Supplementary Material for

Systematic Raman Spectroscopic Study of the Complexation of Uranyl with Fluoride

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1. Tables

Table S1 Experimental results for the UO_2^{2+}-$(CH_3)_4NF$ system. I=2.5 mol/L $(CH_3)_4NF/(CH_3)_4NNO_3$

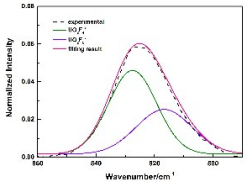
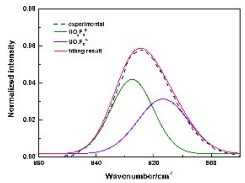
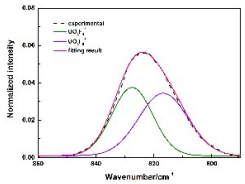
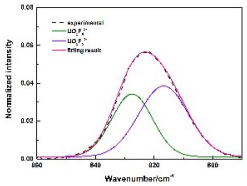
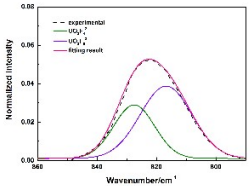
C(F^-) mol/L	C(UO_2^{2+}) mol/L	Curve fitting of normalized Raman spectra of O=U=O moiety	[$UO_2F_4^{2-}$] mol/L	[$UO_2F_5^{3-}$] mol/L	[F^-] mol/L	K_5	$\bar{K}_5$
0.82	0.059		0.039	0.020	0.56	0.93	
1.14	0.059		0.031	0.028	0.88	1.0	
1.63	0.059		0.027	0.032	1.36	0.86	0.92
1.90	0.059		0.024	0.035	1.63	0.88	
2.37	0.059		0.020	0.039	2.10	0.91	

Table S2 U=O bond distances (Å) and UO_2^{2+} symmetric stretching frequencies (1/cm) by the COSMO approach at the B3LYP level and gasphase/COSMO approach at MP2 level calculations of $[UO_2(H_2O)_mF_n]^{2-n}$ complexes with H_2O directly coordinated to U.

Complex	COSMO, B3LYP		gasphase, MP2		COSMO, MP2[a]	
	R(U=O)	ω_e	R(U=O)	ω_e	R(U=O)	ω_e
$[UO_2(H_2O)_5]^{2+}$	1.753	912	1.753	895	1.761	881
$[UO_2(H_2O)_4F]^{+}$	1.770	877	1.774	861	1.777	854
$[UO_2(H_2O)_3F_2]_{meta}$	1.785	847	1.783	845	1.789	833
$[UO_2(H_2O)_2F_3]^{-}_{ortho}$	1.800	819	1.800	814	1.799	815
$[UO_2(H_2O)F_3]^{-}$	1.801	824	1.799	817	1.799	817
$[UO_2F_4]^{2-}$	1.817	819	1.823	782	1.817	795
$[UO_2F_5]^{3-}$	1.830	769	1.834	756	1.826	777

[a]MP2 calculations for the complexes $[UO_2(H_2O)_3F_2]_{meta}$, $[UO_2(H_2O)_2F_3]^{-}_{ortho}$, and $[UO_2(H_2O)F_3]^{-}$, were done using the Resolution-of-the-Identity approximation (RI-MP2). Auxiliary basis sets of def2-TZVPP quality for H, O, and F were used. For U the RI basis set (11s10p9d7f6g6h5i)/[11s10p9d7f6g6h5i] developed by B. Schimmelpfennig was applied. Test calculations have shown that the RI approximation changed the listed MP2 distances and frequencies by at most 0.002 Å and 1 cm^{-1}, respectively.

Table S3 UO_2^{2+} symmetric stretching frequencies (1/cm) from B3LYP COSMO calculations of $[UO_2(H_2O)_mF_n]^{2-n}(H_2O)_k$ complexes in comparison to Raman data for aqueous solution. The additional k H_2O molecules form hydrogen bridges to the F^- ligands.

complex	B3LYP, COSMO	Raman
$[UO_2(H_2O)_5]^{2+}$	912	870
$[UO_2(H_2O)_4F]^{+}(H_2O)$	880	858
$[UO_2(H_2O)_4F]^{+}(H_2O)_2$	878	858
$[UO_2(H_2O)_3F_2](H_2O)_{3\ ortho}$	861	848
$[UO_2(H_2O)_3F_2](H_2O)_{4\ meta}$	859	848
$[UO_2(H_2O)F_3]^{-}(H_2O)_4$	845	837
$[UO_2F_4]^{2-}(H_2O)_4$	830	828
$[UO_2F_5]^{3-}(H_2O)_5$	817	817*

*extrapolated Raman value

Table S4 Bond distances (Å) and UO_2^{2+} stretching frequencies (1/cm) using the COSMO approach in MP2 level calculations of $[UO_2(H_2O)_mF_n]^{2-n}(H_2O)_k$ complexes in comparison to Raman data for aqueous solution. The additional explicitly treated k H_2O molecules form hydrogen bridges to the F^- ligands[a].

complex	R(U=O)	R(U-OH$_2$)	R(U-F)	ω_{sym}	ω_{asym}	Ref[b]
$[UO_2(H_2O)_5]^{2+}$	1.761	2.420	-	881	954	1.76, 2.41, 870, 965
$[UO_2(H_2O)_4F]^+(H_2O)$	1.772	2.469	2.177	860	942	858[c]
$[UO_2(H_2O)_4F]^+(H_2O)_2$	1.770	2.469	2.204	863	942	858[c]
$[UO_2(H_2O)_3F_2](H_2O)_{3\ ortho}$	1.779	2.506	2.226	848	927	848[c]
$[UO_2(H_2O)_3F_2](H_2O)_{4\ meta}$	1.778	2.492	2.252	850	927	848[c]
$[UO_2(H_2O)F_3]^-(H_2O)_4$	1.786	2.476	2.220	837	912	1.80, 2.47, 2.25, 837[c]
$[UO_2F_4]^{2-}(H_2O)_4$	1.798	-	2.231	819	890	828[c]
$[UO_2F_5]^{3-}(H_2O)_5$	1.797	-	2.232	818	892	1.80, 2.26, 784, 850 817[c]

[a]Except for the complexes $[UO_2(H_2O)_5]^{2+}$ and $[UO_2F_4]^{2-}(H_2O)_4$, the MP2 calculations were done using the Resolution-of-the-Identity approximation (RI-MP2). Auxiliary basis sets of def2-TZVPP quality for H, O, and F were used. For U the RI basis set (11s10p9d7f6g6h5i)/[11s10p9d7f6g6h5i] developed by B. Schimmelpfennig was applied. Test calculations have shown that the RI approximation changed the listed MP2 distances and frequencies by at most 0.002 Å and 1 cm^{-1}, respectively.

