08%	0.92%	99.11%	0.89%	-	-
	cc-pVQZ	99.04%	0.96%	99.09%	0.91%	99.08%	0.92%	-	-	-	-
M11	6-31G*	99.07%	0.93%	99.12%	0.88%	99.12%	0.88%	99.15%	0.85%	-	-
	cc-pVDZ	99.07%	0.93%	99.11%	0.89%	99.10%	0.90%	-	-	-	-
	6-311++G**	99.01%	0.99%	99.06%	0.94%	99.03%	0.97%	99.09%	0.91%	-	-
	cc-pVTZ	99.03%	0.97%	99.07%	0.93%	99.04%	0.96%	99.11%	0.89%	-	-
	cc-pVQZ	99.03%	0.97%	99.09%	0.91%	-	-	99.11%	0.89%	-	-

Table S.23: Percentage contribution of enthalpy and entropy terms to the Gibbs free energy of reaction in $[An(NO_3)^{2+}$ with An = Pu to Cf] calculated with LDA, TPSS, B3LYP, PBE0, B972, M06, and M11, the Stuttgart RSC 1997 ECP and associated basis set including all basis functions for the actinides, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ basis set for the non-actinide atoms. [Calculated as ΔH/ΔG x 100 and T.ΔS x 100]

Functional	Non-An basis set	Pu		Am		Cm		Bk		Cf	
		ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$
LDA	6-31G*	99.32%	0.68%	99.21%	0.79%	99.22%	0.78%	99.31%	0.69%	99.33%	0.67%
	cc-pVDZ	99.31%	0.69%	-	-	99.22%	0.78%	99.32%	0.68%	99.33%	0.67%
	6-311++G**	99.25%	0.75%	-	-	99.16%	0.84%	99.28%	0.72%	99.26%	0.74%
	cc-pVTZ	99.29%	0.71%	99.19%	0.81%	99.20%	0.80%	99.30%	0.70%	99.31%	0.69%
	cc-pVQZ	99.28%	0.72%	99.18%	0.82%	99.19%	0.81%	99.30%	0.70%	99.31%	0.69%
TPSS	6-31G*	99.21%	0.79%	-	-	99.18%	0.82%	99.20%	0.80%	99.20%	0.80%
	cc-pVDZ	99.21%	0.79%	-	-	99.18%	0.82%	99.19%	0.81%	-	-
	cc-pVTZ	99.19%	0.81%	-	-	99.16%	0.84%	99.19%	0.81%	99.19%	0.81%
	cc-pVQZ	-	-	-	-	99.16%	0.84%	99.19%	0.81%	99.19%	0.81%
B3LYP	6-31G*	99.20%	0.80%	99.20%	0.80%	99.18%	0.82%	99.20%	0.80%	99.20%	0.80%
	cc-pVDZ	99.22%	0.78%	99.20%	0.80%	99.18%	0.82%	99.20%	0.80%	99.21%	0.79%
