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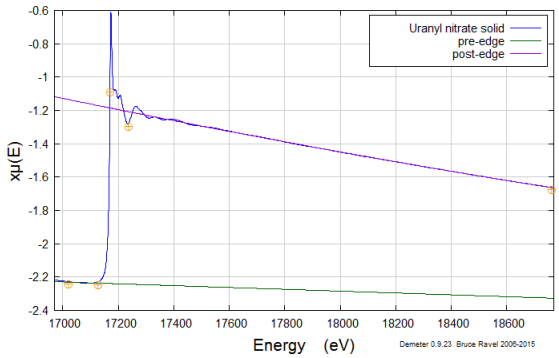
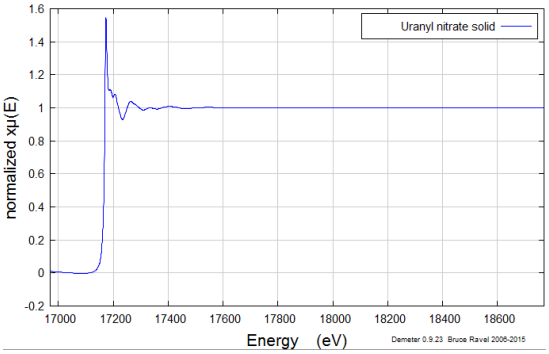
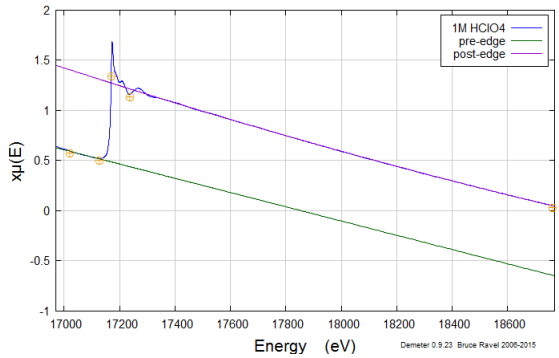
Supporting information for article:

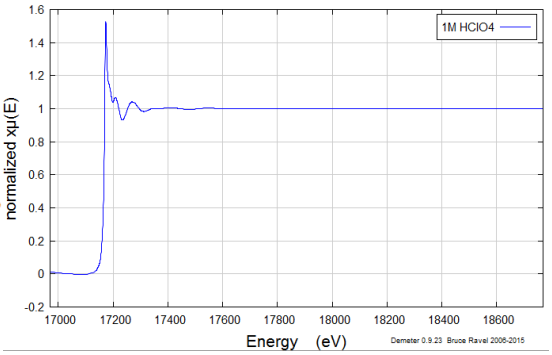
Extraction of Local Coordination Structure in Low Concentration Uranyl System by X-ray Absorption Near-edge Structure

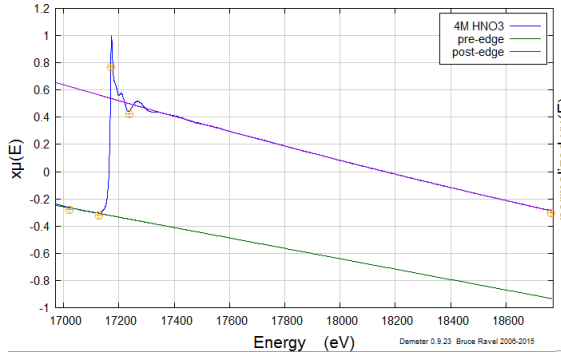
Linjuan Zhang, Jing Zhou, Jianyong Zhang, Jing Su, Shuo Zhang, Ning Chen, Yunpeng Jia, Jiong Li, Yu Wang and Jian-Qiang Wang