[b]EXAFS data from ref. 21, except as otherwise indicated.

[c]present work

Table S5: Cartesian coordinates and total energies of the optimized B3LYP equilibrium structures of selected complexes $[UO_2(H_2O)_mF_n]^{2-n}(H_2O)_k$ in aqueous solutions modeled by COSMO as implemented in TURBOMOLE are listed below. Only those complexes are listed where all water molecules in the second coordination sphere form hydrogen bonds to the fluoride ions in the first coordination sphere, since for these cases the effects on the vibrational modes of uranyl were found to be strongest. Basis sets of def2-TZVPP quality for H, O, and F were used. For U the ECP60MWB PP and the corresponding (14s13p10d8f6g)/[10s9p5d4f3g] basis set were applied. The coordinates are given in Bohr in TURBOMOLE format. Fine grids (5 or higher/better in TURBOMOLE) and total energy convergence to at least 10^{-8} Hartree were required to get all real vibrational frequencies.

（1）$[UO_2(H_2O)_5]^{2+}$ E=-1009.848935139 Hartree

$coord

0.09723598046692	0.04918232103920	0.00253555461392	u
0.10789131351419	0.12457655038580	3.31384762518114	o
0.08763564076838	0.11691148491801	-3.30864387763171	o
2.96676730634930	3.62062474289100	-0.01522871654925	o
3.59263200924106	4.50688135117062	1.45028061880683	h
3.56726481570991	4.51795453057196	-1.48467274458099	h
0.01810500697277	-4.64472305258075	0.06618023425767	o
1.47619380960052	-5.66345547712244	-0.33085613182268	h
-1.46843451805638	-5.62051826110330	-0.33326403569031	h
-4.19817555946266	-1.65821677630119	0.00781397000687	o
-5.26278550833863	-1.78623193909081	1.48372352798875	h
-5.30195731567002	-1.70512849209736	-1.44424970546794	h
-2.65287334701367	3.71443640525791	0.01057147886562	o
-3.30881071954280	4.56828784778360	1.48214412729160	h
-3.29960141299260	4.58938170403743	-1.45268660123937	h
4.33107022264565	-1.80296267799195	-0.01057444168427	o
5.39549315759775	-1.96120039642819	1.46256291501200	h
5.42950435286281	-1.88203595267902	-1.46538514630950	h

$end

（2）$[UO_2(H_2O)_4F]^{+}$ E=-1033.463673271 Hartree

$coord

-0.03417975167756	0.25522887059082	0.17643852989208	u
-0.80467841063537	-0.05597052574963	3.41652055049213	o
0.44008189052169	0.73112493510607	-3.10004966317430	o
1.58609044919904	4.56843854454443	1.01981433082444	o
2.02677239413189	5.13457092222925	2.69561715989950	h
2.69755489373411	5.44425985141999	-0.12956997342942	h
0.90424567928150	-4.39697207375719	-0.13267624093065	o
2.62679067650329	-4.93274606697958	-0.38849355645054	h
-0.17053214965180	-5.55016728933548	-1.04575810298950	h

-3.81182932349778 -2.39780061770551 -0.89268397935349 o
-5.08793754067515 -2.84477931199942 0.32998090396181 h
-4.65296415187611 -2.31592927613976 -2.50803905399425 h
-3.70980393687964 3.17504727995737 -0.17852758040236 o
-4.59536473166362 3.88052668758343 1.24923770962313 h
-3.98489586236455 4.31046654605975 -1.57712442642762 h
3.88884758983673 -0.41501028904694 0.82764315415903 f
$end

（3）$[UO_2(H_2O)_3F_2]_{meta}$ E= -1057.068154422 Hartree

$coord
-0.09792769770668 -0.00505903194197 -0.00601620413523 u
0.01018025758204 -0.01353747277688 3.36510777433263 o
-0.06581067937758 0.00336462443628 -3.37891907958437 o
3.98943062704485 -2.62114241549433 -0.08962542048149 o
5.23608336669592 -2.41333880873257 1.22364371358298 h
3.70684716943902 -4.41743286944936 -0.22749529245106 h
-4.87796937874068 -0.00020065990547 0.09571622883854 o
-5.76080179070513 1.48458972059009 -0.48431428510381 h
-5.78086049166917 -1.43854620707006 -0.56490893375622 h
-1.27931606211051 3.92420132347378 0.04153885873812 f
3.97449035009193 2.63933343338812 0.00472110476867 o
3.66599358071730 4.43192728955455 0.13436579280324 h
5.21261239429254 2.44202763711353 -1.31815969545072 h
-1.27412984879095 -3.93397214432767 -0.03977078448849 f
$end

（4）$[UO_2(H_2O)_3F_2]_{ortho}$ E=-1057.067838170 Hartree

$coord
-0.03263641579030 -0.37184340040329 0.02328480218256 u
-0.11167191919621 -0.26278241304311 3.39493176507299 o
0.01116155497543 -0.14502164710999 -3.34312083014446 o
4.24308211230402 1.95652138903108 0.20133928186461 o
5.64317686035006 0.89931211330904 -0.29223295706762 h
4.49030966616950 3.56464765759219 -0.61679460136664 h
-2.47431003461804 -3.64867234334868 -0.07457211596614 f
-4.70571943006021 0.99237026999897 -0.10261366266030 o
-5.27091696273652 2.51385413705698 0.72337996747570 h
-5.86369544737101 -0.32793877537568 0.38449313317063 h
-0.55262779706413 4.39792502319420 0.10032452164259 o
-0.48206521876510 5.47090762354356 1.56990522302029 h
-0.82827249733623 5.48678092565788 -1.33295163297888 h
3.06147321219162 -3.04423377360343 0.03200651579147 f

$end

(5) $[UO_2H_2OF_3]^-H_2O$ E= -1080.666960775 Hartree

$coord

0.43023951564143	-0.15186906697449	-0.19581302312970	u
1.52533594394453	0.38649670142831	-3.36238487726690	o
-0.77361900371743	-0.57975820871801	2.94864441366919	o
3.70242918743541	-2.47475976552356	0.64058169950525	f
-3.20029167905520	2.80493680614876	-1.13281422668951	o
-4.70245448267857	2.70396056688617	-0.10796680216689	h
-2.64436265436441	4.59415808055261	-1.09616674602476	h
-2.00328273152561	-3.17466282828057	-1.52940143259465	f
-0.88766198648494	7.27553472547316	-0.59222462031085	o
0.58240698172952	6.31688810783711	-0.02174933253365	h
-0.35890587961981	8.17401106490811	-2.08415562888849	h
2.17946585395774	3.49156819581374	0.95134551495601	f