	6-311++G**	99.15%	0.85%	99.15%	0.85%	99.12%	0.88%	99.16%	0.84%	99.15%	0.85%
	cc-pVTZ	99.18%	0.82%	99.18%	0.82%	99.15%	0.85%	99.18%	0.82%	99.18%	0.82%
	cc-pVQZ	99.18%	0.82%	99.18%	0.82%	99.15%	0.85%	99.18%	0.82%	99.18%	0.82%
PBE0	6-31G*	99.20%	0.80%	99.22%	0.78%	99.19%	0.81%	99.22%	0.78%	99.22%	0.78%
	cc-pVDZ	99.21%	0.79%	99.22%	0.78%	99.19%	0.81%	99.21%	0.79%	99.23%	0.77%
	6-311++G**	99.18%	0.82%	99.17%	0.83%	99.14%	0.86%	99.17%	0.83%	99.18%	0.82%
	cc-pVTZ	99.20%	0.80%	99.20%	0.80%	99.17%	0.83%	99.20%	0.80%	99.22%	0.78%
	cc-pVQZ	99.20%	0.80%	99.20%	0.80%	99.17%	0.83%	99.20%	0.80%	99.20%	0.80%
B972	6-31G*	99.21%	0.79%	99.19%	0.81%	99.18%	0.82%	99.20%	0.80%	99.21%	0.79%
	cc-pVDZ	99.21%	0.79%	99.19%	0.81%	99.17%	0.83%	99.20%	0.80%	99.21%	0.79%
	6-311++G**	99.05%	0.95%	99.14%	0.86%	99.13%	0.87%	99.16%	0.84%	99.18%	0.82%
	cc-pVTZ	99.17%	0.83%	99.18%	0.82%	99.16%	0.84%	99.18%	0.82%	99.20%	0.80%
	cc-pVQZ	99.17%	0.83%	99.18%	0.82%	99.16%	0.84%	99.19%	0.81%	99.20%	0.80%
M06	6-31G*	99.18%	0.82%	99.19%	0.81%	99.17%	0.83%	99.19%	0.81%	99.21%	0.79%
	cc-pVDZ	99.18%	0.82%	99.19%	0.81%	99.16%	0.84%	99.17%	0.83%	99.21%	0.79%
	6-311++G**	99.10%	0.90%	99.14%	0.86%	99.11%	0.89%	99.15%	0.85%	99.17%	0.83%
	cc-pVTZ	99.17%	0.83%	99.18%	0.82%	99.15%	0.85%	99.19%	0.81%	99.20%	0.80%
	cc-pVQZ	99.17%	0.83%	99.18%	0.82%	99.15%	0.85%	99.18%	0.82%	-	-
M11	6-31G*	99.20%	0.80%	-	-	99.18%	0.82%	99.19%	0.81%	-	-
	cc-pVDZ	99.21%	0.79%	-	-	99.18%	0.82%	99.20%	0.80%	-	-
	6-311++G**	99.16%	0.84%	-	-	99.13%	0.87%	99.13%	0.87%	99.09%	0.91%
	cc-pVTZ	99.18%	0.82%	-	-	99.16%	0.84%	99.17%	0.83%	99.19%	0.81%
	cc-pVQZ	-	-	-	-	99.15%	0.85%	99.17%	0.83%	-	-