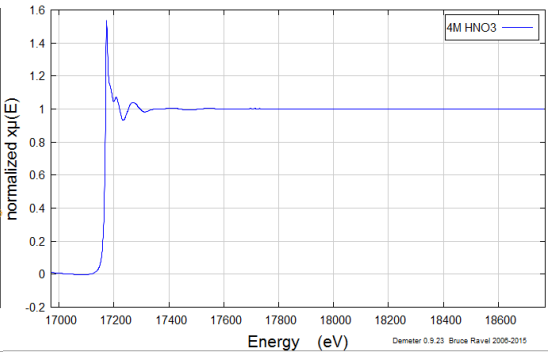

Uranyl nitrate solid
pre-edge
post-edge
χμ(E)
Energy (eV)
Demeter 0.9.23 Bruce Ravel 2006-2015

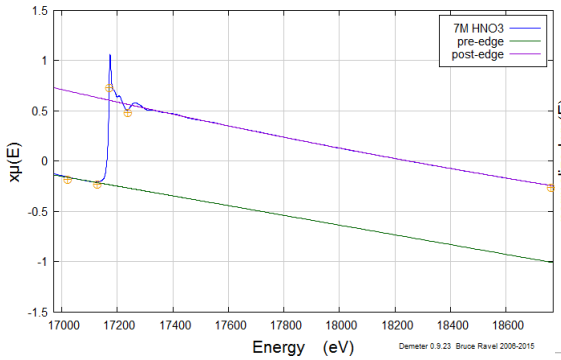

Uranyl nitrate solid
normalized χμ(E)
Energy (eV)
Demeter 0.9.23 Bruce Ravel 2006-2015

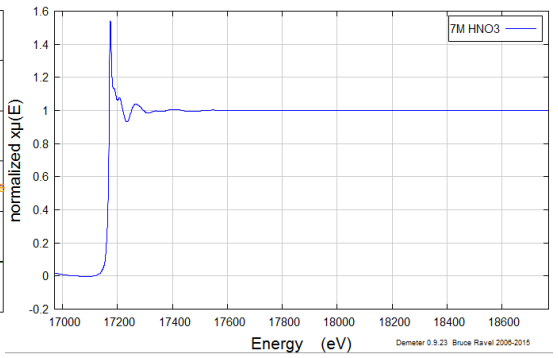

1M HClO4
pre-edge
post-edge
χμ(E)
Energy (eV)
Demeter 0.9.23 Bruce Ravel 2006-2015

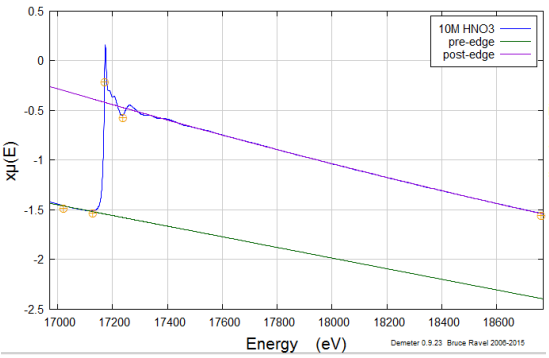

1M HClO4
normalized χμ(E)
Energy (eV)
Demeter 0.9.23 Bruce Ravel 2006-2015


4M HNO3
pre-edge
post-edge
χμ(E)
Energy (eV)
Demeter 0.9.23 Bruce Ravel 2006-2015


4M HNO3
normalized χμ(E)
Energy (eV)
Demeter 0.9.23 Bruce Ravel 2006-2015


7M HNO3
pre-edge
post-edge
χμ(E)
Energy (eV)
Demeter 0.9.23 Bruce Ravel 2006-2015


7M HNO3
normalized χμ(E)
Energy (eV)
Demeter 0.9.23 Bruce Ravel 2006-2015


10M HNO3
pre-edge
post-edge
χμ(E)
Energy (eV)
Demeter 0.9.23 Bruce Ravel 2006-2015

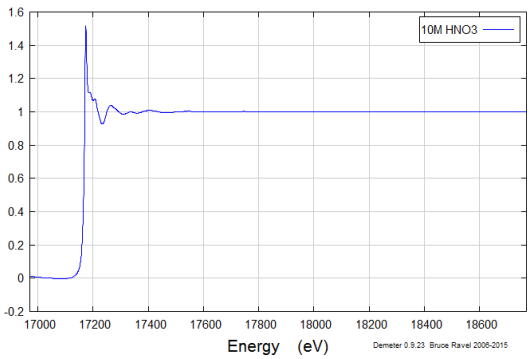

10M HNO3
normalized χμ(E)
Energy (eV)
Demeter 0.9.23 Bruce Ravel 2006-2015

Figure S1 Left panel show the raw data of U L_3-edge XAS, as well as the pre- and post-edge line. Right panel show the normalized uranium L_3-edge x-ray absorption spectra of uranyl species.