$end

(6) $[UO_2(H_2O)_2F_3]^-$ E= -1080.661652465 Hartree

$coord

-0.00223450834213	0.12231106662014	0.00146839194113	u
-0.05880664988229	0.15068489898947	-3.40001455607087	o
0.05462990700332	0.16975769469960	3.40268393503108	o
4.56982284701302	1.95673302366591	-0.12910551032960	o
5.83693776453077	0.88813859260613	0.62532883453760	h
4.73881733704347	3.59890102541215	0.64010160619718	h
-4.58268282577875	1.93783569917888	0.12463808741147	o
-4.70052649027251	3.60949778801019	-0.58890025748318	h
-5.86281699663903	0.92737146143598	-0.68558894082221	h
-2.80751351580112	-2.94396231007219	0.03375032858503	f
2.83914372918718	-2.91449497729199	-0.01560048665055	f
-0.02474036244533	4.31525489543776	-0.00875765289618	f

$end

（7）$[UO_2(H_2O)F_3]^-$ E= -1004.158892453 Hartree

$coord

0.42639613973412	-0.04521807171379	-0.16171871374948	u
1.45421472988486	0.74039118146324	-3.30878004097510	o
-0.81379743946458	-0.67615573622923	2.94329682901015	o
3.71018458500080	-2.42642992263419	0.49073519727036	f
-3.38317096705218	2.68444675423768	-0.91791905324132	o
-4.35103196979772	3.39338005309819	0.49106884315197	h
-3.37988563700167	3.99384664523421	-2.22541785864204	h
-1.78632361414668	-3.16855062778924	-1.67365832567716	f
2.30019141643116	3.32108080119441	1.28189031906620	f

$end

（8）$[UO_2F_4]^{2-}$ E= -1027.768185139 Hartree

$coord

-0.48151331297536	0.55458620168436	-0.27169995534497	u
-1.27258341400711	-0.29129250847092	2.96011764216251	o
0.30966158010447	1.40057360470798	-3.50353774198257	o
1.03649744345223	4.20725381010496	1.05560095286496	f
-1.99948396649125	-3.09805557741490	-1.59925637463359	f
-4.24603437244238	2.28959910634275	-0.73773533686870	f
3.28314618163075	-1.18044640072717	0.19413295284029	f

$end

（9）$[UO_2F_5]^{3-}$ E=-1127.797406182 Hartree

$coord

0.00013306133062	-0.00038218879070	0.00000384943373	u
0.00071254936890	-0.00115662879319	3.45882830294146	o
0.00053447422831	-0.00129632349816	-3.45883727728722	o
3.96146506286785	-1.68384246834872	-0.00018476824787	f
-4.19430180513822	-0.96452947126781	0.00018351223591	f
-2.21353420825312	3.69044931512668	-0.00003623109475	f
2.82432014246391	3.24700695866812	-0.00009113339459	f
-0.37837874466512	-4.28760412667401	0.00012051733103	f

$end

（10）$[UO_2(H_2O)_4F]^{+}(H_2O)$ E=-1109.951012513 Hartree

$coord

-0.42662983759966	0.00768881570141	0.03088133752573	u
-0.58881681181419	-0.04519125349342	3.36859476313795	o
-0.56515229108594	0.04364790805404	-3.30793242191758	o
-2.59617323499290	4.12572238664177	0.08168073596549	o
-2.77860317878305	5.15780680410993	1.57300191938779	h
-2.69912163725832	5.24859505875046	-1.35024141757524	h
3.40477348087051	-2.68731793670177	0.01060107486074	o
5.06405644802590	-1.80119613542591	-0.02691863882611	h
3.52621022253750	-4.13794360843948	-1.08321670378348	h
-1.28728626664432	-4.71820408670427	-0.03609416187401	o
-0.61881728682318	-5.81285932007576	1.25994268933880	h
-3.02881223114732	-5.20903594907033	-0.26731545724461	h
-5.05478454520629	-0.97791408321382	-0.12098654252736	o
-6.14858833536931	-0.90795557832614	1.33593070554974	h
-6.10139749410486	-0.62832334141203	-1.57231514382034	h

2.75818561065894 2.55055275109983 0.11223720716852 f
7.45688522119458 0.25075717330051 0.14393051171268 o
8.54940619701963 0.27629147373212 -1.31333132834868 h
6.37983605010644 1.72540044204574 -0.01252559385585 h
$end

（11）$[UO_2(H_2O)_4F]^+(H_2O)_2$ E=-1186.438841571 Hartree

$coord
-0.45608633278510 -0.01237817462606 0.06590219276724 u
-0.25367912983203 -0.37567685197765 3.37961575551222 o
-0.87288468480612 0.38168283523422 -3.22456528571842 o
1.31229346591465 4.29548690464304 0.44736647098403 o
3.14757431549306 4.50808678639783 0.09340533975450 h
0.44229692785358 5.77444779776609 -0.15921832982759 h
0.06613025885971 -4.61578860279400 -0.44907762000892 o
1.79632218329377 -5.24878868145583 -0.04217586109835 h
-0.45456444081376 -5.43510863537130 -1.99098879549907 h
-4.61240468854866 -2.39414300348054 0.14885614921605 o
-4.69127886773443 -4.02509437447524 0.95979972588491 h
-6.14941403907881 -1.51977897801432 0.59254952668911 h
-4.01386793523933 2.97693560303134 0.63610308372968 o
-4.58831925492763 3.67231218766467 2.22041608843221 h
-4.91163218937219 3.85461977916007 -0.68556666559520 h
3.63102668454317 -0.47572488687750 -0.29005638504317 f
4.79720980798103 -5.54182293987184 0.80954490020483 o
5.79966537602358 -6.62844872075133 -0.25472566869497 h
5.31434690506701 -3.83117488313382 0.40930311050559 h
6.20851156781745 4.11939194383364 -0.56227422819232 o
7.33889776281859 4.62178319294037 0.77496870081574 h
6.15045417091159 2.28892632474589 -0.50340349168342 h
$end

（12）$[UO_2(H_2O)_3F_2](H_2O)_3$ E=-1286.525913833Hartree

$coord
0.01549563024380 -0.44794901083072 0.08264880398733 u
-0.05016531204313 -0.61315944436688 -3.27215100995915 o
0.08252824325392 -0.56048110976126 3.43949691599722 o
-2.70635536485505 2.73330242167546 0.10996705499993 f
0.00962613956437 7.36611865252715 0.43306064776157 o
-1.40617906335679 6.20980897313125 0.37418679161094 h
-7.62346158126585 1.56235473396092 -0.75653058906727 o
-6.09349772153701 2.57826839823802 -0.65254565527166 h
6.13066865091409 2.60331862979207 -0.63431113001291 h