Table S.24: Percentage contribution of enthalpy and entropy terms to the Gibbs free energy of reaction in $[An(NO_3)^{2+}$ with An = Es to Lr] calculated with LDA, TPSS, B3LYP, PBE0, B972, M06, and M11, the Stuttgart RSC 1997 ECP and associated basis set including all basis functions for the actinides, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ basis set for the non-actinide atoms. [Calculated as ΔH/ΔG x 100 and T.ΔS x 100]

Functional	Non-An basis set	Es		Fm		Md		No		Lr	
		ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$
LDA	6-31G*	99.33%	0.67%	99.32%	0.68%	99.30%	0.70%	99.21%	0.79%	99.28%	0.72%
	cc-pVDZ	-	-	99.32%	0.68%	99.30%	0.70%	99.21%	0.79%	99.28%	0.72%
	6-311++G**	99.29%	0.71%	99.28%	0.72%	99.26%	0.74%	99.15%	0.85%	99.23%	0.77%
	cc-pVTZ	99.31%	0.69%	99.31%	0.69%	99.28%	0.72%	99.18%	0.82%	99.26%	0.74%
	cc-pVQZ	99.30%	0.70%	99.30%	0.70%	-	-	99.18%	0.82%	99.25%	0.75%
TPSS	6-31G*	99.24%	0.76%	-	-	99.06%	0.94%	99.07%	0.93%	99.23%	0.77%
	cc-pVDZ	99.19%	0.81%	-	-	-	-	99.07%	0.93%	99.23%	0.77%
	cc-pVTZ	99.23%	0.77%	99.21%	0.79%	-	-	99.05%	0.95%	99.22%	0.78%
	cc-pVQZ	99.23%	0.77%	99.21%	0.79%	-	-	-	-	99.22%	0.78%
B3LYP	6-31G*	99.26%	0.74%	-	-	99.02%	0.98%	99.07%	0.93%	99.23%	0.77%
	cc-pVDZ	99.25%	0.75%	-	-	-	-	99.07%	0.93%	99.24%	0.76%
	6-311++G**	99.16%	0.84%	98.89%	1.11%	98.91%	1.09%	98.99%	1.01%	99.19%	0.81%
	cc-pVTZ	99.23%	0.77%	98.91%	1.09%	98.97%	1.03%	99.03%	0.97%	99.21%	0.79%
	cc-pVQZ	99.19%	0.81%	98.92%	1.08%	98.96%	1.04%	99.02%	0.98%	99.21%	0.79%
PBE0	6-31G*	99.24%	0.76%	98.94%	1.06%	99.02%	0.98%	99.08%	0.92%	99.24%	0.76%
	cc-pVDZ	99.24%	0.76%	98.96%	1.04%	99.02%	0.98%	99.08%	0.92%	99.24%	0.76%
	6-311++G**	99.20%	0.80%	98.93%	1.07%	98.94%	1.06%	99.01%	0.99%	99.21%	0.79%
	cc-pVTZ	99.22%	0.78%	98.96%	1.04%	98.99%	1.01%	99.06%	0.94%	99.23%	0.77%
	cc-pVQZ	99.22%	0.78%	98.95%	1.05%	98.99%	1.01%	99.06%	0.94%	99.23%	0.77%
B972	6-31G*	99.24%	0.76%	-	-	99.03%	0.97%	99.06%	0.94%	99.23%	0.77%
	cc-pVDZ	99.24%	0.76%	98.89%	1.11%	99.01%	0.99%	99.05%	0.95%	99.23%	0.77%
	6-311++G**	99.19%	0.81%	98.89%	1.11%	98.94%	1.06%	98.99%	1.01%	99.20%	0.80%
	cc-pVTZ	99.23%	0.77%	98.89%	1.11%	98.98%	1.02%	99.03%	0.97%	99.22%	0.78%
	cc-pVQZ	99.17%	0.83%	98.93%	1.07%	98.96%	1.04%	99.03%	0.97%	99.21%	0.79%
M06	6-31G*	99.21%	0.79%	98.98%	1.02%	99.02%	0.98%	99.07%	0.93%	99.22%	0.78%
	cc-pVDZ	99.21%	0.79%	98.96%	1.04%	99.01%	0.99%	99.07%	0.93%	99.22%	0.78%
	6-311++G**	99.17%	0.83%	98.91%	1.09%	98.96%	1.04%	99.01%	0.99%	99.19%	0.81%
	cc-pVTZ	99.19%	0.81%	98.95%	1.05%	99.00%	1.00%	99.05%	0.95%	99.21%	0.79%
	cc-pVQZ	-	-	98.95%	1.05%	99.00%	1.00%	99.03%	0.97%	99.21%	0.79%
M11	6-31G*	99.23%	0.77%	-	-	99.06%	0.94%	99.12%	0.88%	99.23%	0.77%
	cc-pVDZ	99.22%	0.78%	-	-	99.06%	0.94%	99.13%	0.87%	99.23%	0.77%
	6-311++G**	99.17%	0.83%	-	-	-	-	99.05%	0.95%	99.19%	0.81%
	cc-pVTZ	99.19%	0.81%	-	-	99.01%	0.99%	99.07%	0.93%	99.20%	0.80%
	cc-pVQZ	99.19%	0.81%	-	-	99.01%	0.99%	99.07%	0.93%	99.20%	0.80%

