Supplementary information

for

Structure and Thermodynamics of Uranium(VI) Complexes in Gas Phase: A Comparison of Experimental and *Ab Initio* Data.

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Table S1. Calculated entropy and heat capacity contributions in J/K.mol for UO_2F_2 at 298.15°K and T=1500°K. The numbers in parentheses are obtained from partition functions calculated using vibration frequencies scaled with 0.9 (see text).

Property	Temperature	Value
S_{trans}	298.15	180
S_{rot}	298.15	107
S_{vib}	298.15	48 (53)
S_{tot}	298.15	334 (339)
$C_{p,vib}$	298.15	49 (52)
C_p	298.15	82 (85)
S_{trans}	1500	200
S_{rot}	1500	127
S_{vib}	1500	152 (160)
S_{tot}	1500	479 (487)
$C_{p,vib}$	1500	73 (73)
C_p	1500	106 (106)

Table S2. A summary of total energies (with p-functions on hydrogen atoms). Energies in a.u.

Complex	SCF	MP2	CCSD(T)	B3LYP
UF_6	-619.3712336	-620.9137626	-620.94016241	-623.23717213
trans-$UO_2(OH)_2$	-539.6197394	-540.83294968	-540.85485085	-542.61837370
UO_3	-522.5874913	-523.62917897	-523.62622914	-525.29048855
HF	-24.5649355	-24.753409633	-24.7601377	-24.914514027
H_2O	-16.9396398	-17.14305792	-17.1557647	-17.262406641
UO_2F_2	-554.9211849	-556.09315629	-556.10640772	-557.97304939

Table S3. The enthalpy function ΔH_T(=ΔH_{trans}+ΔH_{rot}+ΔH_{vib}) for reactions 1-5 at T=298.15~K

Reaction	ΔH_T kJ/mol
$2UO_3 + UF_6 \rightarrow 3UO_2F_2$	3
$UO_2F_2 + 2H_2O \rightarrow UO_2(OH)_2 + 2HF$	-2
$UF_6 + 2H_2O \rightarrow UO_2F_2 + 4HF$	-12
$UO_3 + H_2O \rightarrow UO_2(OH)_2$	5
$UF_6 + 3H_2O \rightarrow UO_3 + 6HF$	-20