1.42568028970679 6.21578180586725 0.30457210617681 h
-4.41291281335999 -2.24599726597755 0.26263201212480 o
-5.80969923855365 -1.11295184512377 -0.23747619485581 h
-4.76486186590045 -3.90016065983256 -0.40751874004236 h
-8.23928428298090 1.76003591481217 -2.45830128962741 h
0.00747439808007 -5.22588238795235 0.00970766813254 o
0.11027832768126 -6.32201907957639 1.46072603214666 h
-0.01092836039125 -6.29904532322348 -1.46163536491084 h
4.44734894196666 -2.24692935192539 0.09708367883961 o
5.84470786984298 -1.09045153208987 -0.34853593571882 h
4.79461029099925 -3.86195930417656 -0.66440404903830 h
7.60718753240924 1.57517467594909 -1.01776534757224 o
8.84874545225515 1.99500155132626 0.24595123463544 h
2.72545063162841 2.74268514555105 0.00849837343241 f
$end

（13）$[UO_2(H_2O)_3F_2](H_2O)_4$ E=-1363.019120740Hartree

$coord
0.01612126409775 -0.32123986907938 0.05048836134474 u
0.17387807511565 -0.87304841668858 3.35963301589231 o
-0.12446597626472 0.10847232407541 -3.27702054861789 o
4.05502940330389 0.94982018474166 0.07422281580721 f
-4.04121561129539 0.84539375654565 0.35865960596970 f
4.58755711255545 6.01807811799152 -0.74789671329991 o
5.01889996814045 4.24818256859532 -0.49726546697145 h
-7.43458272270242 -2.99855828811269 0.11723261300080 o
-6.78031826890451 -1.28087996601033 0.19050818564917 h
7.37532885973584 -2.93401890189244 0.52335241065778 o
6.75147756927617 -1.20926592137479 0.37669765691119 h
-4.76256302123276 5.91287609578050 -0.34552491133358 o
-5.12608339023512 4.12972761674219 -0.08164381597040 h
-0.01286669190510 4.31111652553068 0.76908946044687 o
-1.56144589152204 5.25234452002378 0.35730150875543 h
1.47332030592976 5.28731771097533 0.22866018002737 h
-5.64878862429880 6.79893477028964 0.97495318546301 h
5.55971873854706 6.94334943213140 0.48190325538558 h
2.62015834462266 -4.28262885112078 -0.56438648290686 o
4.45980230258253 -4.07801462590031 -0.27712488054104 h
2.42796820481163 -5.23178317633826 -2.10642142868207 h
8.63613775607467 -3.12075507590500 -0.77687135100974 h
-2.57697751236363 -4.30044621173909 -0.48441972956894 o
-4.43183526152718 -4.12110775361485 -0.31291253735624 h
-2.08740121078503 -5.78945546237862 0.44111334768820 h
-8.49225111917962 -3.06795092430119 -1.36310547245554 h

$end

（14）$[UO_2(H_2O)F_3]^-(H_2O)_4$ E=-1310.1168338980 Hartree

$coord

0.84476337087286	0.14603629569650	-0.33284970921661	u
1.44490436581275	0.01295469383694	-3.65317509181320	o
0.20005782331632	0.31348537152208	2.97625651292238	o
4.16862326904630	-2.25795223987933	0.39876679822884	f
7.93448831609755	1.75117578189078	0.95490655037573	o
7.13725320370325	0.10812773279130	0.86667794242565	h
-5.29168907626829	-1.84702878215096	-0.04750828541425	h
0.10658662483638	-6.23324141084658	-0.05688033050903	h
6.55472307369165	2.91410914419340	0.66396659360900	h
-2.89377629095899	2.86409930900150	-1.14362982231370	o
-4.47499757804984	2.07419937445975	-0.56625092367378	h
-2.71505078638525	4.56151967689637	-0.40298979271275	h
-6.56531051576972	-0.52506170426006	0.00594972233568	o
1.63782118636399	-7.16751241277579	0.29371656826412	o
2.91342909781759	-5.86494375887904	0.43308805224986	h
-7.30067511402927	-0.61698577399189	1.66861590829239	h
-1.90552505015595	-2.99632802709287	-0.65192254041330	f
-1.06462384847108	7.25157179268633	0.79393087245089	o
0.62267523873066	6.56284915114127	0.56548336550987	h
-1.15060740763077	8.70385342592622	-0.30085623268630	h
2.88033042770124	3.79621805670138	-0.05959116641714	f

$end

（15）$[UO_2F_4]^{2-}(H_2O)_4$ E=-1333.706427662 Hartree

$coord

-0.32224767683968	0.69108366939790	-0.45110843535336	u
0.58812057428963	0.70271559201432	2.81823980365896	o
-1.23237818898067	0.67982792699492	-3.72049530657687	o
2.68175865518900	-2.16189723756575	-1.28274478384959	f
-3.06761078411152	-2.42484283293991	0.33182663437331	f
-3.32779881109062	3.54293855431566	0.37970784111896	f
2.42290200118443	3.80736479045596	-1.23333597304040	f
0.01531030583969	-6.77701345683342	-0.49833634901504	o
-1.41209958671966	-5.69745541040916	-0.11123293278201	h
1.33101085284258	-5.55983478812581	-0.87466690362505	h
-0.67900604866984	8.15686417383894	-0.45020281049645	o
0.75225511860768	7.07964237938210	-0.82933500303675	h
-1.98781295647059	6.93802303302272	-0.05539145437713	h
6.64255803349010	1.02836852551444	-3.10443370058684	o
5.53504372493804	2.39869385405577	-2.60448779733246	h

5.66572660501125	-0.44956433064311	-2.64112286863794	h
-7.28378500697294	0.35366636319341	2.20955762991267	o
-6.31093536366276	1.83081497771103	1.73534994591898	h
-6.17419157358271	-1.01723379167854	1.71587105550266	h