Table S.25: Percentage contribution of enthalpy and entropy terms to the Gibbs free energy of reaction in $[An(NO_3)^{2+}$ with An = Ac to Np] calculated with LDA, TPSS, B3LYP, PBE0, B972, M06, and M11, the Stuttgart RSC 1997 ECP and associated basis set excluding the most diffuse basis functions for the actinides, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ basis set for the non-actinide atoms. [Calculated as ΔH/ΔG x 100 and T.ΔS x 100]

Functional	Non-An basis set	Ac		Th		Pa		U		Np	
		ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$
LDA	6-31G*	99.12%	0.88%	-	-	99.25%	0.75%	99.27%	0.73%	-	-
	cc-pVDZ	99.12%	0.88%	-	-	99.25%	0.75%	-	-	99.30%	0.70%
	6-311++G**	99.05%	0.95%	-	-	99.20%	0.80%	99.22%	0.78%	99.25%	0.75%
	cc-pVTZ	99.08%	0.92%	-	-	99.23%	0.77%	99.26%	0.74%	99.28%	0.72%
	cc-pVQZ	99.07%	0.93%	-	-	99.22%	0.78%	99.26%	0.74%	99.27%	0.73%
TPSS	6-31G*	99.07%	0.93%	99.12%	0.88%	-	-	-	-	-	-
	cc-pVDZ	99.07%	0.93%	99.11%	0.89%	-	-	-	-	-	-
	6-311++G**	99.01%	0.99%	-	-	99.06%	0.94%	99.10%	0.90%	99.13%	0.87%
	cc-pVTZ	99.04%	0.96%	99.10%	0.90%	-	-	99.13%	0.87%	99.16%	0.84%
	cc-pVQZ	99.04%	0.96%	99.10%	0.90%	99.08%	0.92%	99.13%	0.87%	99.16%	0.84%
B3LYP	6-31G*	99.07%	0.93%	99.12%	0.88%	99.12%	0.88%	99.15%	0.85%	99.18%	0.82%
	cc-pVDZ	99.07%	0.93%	99.11%	0.89%	99.12%	0.88%	99.15%	0.85%	99.17%	0.83%
	6-311++G**	99.00%	1.00%	99.05%	0.95%	99.06%	0.94%	99.09%	0.91%	99.11%	0.89%
	cc-pVTZ	99.03%	0.97%	99.09%	0.91%	99.09%	0.91%	99.13%	0.87%	99.15%	0.85%
	cc-pVQZ	99.03%	0.97%	99.09%	0.91%	99.08%	0.92%	99.12%	0.88%	99.14%	0.86%
PBE0	6-31G*	99.08%	0.92%	99.14%	0.86%	99.12%	0.88%	99.17%	0.83%	99.18%	0.82%
	cc-pVDZ	99.08%	0.92%	-	-	99.12%	0.88%	99.16%	0.84%	99.19%	0.81%
	6-311++G**	99.03%	0.97%	99.08%	0.92%	99.09%	0.91%	99.12%	0.88%	99.15%	0.85%
	cc-pVTZ	99.06%	0.94%	99.12%	0.88%	99.09%	0.91%	99.15%	0.85%	99.19%	0.81%
	cc-pVQZ	99.05%	0.95%	99.12%	0.88%	99.11%	0.89%	99.15%	0.85%	99.12%	0.88%
B972	6-31G*	99.07%	0.93%	99.11%	0.89%	99.12%	0.88%	99.15%	0.85%	99.19%	0.81%
	cc-pVDZ	99.07%	0.93%	99.10%	0.90%	99.12%	0.88%	99.15%	0.85%	99.18%	0.82%
	6-311++G**	99.01%	0.99%	99.05%	0.95%	99.06%	0.94%	99.10%	0.90%	99.13%	0.87%
	cc-pVTZ	99.04%	0.96%	99.09%	0.91%	99.10%	0.90%	99.14%	0.86%	99.17%	0.83%
	cc-pVQZ	99.04%	0.96%	99.09%	0.91%	99.08%	0.92%	99.13%	0.87%	99.17%	0.83%
M06	6-31G*	99.06%	0.94%	99.09%	0.91%	99.12%	0.88%	99.14%	0.86%	-	-
	cc-pVDZ	99.06%	0.94%	99.09%	0.91%	99.10%	0.90%	99.14%	0.86%	-	-
	6-311++G**	99.00%	1.00%	99.04%	0.96%	99.07%	0.93%	-	-	-	-
	cc-pVTZ	99.04%	0.96%	99.08%	0.92%	99.10%	0.90%	99.13%	0.87%	-	-
	cc-pVQZ	99.04%	0.96%	99.08%	0.92%	99.08%	0.92%	99.09%	0.91%	-	-
M11	6-31G*	99.07%	0.93%	99.13%	0.87%	99.11%	0.89%	99.16%	0.84%	-	-
	cc-pVDZ	99.07%	0.93%	99.10%	0.90%	99.12%	0.88%	99.15%	0.85%	-	-
	6-311++G**	99.00%	1.00%	99.05%	0.95%	99.04%	0.96%	99.09%	0.91%	-	-
	cc-pVTZ	99.03%	0.97%	-	-	99.11%	0.89%	99.14%	0.86%	-	-
	cc-pVQZ	99.03%	0.97%	99.09%	0.91%	99.06%	0.94%	99.09%	0.91%	-	-