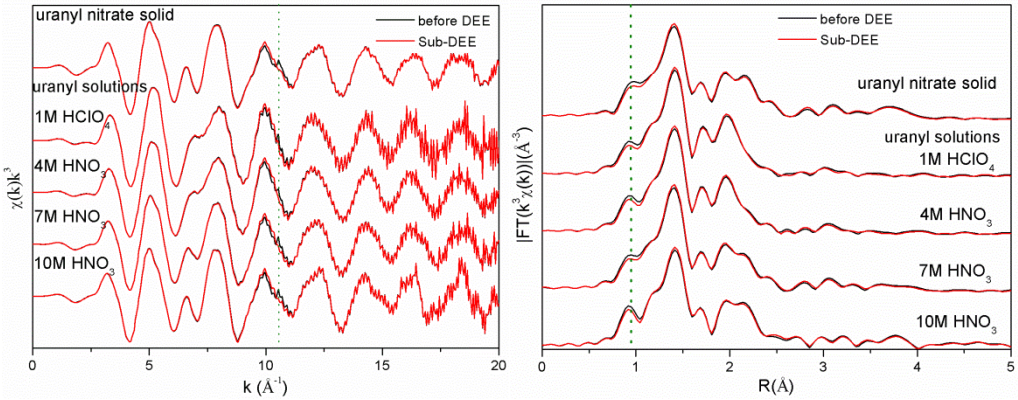


Figure S2 Comparison of experimental uranium L_3-edge k^3-weighted EXAFS oscillation data (a) and Fourier Transform data (b) before and after subtracting the double-electron exciation.