$end

（16）$[UO_2F_5]^{3-}(H_2O)_5$ E=-1510.244667967Hartree

$coord

0.03460777805748	0.15143249114421	0.52034393247473	u
0.00474201240534	0.09166447977219	3.92486423043231	o
0.06454488469833	0.21175234549383	-2.88354710076355	o
3.66661634510442	-2.30039407337936	0.50860896287908	f
-4.34522144643497	0.00167559293602	0.48831983492755	f
-1.46088395691794	4.26980558250154	0.57878514983040	f
3.48931196827579	2.84678882369353	0.59751946640225	f
-1.17534170673697	-4.05851972742819	0.43422988284653	f
7.95887016342653	0.43598813396970	0.07243687437312	o
6.70897848916599	1.77929636759636	0.22213882714484	h
6.81132826974627	-0.99966134723362	0.17268412395550	h
-6.52747260800105	4.59468242421186	0.02459555909935	o
-4.75777685679197	5.07617451187885	0.18161836054171	h
-6.31148408722223	2.76986739824388	0.13543102331996	h
-6.20977479279953	-4.70869804524412	-0.13681366921818	o
-6.11484808896305	-2.87836369249097	0.03735095565368	h
-4.41197091310657	-5.07723019685094	0.00485644386211	h
2.73635769208164	-7.30346607134903	0.93271619951863	o
3.63315546626633	-5.69893593458257	0.83201140920667	h
1.02055123511735	-6.65204501070247	0.79125366811077	h
2.21927276811831	7.75626359303061	1.19354386974761	o
3.22548136027052	6.22251491739862	1.03879000356943	h
0.55278170158553	6.99167218858789	1.02995006620132	h

$end

Table S6: Cartesian coordinates and total energies of optimized MP2/RIMP2 equilibrium structures of selected complexes $[UO_2(H_2O)_mF_n]^{2-n}(H_2O)_k$ in aqueous solution modeled by COSMO as implemented in TURBOMOLE are listed below. Only those complexes are listed where all water molecules in the second coordination sphere form hydrogen bonds to the fluoride ions in the first coordination sphere, since for these cases the effects on the vibrational modes of uranyl were found to be strongest. Basis sets of def2-TZVPP quality for H, O, and F were used. For U the ECP60MWB PP and the corresponding (14s13p10d8f6g)/[10s9p5d4f3g] basis set, as well as the RI basis set (11s10p9d7f6g6h5i)/[11s10p9d7f6g6h5i] by B. Schimmelpfennig, were applied. All occupied orbitals with orbital energies below -3.0 Hartree were kept frozen in RIMP2. The coordinates are given in Bohr in TURBOMOLE format.

（1）$[UO_2(H_2O)_5]^{2+}$ E_{MP2}= -1008.7164258472 Hartree

$coord

0.01070747024414	0.01434820717103	0.00241074520902	u
0.02315641235354	0.14558803731781	3.32820056533481	o
0.01342699537155	0.03472936089164	-3.32614458621425	o
3.12885447075212	3.30307473724403	-0.05614339126558	o
3.84131056750535	4.14393598958361	1.39316686896963	h
3.81420382962489	4.13886400914651	-1.52142227929927	h
-0.43281214550846	-4.60745673348044	0.18598920425040	o
0.93433364294042	-5.71971207614677	-0.27016369873224	h
-1.97621481886898	-5.45002780601436	-0.28566847022273	h
-4.35933296426168	-1.34647840574841	0.02933146966821	o
-5.42647376258285	-1.29644866424197	1.50542961746254	h
-5.48217210437021	-1.21443368779577	-1.39995722808165	h
-2.41283383238689	3.84362296884335	-0.04497926022363	o
-2.96177110107275	4.79416484957934	1.40768778733779	h
-2.92476001567584	4.80096605496308	-1.50673277983939	h
4.04080912379164	-2.15900833379953	0.03912756838527	o
5.09282038880856	-2.30755047697152	1.51947571825495	h
5.17510252922983	-2.23339320701088	-1.38532208443006	h

$end

（2）$[UO_2(H_2O)_4F]^{+}$ E_{MP2}= -1032.3086252331 Hartree

$coord

-0.04899799916213	0.23290777184300	0.17073627015105	u
-0.82981979834787	-0.06375538734103	3.42372583206170	o
0.44851810772742	0.69530856480997	-3.11872351387691	o
1.48234388610050	4.53606383504366	0.94442435629676	o
1.89968017144093	5.08719088541898	2.62917518478788	h
2.75463317328366	5.27363558401538	-0.12998714534769	h
0.86886919595854	-4.36028141753881	-0.07381895275890	o

2.61360993943117 -4.81279189703246 -0.33056350173972 h
-0.10841328858339 -5.45111902684642 -1.15431338129592 h
-3.76988312414887 -2.40089695235652 -0.90161656340744 o
-4.98040605869998 -2.89729678026529 0.36517711145090 h
-4.75170056802024 -2.06951603427822 -2.39931454518854 h
-3.70640560218802 3.08621090282427 -0.22140785233832 o
-4.59110582998294 3.76205346577435 1.21860242258774 h
-3.83489298176092 4.36019764066847 -1.51534599625692 h
3.87216848865178 -0.38762296703046 0.85558003652882 f
$end

（3）$[UO_2(H_2O)_3F_2]_{meta}$ E_{RIMP2}= -1054.8160938680 Hartree

$coord
-0.06842061436643 -0.00282048388532 0.00150379009898 u
0.05284070509707 -0.01840915602090 3.38042223059159 o
-0.04917213160433 0.01283860462821 -3.38006968060786 o
3.94499530007062 -2.57624464467143 -0.13394299172377 o
5.15506489661879 -2.39755782024502 1.21481052599669 h
3.57469014510541 -4.35966933557734 -0.19642482984495 h
-4.77733952816150 -0.00251897708462 0.14081671407738 o
-5.61429403033857 1.47152577953663 -0.52243155135732 h
-5.63880182883498 -1.43074615431999 -0.58803569126158 h
-1.27265529103185 3.91787144644641 0.05584030259067 f
3.93359024435951 2.59458829319774 0.03869061883228 o
3.55371410817237 4.37560218011095 0.11057053752523 h
5.12189903903284 2.42465138259185 -1.33032183951428 h
-1.25728921739242 -3.92689669584776 -0.03554435778472 f
$end

（4）$[UO_2(H_2O)_3F_2]_{ortho}$ E_{RIMP2}= -1054.8156909690 Hartree

$coord
-0.03988972322323 -0.31515982460877 0.02147288732956 u
-0.13386188043817 -0.20927901927059 3.39617730856525 o
0.01970690648036 -0.08902860118610 -3.34808136065191 o
4.20184530833239 1.96741187633974 0.24168876681440 o
5.54131598320785 0.82929712787820 -0.23889668781867 h
4.49315642603126 3.49275382659250 -0.70787395517294 h
-2.47087765350415 -3.61728674789644 -0.09180316878178 f
-4.67330420120606 1.00290724957231 -0.14362309682927 o
-5.25190646243778 2.44690666937906 0.80141269599536 h
-5.75822983766950 -0.37431894500997 0.35218528750748 h
-0.56693071776242 4.38993633701167 0.10156843730035 o
-0.42378437809168 5.47589831425046 1.55430152368602 h
-0.86742059394401 5.48913447142096 -1.31686143669718 h