Table S.26: Percentage contribution of enthalpy and entropy terms to the Gibbs free energy of reaction in $[An(NO_3)^{2+}$ with An = Pu to Cf] calculated with LDA, TPSS, B3LYP, PBE0, B972, M06, and M11, the Stuttgart RSC 1997 ECP and associated basis set excluding the most diffuse basis functions for the actinides, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ basis set for the non-actinide atoms. [Calculated as ΔH/ΔG x 100 and T.ΔS x 100]

Functional	Non-An basis set	Pu		Am		Cm		Bk		Cf	
		ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$
LDA	6-31G*	99.31%	0.69%	99.21%	0.79%	99.22%	0.78%	99.32%	0.68%	99.33%	0.67%
	cc-pVDZ	99.31%	0.69%	99.21%	0.79%	99.22%	0.78%	99.32%	0.68%	99.33%	0.67%
	6-311++G**	99.25%	0.75%	99.15%	0.85%	99.16%	0.84%	99.28%	0.72%	99.28%	0.72%
	cc-pVTZ	99.29%	0.71%	99.19%	0.81%	99.20%	0.80%	99.30%	0.70%	99.31%	0.69%
	cc-pVQZ	99.28%	0.72%	99.18%	0.82%	99.19%	0.81%	99.30%	0.70%	99.31%	0.69%
TPSS	6-31G*	99.21%	0.79%	99.19%	0.81%	99.18%	0.82%	99.20%	0.80%	99.20%	0.80%
	cc-pVDZ	99.20%	0.80%	99.19%	0.81%	99.18%	0.82%	99.20%	0.80%	99.20%	0.80%
	6-311++G**	99.16%	0.84%	-	-	99.13%	0.87%	99.16%	0.84%	99.16%	0.84%
	cc-pVTZ	99.19%	0.81%	99.18%	0.82%	99.16%	0.84%	99.19%	0.81%	99.19%	0.81%
	cc-pVQZ	99.19%	0.81%	99.18%	0.82%	99.16%	0.84%	99.19%	0.81%	99.19%	0.81%
B3LYP	6-31G*	99.20%	0.80%	99.20%	0.80%	99.18%	0.82%	99.20%	0.80%	99.20%	0.80%
	cc-pVDZ	99.20%	0.80%	99.20%	0.80%	99.18%	0.82%	99.20%	0.80%	99.20%	0.80%
	6-311++G**	99.15%	0.85%	99.15%	0.85%	99.12%	0.88%	99.15%	0.85%	99.16%	0.84%
	cc-pVTZ	99.18%	0.82%	99.18%	0.82%	99.16%	0.84%	99.18%	0.82%	99.18%	0.82%
	cc-pVQZ	99.12%	0.88%	99.18%	0.82%	99.15%	0.85%	99.18%	0.82%	99.19%	0.81%
PBE0	6-31G*	99.21%	0.79%	99.22%	0.78%	99.19%	0.81%	99.22%	0.78%	99.23%	0.77%
