Covalency hinders $AnO_2(H_2O)^+$ isomerisation (An = Pa–Pu)

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Supplementary Information

Table S1. $\langle S^2 \rangle$, before annihilation of the first spin contaminant, for AnO^+, AnO_2^+, $AnO_2(H_2O)^+$, $AnO(OH)_2^+$ and the isomerisation TS. Formal values in parentheses.

	AnO^+	AnO_2^+	$AnO_2(H_2O)^+$	TS	$AnO(OH)_2^+$
Pa	2.01 (2.00)				
U	3.76 (3.75)	0.75 (0.75)	0.75 (0.75)	0.75 (0.75)	0.76 (0.75)
Np	6.01 (6.00)	2.01 (2.00)	2.02 (2.00)	2.02 (2.00)	2.03 (2.00)
Pu	8.78 (8.75)	3.79 (3.75)	3.79 (3.75)	3.80 (3.75)	3.83 (3.75)

Cartesian atomic coordinates and total energies (H)

PaO^+ -516.4449534
Pa 0.000000 0.000000 0.143532
O 0.000000 0.000000 -1.632681

UO^+ -551.9965166
U 0.000000 0.000000 0.144063
O 0.000000 0.000000 -1.656724

NpO^+ -589.5156462
Np 0.000000 0.000000 0.142109
O 0.000000 0.000000 -1.652012

PuO^+ -628.9949106
Pu 0.000000 0.000000 0.139539
O 0.000000 0.000000 -1.639583

PaO_2^+ -591.8401929
Pa 0.000000 0.000000 0.000000
O 0.000000 0.000000 1.766496
O 0.000000 0.000000 -1.766496

UO_2^+ -627.3555306
U 0.000000 0.000000 0.000000
O 0.000000 0.000000 1.752904
O 0.000000 0.000000 -1.752904

NpO^+ -664.8455542
Np 0.000000 0.000000 0.000000
O 0.000000 0.000000 1.719856
O 0.000000 0.000000 -1.719856

PuO_2^+ -704.2904163
Pu 0.000000 0.000000 0.000000
O 0.000000 0.000000 1.714284
O 0.000000 0.000000 -1.714284

$PaO_2(H_2O)^+$ -668.3583854
Pa 0.223127 0.000054 -0.000005
O 0.210657 -1.782323 0.000028
O 0.209626 1.782424 0.000030
O -2.252389 -0.000547 -0.000006
H -2.824022 0.781703 0.000002
H -2.823646 -0.783071 0.000036

$UO_2(H_2O)^+$ -703.8709966
U 0.220372 0.000071 0.000000
O 0.212397 -1.767079 -0.000007
O 0.211010 1.767217 0.000007
O -2.251481 -0.000738 0.000000
H -2.824990 0.779706 -0.000123
H -2.824623 -0.781450 0.000122

$NpO_2(H_2O)^+$ -741.3614439
Np 0.216046 -0.014925 0.000000
O 0.061607 -1.752854 -0.000025
O 0.355389 1.724369 0.000030
O -2.228642 0.153844 -0.000008
H -2.746712 0.971808 -0.000462
H -2.852439 -0.586651 0.000518

$PuO_2(H_2O)^+$ -780.8056369
Pu -0.213559 -0.000045 0.000003
O -0.207203 1.727520 -0.000022
O -0.206316 -1.727604 -0.000019
O 2.223716 0.000452 0.000012
H 2.796728 -0.779834 -0.000063
H 2.796214 0.781116 -0.000013

$Ts\text{-}Pu^+$ -780.729854
Pu 0.153428 -0.127135 0.000002
O 1.616377 0.780145 -0.000008
O -1.520089 -0.944345 0.000011
O -1.429604 1.362778 -0.000025
H -2.007532 0.089481 -0.000006
H -1.748215 2.272546 -0.000041

$Ts\text{-}Pa^+$ -668.303836
Pa -0.166328 -0.128384 0.000000
O -1.671700 0.803209 0.000000
O 1.544091 -0.962157 0.000000
O 1.524901 1.327147 0.000001
H 2.041873 0.127912 0.000001
H 1.915623 2.209478 0.000002

$PaO(OH)_2^+$ -668.3557574
Pu 0.055942 -0.145071 0.000001
O 1.791360 -0.108396 -0.000006
O -1.894916 -0.412448 -0.000008
O -0.235857 1.849052 0.000003
H -2.613653 0.245590 0.000012
H 0.070379 2.765409 -0.000025

$Ts\text{-}U^+$ -703.8082436
U -0.162320 -0.117079 -0.000008
O -1.689781 0.751054 -0.000022
O 1.518464 -0.985862 -0.000001
O 1.540231 1.301272 0.000092
H 2.044236 0.059643 0.000054
H 1.937898 2.179933 0.000147

$UO(OH)_2^+$ -703.8486403
U 0.085877 -0.105297 0.000001
O 1.856831 -0.084468 -0.000029
O -1.808731 -0.772623 0.000009
O -0.569221 1.810187 0.000003
H -2.768509 -0.629661 -0.000075
H -0.963202 2.692204 0.000145

$Ts\text{-}Np^+$ -741.2920609
Np -0.157412 -0.126536 0.000000
O -1.632968 0.785531 0.000000
O 1.522196 -0.955186 0.000000
O 1.461228 1.348376 0.000002
H 2.013320 0.095428 0.000001
H 1.822396 2.242651 -0.000001

$NpO(OH)_2^+$ -741.3258815
Np -0.056107 -0.141510 -0.000003
O -1.802456 -0.158028 0.000013
O 1.905988 -0.415365 0.000008
O 0.215954 1.857459 0.000004
H 2.727190 0.105800 0.000035
H -0.065133 2.782116 0.000008

$PuO(OH)_2^+$ -780.7582018

Pu 0.055942 -0.145071 0.000001

O 1.791360 -0.108396 -0.000006

O -1.894916 -0.412448 -0.000008

O -0.235857 1.849052 0.000003

H -2.613653 0.245590 0.000012

H 0.070379 2.765409 -0.000025