3.05746850802067 -3.00734595250966 0.04571220861709 f
$end

(5) $[UO_2H_2OF_3]^- H_2O$ E_{RIMP2}= -1078.3894623877 Hartree

$coord chem.pot.
0.40884819436009 -0.09230068358855 -0.16791667667633 u
1.50768378971330 0.68982273022673 -3.27979451736856 o
-0.81858656503316 -0.74247028650501 2.92694845991265 o
3.75955672384092 -2.34502888689596 0.56970596171321 f
-3.26443635232640 2.78005331381609 -1.00411409571477 o
-4.65507946001230 2.78110895996798 0.17025400910927 h
-2.67255694865650 4.55009240379527 -1.09229491804943 h
-1.98754020663774 -3.04703448799492 -1.73239494476365 f
-0.82603072087995 7.17100000051389 -0.79380823995278 o
0.54979137781397 6.17248753337834 -0.06818316904860 h
-0.15161534808130 7.88000236566663 -2.32568458302405 h
1.99926458670042 3.56877139074274 1.21517765740992 f
$end

(6) $[UO_2(H_2O)_2F_3]^-$ E_{RIMP2}= -1078.3881254349 Hartree

$coord
-0.00238365264440 0.15462737032267 0.00227486296652 u
-0.07027770800423 0.18668814996997 -3.39633227614235 o
0.06466115785484 0.20264585278429 3.40081542467283 o
4.51344488649230 1.94048713789837 -0.16931997446716 o
5.73544587660661 0.85923807323440 0.63706653681606 h
4.66519241869333 3.56034845206253 0.64658939661342 h
-4.52652198368711 1.92185075474832 0.16488075064164 o
-4.62541019974013 3.56972527390419 -0.60214334475818 h
-5.75908513546394 0.89618492675728 -0.69582599688330 h
-2.80474398959660 -2.92993349565629 0.03180043340529 f
2.83470810788877 -2.90124797449137 -0.01416634463019 f
-0.02499954278261 4.35741433700937 -0.00563568878317 f
$end

（7）$[UO_2(H_2O)F_3]^-$ E_{RIMP2}= -1002.0419588744 Hartree

$coord
0.44710320452188 -0.04771130137160 -0.15518896401699 u
1.47712006207253 0.73541833467295 -3.29949362075176 o
-0.79075745167059 -0.67528768107627 2.94889132207112 o
3.72702652821801 -2.44332523930184 0.49519632391036 f
-3.30776973353901 2.70300043720591 -0.89187244461351 o
-4.36779068120863 3.33046782403710 0.44779779872802 h
-3.40653351394119 3.94715755452405 -2.21654584473036 h
-1.88396803534723 -3.09346297583980 -1.68889329791429 f
2.28234686483716 3.36053412353406 1.27960592371676 f

$end

（8）$[UO_2F_4]^{2-}$ E= -1026.6659531105 Hartree

$coord

-0.48154005103873	0.55433765996816	-0.27156579189265	u
-1.27142941356562	-0.29263981219974	2.95980358060297	o
0.30837446186105	1.40127579847577	-3.50296623631666	o
1.02795426521721	4.19620836731828	1.05212457935115	f
-1.99114287770919	-3.08743031114593	-1.59543864003393	f
-4.23580833244041	2.28014462044796	-0.73715780445620	f
3.27296881857791	-1.17093117916023	0.19404929842271	f

$end

（9）$[UO_2F_5]^{3-}$ E= -1126.5567870050 Hartree

$coord

0.00009879661801	-0.00019263851592	0.00000342449942	u
0.00026533526897	-0.00003991954534	3.45136316160116	o
-0.00003594078690	-0.00019494032277	-3.45136829578592	o
3.94339564945142	-1.67698440021125	0.00051540467745	f
-4.17564905381050	-0.96144968798911	-0.00027981307242	f
-2.20447990005619	3.67417177396532	0.00078470959712	f
2.81342496134454	3.23194869347651	-0.00099339571853	f
-0.37606931582148	-4.26861381444196	-0.00003842388064	f

$end

（10）$[UO_2(H_2O)_4F]^+(H_2O)$ E_{RIMP2}= -1107.5814561390 Hartree

$coord

-0.35323998134839	-0.15068688292918	0.12432087002315	u
-0.54863885577134	-0.21847979803533	3.46671866903183	o
-0.40744672349798	-0.27633349125736	-3.22211932377133	o
-2.14420348232013	4.20483129041276	-0.04352844082412	o
-3.79975302124066	4.57209469319811	0.62072662874064	h
-1.06006584921101	5.54983379645743	0.53638136397516	h
3.55383606940045	-2.66878551297322	0.30545958318129	o
5.12806609130473	-1.77337365589236	-0.16727400812960	h
3.61849418933942	-4.31260506514848	-0.47305152239036	h
-1.37295237196432	-4.68159756999553	0.08076474482962	o
-1.78544128970543	-5.68724384537225	1.54201889678067	h
-1.99154301025984	-5.61339934452199	-1.35665049856308	h
-5.00454129543722	-0.21743853171433	0.09159486547062	o
-6.03475213234030	-0.66293117606068	1.52632616784562	h
-6.04425579151629	-0.50877269002094	-1.37540340626752	h
2.72922084773000	2.57289517474960	0.17039770423890	f

7.30753858828562 0.49007100504743 -0.61962561116453 o
8.01286680684700 0.59551724329013 -2.29253457539628 h
6.01119530715609 1.78880384429942 -0.56359044242826 h
$end