	cc-pVDZ	99.22%	0.78%	99.22%	0.78%	99.19%	0.81%	99.21%	0.79%	99.22%	0.78%
	6-311++G**	99.05%	0.95%	99.18%	0.82%	99.14%	0.86%	99.17%	0.83%	99.18%	0.82%
	cc-pVTZ	99.20%	0.80%	99.21%	0.79%	99.17%	0.83%	99.20%	0.80%	99.21%	0.79%
	cc-pVQZ	99.12%	0.88%	99.21%	0.79%	99.17%	0.83%	99.20%	0.80%	99.21%	0.79%
B972	6-31G*	99.21%	0.79%	99.19%	0.81%	99.18%	0.82%	99.20%	0.80%	99.20%	0.80%
	cc-pVDZ	99.19%	0.81%	99.19%	0.81%	99.17%	0.83%	99.20%	0.80%	99.20%	0.80%
	6-311++G**	99.16%	0.84%	99.14%	0.86%	99.13%	0.87%	99.16%	0.84%	99.15%	0.85%
	cc-pVTZ	99.19%	0.81%	99.18%	0.82%	99.16%	0.84%	99.19%	0.81%	99.19%	0.81%
	cc-pVQZ	99.17%	0.83%	99.18%	0.82%	99.16%	0.84%	99.19%	0.81%	99.19%	0.81%
M06	6-31G*	99.18%	0.82%	99.20%	0.80%	99.17%	0.83%	99.20%	0.80%	99.22%	0.78%
	cc-pVDZ	99.19%	0.81%	99.20%	0.80%	99.16%	0.84%	-	-	99.22%	0.78%
	6-311++G**	99.14%	0.86%	99.15%	0.85%	99.12%	0.88%	99.15%	0.85%	99.12%	0.88%
	cc-pVTZ	99.17%	0.83%	99.19%	0.81%	99.15%	0.85%	-	-	99.21%	0.79%
	cc-pVQZ	99.18%	0.82%	99.19%	0.81%	99.15%	0.85%	99.19%	0.81%	-	-
M11	6-31G*	99.21%	0.79%	-	-	99.18%	0.82%	-	-	99.18%	0.82%
	cc-pVDZ	99.20%	0.80%	-	-	99.17%	0.83%	99.19%	0.81%	99.19%	0.81%
	6-311++G**	99.10%	0.90%	-	-	99.13%	0.87%	99.14%	0.86%	99.13%	0.87%
	cc-pVTZ	99.13%	0.87%	-	-	99.16%	0.84%	99.16%	0.84%	99.18%	0.82%
	cc-pVQZ	-	-	-	-	99.15%	0.85%	99.16%	0.84%	-	-

Table S.27: Percentage contribution of enthalpy and entropy terms to the Gibbs free energy of reaction in $[An(NO_3)^{2+}$ with An = Es to Lr] calculated with LDA, TPSS, B3LYP, PBE0, B972, M06, and M11, the Stuttgart RSC 1997 ECP and associated basis set excluding the most diffuse basis functions for the actinides, and the 6-31G*, cc-pVDZ, 6-311++G**, cc-pVTZ, and cc-pVQZ basis set for the non-actinide atoms. [Calculated as ΔH/ΔG x 100 and T.ΔS x 100]

Functional	Non-An basis set	Es		Fm		Md		No		Lr	
		ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$	ΔH_{rxn}	$-T.\Delta S_{rxn}$
LDA	6-31G*	99.33%	0.67%	99.32%	0.68%	99.30%	0.70%	99.20%	0.80%	99.28%	0.72%
	cc-pVDZ	-	-	99.32%	0.68%	99.30%	0.70%	-	-	99.28%	0.72%
	6-311++G**	99.28%	0.72%	99.27%	0.73%	99.26%	0.74%	-	-	99.23%	0.77%
	cc-pVTZ	99.31%	0.69%	99.30%	0.70%	-	-	-	-	99.26%	0.74%
	cc-pVQZ	99.31%	0.69%	99.31%	0.69%	99.26%	0.74%	99.17%	0.83%	99.25%	0.75%
TPSS	6-31G*	99.25%	0.75%	99.21%	0.79%	99.13%	0.87%	-	-	99.23%	0.77%
	cc-pVDZ	-	-	-	-	99.12%	0.88%	-	-	99.23%	0.77%
	6-311++G**	99.19%	0.81%	-	-	-	-	99.00%	1.00%	99.19%	0.81%
	cc-pVTZ	-	-	99.12%	0.88%	99.11%	0.89%	-	-	-	-
	cc-pVQZ	99.23%	0.77%	99.21%	0.79%	-	-	99.05%	0.95%	99.22%	0.78%
B3LYP	6-31G*	99.25%	0.75%	-	-	98.97%	1.03%	99.07%	0.93%	99.23%	0.77%
	cc-pVDZ	99.25%	0.75%	98.96%	1.04%	98.95%	1.05%	99.07%	0.93%	99.23%	0.77%
	6-311++G**	99.19%	0.81%	98.89%	1.11%	98.91%	1.09%	98.98%	1.02%	99.19%	0.81%
	cc-pVTZ	99.22%	0.78%	98.90%	1.10%	98.96%	1.04%	99.03%	0.97%	99.21%	0.79%
	cc-pVQZ	99.22%	0.78%	98.92%	1.08%	98.98%	1.02%	99.02%	0.98%	99.21%	0.79%
PBE0	6-31G*	99.27%	0.73%	98.96%	1.04%	99.02%	0.98%	99.08%	0.92%	99.24%	0.76%
	cc-pVDZ	99.26%	0.74%	98.96%	1.04%	99.02%	0.98%	99.08%	0.92%	99.24%	0.76%
	6-311++G**	99.23%	0.77%	98.93%	1.07%	98.97%	1.03%	99.01%	0.99%	99.20%	0.80%
	cc-pVTZ	99.26%	0.74%	98.96%	1.04%	99.01%	0.99%	99.06%	0.94%	99.23%	0.77%
	cc-pVQZ	99.25%	0.75%	98.95%	1.05%	98.98%	1.02%	99.06%	0.94%	99.23%	0.77%
B972	6-31G*	99.23%	0.77%	98.94%	1.06%	99.00%	1.00%	99.06%	0.94%	99.23%	0.77%
	cc-pVDZ	99.24%	0.76%	-	-	98.98%	1.02%	99.05%	0.95%	99.23%	0.77%
	6-311++G**	99.14%	0.86%	98.89%	1.11%	-	-	98.99%	1.01%	99.19%	0.81%
	cc-pVTZ	99.23%	0.77%	98.92%	1.08%	98.98%	1.02%	99.03%	0.97%	99.22%	0.78%
	cc-pVQZ	99.18%	0.82%	98.93%	1.07%	98.99%	1.01%	99.03%	0.97%	99.21%	0.79%
M06	6-31G*	99.21%	0.79%	98.97%	1.03%	99.03%	0.97%	99.06%	0.94%	99.22%	0.78%
	cc-pVDZ	99.22%	0.78%	98.96%	1.04%	99.01%	0.99%	99.06%	0.94%	99.22%	0.78%
	6-311++G**	99.17%	0.83%	98.89%	1.11%	98.94%	1.06%	99.01%	0.99%	99.18%	0.82%
	cc-pVTZ	-	-	-	-	98.97%	1.03%	99.05%	0.95%	99.21%	0.79%
	cc-pVQZ	99.20%	0.80%	98.95%	1.05%	99.00%	1.00%	99.05%	0.95%	99.21%	0.79%
M11	6-31G*	-	-	-	-	99.05%	0.95%	99.11%	0.89%	99.23%	0.77%
	cc-pVDZ	-	-	-	-	99.05%	0.95%	99.11%	0.89%	99.23%	0.77%
	6-311++G**	-	-	-	-	98.98%	1.02%	99.05%	0.95%	99.18%	0.82%
	cc-pVTZ	99.17%	0.83%	-	-	99.01%	0.99%	99.09%	0.91%	99.20%	0.80%
	cc-pVQZ	99.20%	0.80%	-	-	99.01%	0.99%	99.07%	0.93%	99.20%	0.80%