（11）$[UO_2(H_2O)_4F]^+(H_2O)_2$ E_{RIMP2}= -1183.9276034897 Hartree

$coord
0.24743223788221 0.57642456232603 0.25971356517532 u
-0.56587387562274 -0.66862948389297 3.25632712150356 o
0.89655997603935 1.80706960926164 -2.78287175538672 o
0.88922081172830 4.75768876441694 2.19806954751927 o
2.67026675485804 5.32969703918564 2.32936428791183 h
-0.08529348521797 6.19181590759624 1.64502588437091 h
1.76513451675268 -3.62597854241197 -0.95331377896295 o
3.21008787929647 -4.17249683008712 0.12074484812228 h
2.26427373240364 -3.94562252013205 -2.67627807201910 h
-3.10356372076247 -2.12102237959121 -1.74038043185264 o
-2.99040180016009 -3.92110635596772 -1.47846280332802 h
-4.86033278039177 -1.70086346270167 -1.50067769072395 h
-3.77958799191129 2.87144074655070 0.15998719543570 o
-4.96713386932692 3.03985941639617 1.53116319658598 h
-4.40740751521527 3.93011985003186 -1.18297785046846 h
4.26467188975496 0.69312852556190 1.35001098887884 f
5.44115288023532 -4.23978276935303 2.29971398099929 o
7.02586211128787 -4.97324342706604 1.78944606302005 h
5.72200642441493 -2.42821530741450 2.28073783080839 h
5.81644057675209 5.47742791294653 2.30939434999066 o
6.58830724143587 5.83420891315735 3.91736270543896 h
5.95986819267077 3.66016156954813 2.10551176264880 h
$end

（12）$[UO_2(H_2O)_3F_2](H_2O)_3$ E_{RIMP2}= -1283.8520627938 Hartree

$coord
0.01223681917922 -0.38108228729092 0.08957551536839 u
0.03238504180587 -0.74247505946470 -3.25392112525154 o
-0.00679975129596 -0.27854883585594 3.44923279610318 o
-2.70942134498986 2.81851745403848 -0.13817632889331 f
0.00833994641272 6.97635282738738 1.66100858163159 o
-1.39644331982618 5.91050351098367 1.18169987144435 h
-7.41624404432714 1.56059418163455 -1.32900183592922 o
-5.85588771807882 2.52164456326807 -1.14871109779116 h
5.85295463598797 2.50562136000955 -1.22420097812992 h
1.40684988247947 5.90379864217067 1.17725235882641 h
-4.38802294742882 -2.14476058635191 0.27079169618814 o
-5.71919660722380 -1.05430547099609 -0.44461495279004 h

-4.68670536767998 -3.81937616345154 -0.37340112531259 h
-7.84502029321214 1.67658255161052 -3.09196864367307 h
0.02787796332747 -5.09158328967397 0.32720711205370 o
-0.00735970449752 -6.10440424385484 1.83921243677230 h
0.03149791456075 -6.26674201056639 -1.06270943602466 h
4.42135736739748 -2.11947383961696 0.32585593020129 o
5.73907818345819 -1.05201475732697 -0.44784573376043 h
4.71352185276614 -3.81673680129667 -0.25902447991562 h
7.29233618895604 1.47894411570718 -1.73808836645058 o
8.69465545723331 2.11127818676211 -0.76883433472718 h
2.73046664092693 2.82253054613180 -0.12398185504376 f
$end

（13）$[UO_2(H_2O)_3F_2](H_2O)_4$ E_{RIMP2}= -1360.2044184390 Hartree -7.52 cm^{-1} was obtained

$coord
0.02438172442310 -0.30895471339889 0.06954278724524 u
0.18712866226709 -1.01863653469217 3.35007820987024 o
-0.11526636469331 0.30549865789480 -3.23169713175375 o
4.08224085174461 0.96948247191905 0.16095707557111 f
-4.04838467847174 0.85606117277977 0.45885406837959 f
4.44961503982530 5.90126434790323 -0.86684058895810 o
4.87075694375870 4.13795045300503 -0.55284082972086 h
-7.31133492087145 -2.96803723989196 0.31822118751086 o
-6.60820267671845 -1.26787296872950 0.37813507255405 h
7.20920935544232 -2.93331236780169 0.68114986437285 o
6.56895972854360 -1.21459747056057 0.51925672079106 h
-4.62980558063691 5.78225555449354 -0.51428828235472 o
-4.97686490373652 4.00791630609281 -0.17426332405685 h
-0.00374555397767 4.26619168612561 1.00118459718913 o
-1.53693793274929 5.16851767456196 0.46989586653795 h
1.45940885078821 5.20880161968245 0.35465154455738 h
-5.65725585134433 6.69807384441020 0.67362999342799 h
5.54263040702797 6.84822243771936 0.23492779497383 h
2.56517225931809 -4.21941706426740 -0.69284729598156 o
4.38920809692559 -4.03264332920380 -0.31935897102764 h
2.47005507075626 -4.99880951972400 -2.33546620675444 h
8.55541267854296 -3.05552618208951 -0.53531635015012 h
-2.54732489502490 -4.20720663643441 -0.68985450991647 o
-4.38147567400377 -4.05455485410177 -0.36922888550160 h
-2.00332986774839 -5.72435747999429 0.15579962645921 h
-8.47963331261852 -2.95002784245187 -1.07499338461767 h
$end

（14）$[UO_2(H_2O)F_3]^-(H_2O)_4$ E_{RIMP2}= -1307.4188811686 Hartree

$coord

0.91574460272709	0.12482600959940	-0.34048736891824	u
1.80868065358471	-0.00295126298877	-3.59240263612379	o
-0.02565927108944	0.28976433212474	2.89656065336792	o
4.16274790465905	-2.28988340791150	0.68750331844911	f
7.71669124849447	1.75592428227036	1.27148744323314	o
6.91892255551216	0.11479689707186	1.18934583430779	h
-5.05576447250578	-1.79426108149704	-0.31470644697771	h
0.01366287967098	-6.01089524421348	-0.04497047567113	h
6.32999767064995	2.89354183168189	0.92819240673321	h
-2.72923407783224	2.84049455348636	-1.44779337749787	o
-4.30644369245162	2.04072423086819	-0.88135582723960	h
-2.58300000275367	4.49742686210542	-0.61780686109993	h
-6.37006924154359	-0.51115023151653	-0.25324651893303	o
1.46405428664876	-6.97935363387332	0.49683539016828	o
2.75073152773495	-5.69803563706187	0.69469484333351	h
-7.06268711098237	-0.62857619276835	1.42354587020107	h
-1.88549391826804	-2.95591677891852	-0.94069369135019	f
-1.09142934244958	7.05947870624062	0.87789428637287	o
0.59736711645241	6.36026161654920	0.68540418497566	h
-1.06802982141893	8.60366536817367	-0.08188668042721	h
2.84760231729212	3.83659608967731	0.07998859103628	f