Table S.28: Percentage contribution of enthalpy and entropy terms to the Gibbs free energy of reaction in $[An(NO_3)^{2+}_{(aq)}$ with An = Ac to Cf] calculated with B3LYP and PBE0, the Stuttgart RSC 1997 ECP and associated basis set including all functions in the basis set for the actinides, and the 6-31G*, and 6-311++G**, cc-pVTZ, and cc-pVQZ basis set for the non-actinide atoms. [Calculated as ΔH/ΔG x 100 and T.ΔS x 100].

	ΔH_{rxn}				$-T.\Delta S_{rxn}$			
	B3LYP		PBE0		B3LYP		PBE0	
	6-31G*	cc-pVDZ	6-311++G**	cc-pVTZ	6-31G*	cc-pVDZ	6-311++G**	cc-pVTZ
Ac	91.45%	84.37%	91.48%	86.02%	8.55%	15.63%	8.52%	13.98%
Th	94.89%	93.05%	95.09%	-	5.11%	6.95%	4.91%	-
Pa	93.15%	89.24%	93.30%	90.35%	6.85%	10.76%	6.70%	9.65%
U	93.94%	90.87%	94.69%	92.76%	6.06%	9.13%	5.31%	7.24%
Np	95.20%	93.37%	94.65%	92.72%	4.80%	6.63%	5.35%	7.28%
Pu	95.47%	93.51%	95.13%	93.20%	4.53%	6.49%	4.87%	6.80%
Am	95.33%	93.22%	95.33%	93.63%	4.67%	6.78%	4.67%	6.37%
Cm	95.09%	92.76%	95.04%	93.12%	4.91%	7.24%	4.96%	6.88%
Bk	95.66%	-	95.61%	94.24%	4.34%	-	4.39%	5.76%
Cf	95.81%	94.16%	95.78%	94.45%	4.19%	5.84%	4.22%	5.55%

Table S.29: T1 and D1 values for T1/D1 diagnostic for $[An(NO_3)]^{2+}$ (with An = Ac-Lr).

	T1	D1
Ac	0.020	0.065
Th	0.027	0.100
Pa	0.075	0.358
U	0.023	0.067
Np	0.024	0.067
Pu	0.025	0.074
Am	0.024	0.065
Cm	0.021	0.066
Dk	0.023	0.066
Cf	0.027	0.108
Es	0.029	0.121
Fm	0.025	0.128
Md	0.029	0.136
No	0.026	0.099
Lr	0.018	0.066

Table S.30: Calculated values for the B1 diagnostic for $[An(NO_3)]^{2+}$ (with An = Ac-Es).

An	B1 (n=1)	B1 (n=2)	B1 (n=3)	B1 (n=4)
Ac	5.77	2.88	1.92	1.44
Th	7.00	3.50	2.33	1.75
Pa	16.26	8.13	5.42	4.07
U	10.92	5.46	3.64	2.73
Np	12.13	6.07	4.04	3.03
Pu	15.90	7.95	5.30	3.97
Am	18.70	9.35	6.23	4.68
Cm	14.29	7.15	4.76	3.57
Bk	17.94	8.97	5.98	4.48
Cf	19.83	9.91	6.61	4.96
Es	21.36	10.68	7.12	5.34