$end

（15）$[UO_2F_4]^{2-}(H_2O)_4$ E= -1332.1213709365 Hartree

$coord

-0.23214961040154	0.69848787217735	-0.13154155784012	u
0.50748591764007	0.62489540735595	3.18446346029548	o
-0.97304983568184	0.76989949918749	-3.44671589676580	o
2.78758048073973	-2.14636576061663	-0.88251324206397	f
-3.02488874479793	-2.40772048893113	0.42749549964488	f
-3.25234909681200	3.54372022560126	0.60561222003454	f
2.55447877378154	3.81006955320518	-0.70786148227365	f
-0.12140549059730	-6.54670067737145	-1.09852825163245	o
-1.49592924285623	-5.47384317369685	-0.54831506311238	h
1.25449177551944	-5.34419978773475	-1.16531454398111	h
-0.79443289758628	7.87108602669863	-1.17171708974762	o
0.69869249884761	6.81568544785440	-1.17288574365793	h
-2.05125747079128	6.69891195594567	-0.54780453033008	h
6.37168708959330	1.04121506164217	-3.09807867415753	o
5.33908981109393	2.39495953421038	-2.43179639194565	h
5.44773963670411	-0.42470519520117	-2.51460243783438	h
-7.09225893287091	0.36236701831625	2.34396572808855	o
-6.09843757478553	1.82657080488157	1.88390063915869	h
-5.98826721250966	-0.99217133168862	1.80589594980653	h

$end

（16）$[UO_2F_5]^{3-}(H_2O)_5$ E_{RIMP2}=-1507.2284447222 Hartree

$coord

-0.06515694653928	0.03723782973232	-0.38398592659355	u
0.44226474534307	0.42954222632036	2.94749660423128	o
-0.56992670327574	-0.35375381714380	-3.72061786592445	o
3.56006565489201	-2.42897266521622	-0.64676923914766	f
-4.39432487800848	-0.09843968920711	0.28070603043745	f
-1.60929404650298	4.14289716157612	-0.63019408747793	f
3.31897769144071	2.70600988485461	-1.21808743482343	f
-1.21440536689651	-4.13924854148389	0.27608028373451	f
7.41348292339770	0.51174248875130	0.73758075442763	o
6.22283855888736	1.77065406846459	0.12003578325479	h
6.35230070208467	-0.95791522811318	0.42471066737664	h
-5.61370243535524	4.28599888869418	2.44360052941642	o
-4.13622510163673	4.75728278924792	1.45380112134327	h
-5.62530891923621	2.50588424637610	1.98178097052877	h
-5.93708161276863	-4.57748175845453	-1.44708302682817	o
-5.89812961169269	-2.80075236135363	-0.97140041996528	h
-4.20401936784576	-4.96955910415326	-0.97170848688278	h
2.76648760411793	-6.35011406452659	2.43526541462638	o
3.56929343686429	-5.02460641977926	1.44364316293297	h
1.03255514373480	-5.94630570096419	1.97403319113987	h
2.15183207752079	7.12747935519878	0.91379419767335	o
3.09999157988450	5.69898482818426	0.24816314182899	h
0.47606176244313	6.45878910139965	0.55520373211908	h

$end

2. Additional Explanations

2.1 Imaginary frequency for $[UO_2(H_2O)_3F_2](H_2O)_4$

For $[UO_2(H_2O)_3F_2](H_2O)_4$ a single imaginary value of 7.52i cm^{-1} appeared in the numerical evaluation of the vibrational frequencies at the MP2 level when COSMO was included to model the aqueous solution. This is most likely an artefact due to the combination of COSMO and numerical frequency evaluation, which in this case also could not be avoided by changing various computational parameters as suggested in the Turbomole manual. The problem does not appear at the MP2 level for the gasphase, i.e. without COSMO, and also not at the DFT level with and without COSMO. The imaginary frequency mainly corresponds to the simultaneous stretching of two F…H_2O hydrogen bridges connecting the first and second coordination sphere. The symmetric and antisymmetric O=U=O stretching frequencies were found to be 850 and 927 cm^{-1},

respectively, in comparison to values of 848 and 927 cm^{-1} for $[UO_2(H_2O)_3F_2](H_2O)_3$ where one water molecule less is treated explicitly in the second hydration sphere and no imaginary frequency resulted.

2.2 Addition of water molecules

Initially it was attempted to add water molecules one at a time to the $UO_2F_n^{2-n}$ complexes and to build up the first (F, H_2O) and then the second (H_2O) coordination sphere systematically by searching for the minimum energy structure for each case, before adding to this the next water molecule. Two problems made this strategy impracticable and led to the prescription described in the main text:

1) There are in most cases multiple possibilities to add a single water molecule and the necessary search for the minimum energy structure requires often several lengthy geometry optimizations, all but one of them finally related to a "dead end".
2) It is quite often not possible to first construct the first coordination sphere, and after completion of this task the second. Even for a small number of F^- ligands the added water molecules prefer to go to the second coordination sphere where they can form hydrogen bridges with the F^- ligand(s). In a certain way this spoils a systematic construction strategy and moreover it leads to too numerous possibilities to add the next water molecule (first or second coordination sphere?).

2.3 Structure of $UO_2(H_2O)F_4^{2-}$ complex

The construction of a UO_2^{2+} complex with four F^- ligands and one H_2O ligand in the first coordination sphere was attempted. Addition of up to totally six water molecules was investigated. In all cases, independent from the starting geometry, the structure optimizations led to (local/global) minimum structures were all water molecules are located in the second coordination sphere, i.e., despite the "smallness" of the F^- ligand a direct coordination of the water O to U was not observed. Table S7 summarizes selected results for the addition of the first water molecule.

Table S7. Selected def2-TZVPP U SPP results for $UO_2F_4^{2-}$ and $UO_2F_4^{2-}$ + H2O

	-E(a.u.)	R(U=O)	R(U-O)	R(U-F)	R(F…H)	ν(cm-1)
$[UO_2F_4]^{2-}$						
MP2,COSMO	1026.665953	1.817	-	2.201	-	794.94
						851.71
MP2,gasphase	1026.352958	1.823	-	2.219	-	781.97
						868.95
BP86,COSMO	1027.988069	1.848	-	2.197	-	742.34
						770.03

BP86,gasphase	1027.675010	1.848	-	2.222	-	742.09
						801.81
Formation of $[UO_2F_4(H_2O)]^{2-}$ attempted, but $[UO_2F_4](H_2O]^{2-}$ resulted from all optimizations.						
MP2,COSMO	1102.736188	1.811	3.865	2.190	1.827	801.92
				2.230		862.9
MP2,gasphase	1104.476846	1.815	3.939	2.202	1.815	793.59
				2.253		886.32
BP86,COSMO	1104.476846	1.840	3.923	2.187	1.866	751.52
				2.227		783.4
BP86,gasphase	1104.181942	1.838	3.972	2.204	1.842	755.60
				2.260		822.2

The listed distances partly refer to averages and are just meant to get a rough impression of the structure. In case of R(U-F) the two values refer to F atoms without (shorter) and with (longer) hydrogen bonds to water.