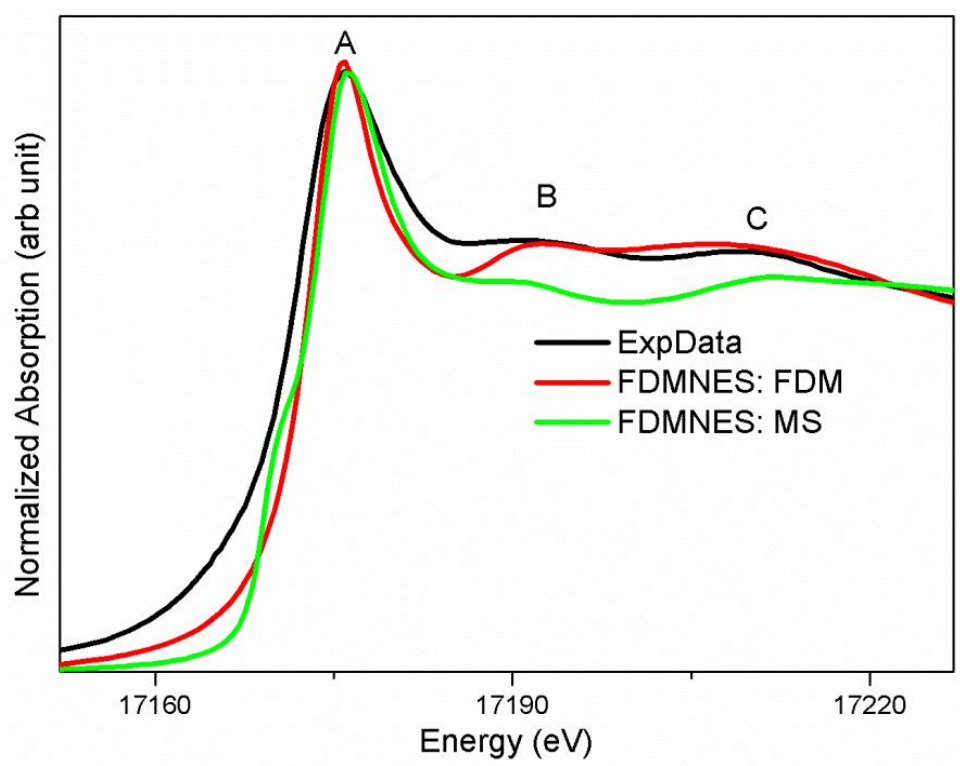


Figure S3 Uranium L_3 edge XANES spectra in the $UO_2(NO_3)_2(H_2O)_2$ solid. Experiment (black line), FDM calculation (red line) and MS calculation based on the muffin-tin approximation (green line). MS and FDM calculations are comparable, but the edge jump and continuum resonance part of XANES ExpData can be better reproduced by FDM calculation.

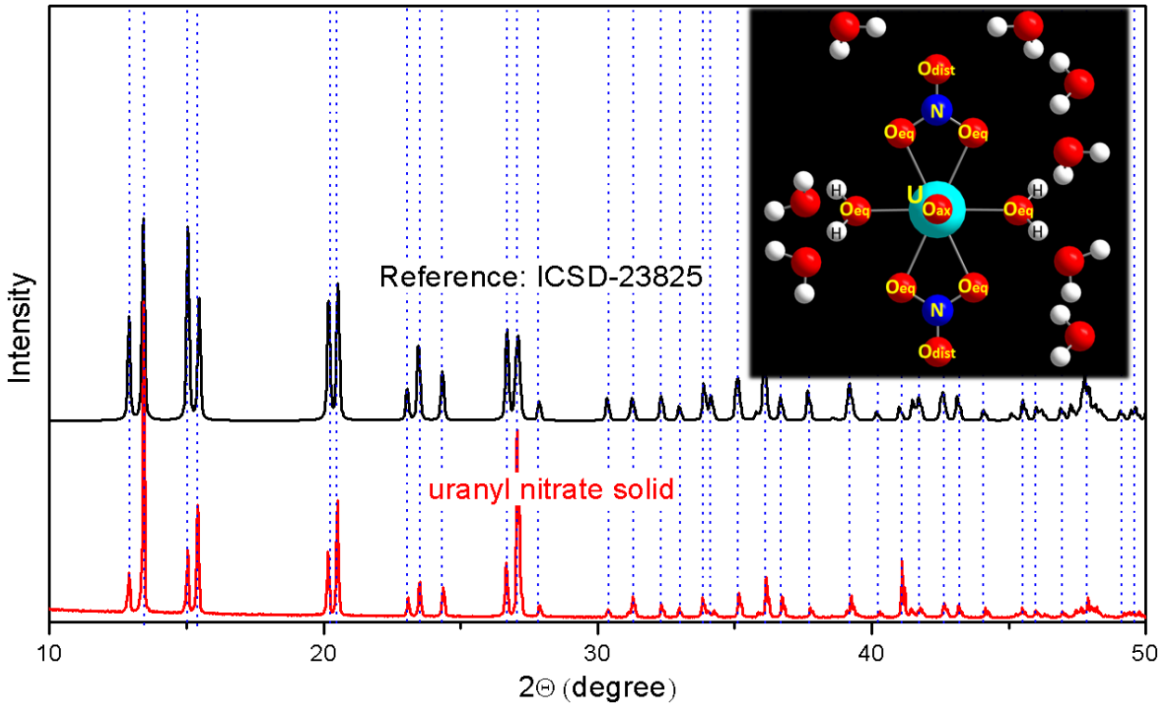


Figure S4 X-ray diffraction patterns of $UO_2(NO_3)_2(H_2O)_2$ solid powder and referenced standard model of ICSD-23825. The inset shows the ball-and-stick figure of uranyl nitrate species (ICSD-23825). The uranyl moiety (UO_2) is made up of a uranium atom bound to two axial oxygen (O_{ax}) atoms above and below the uranium atom.

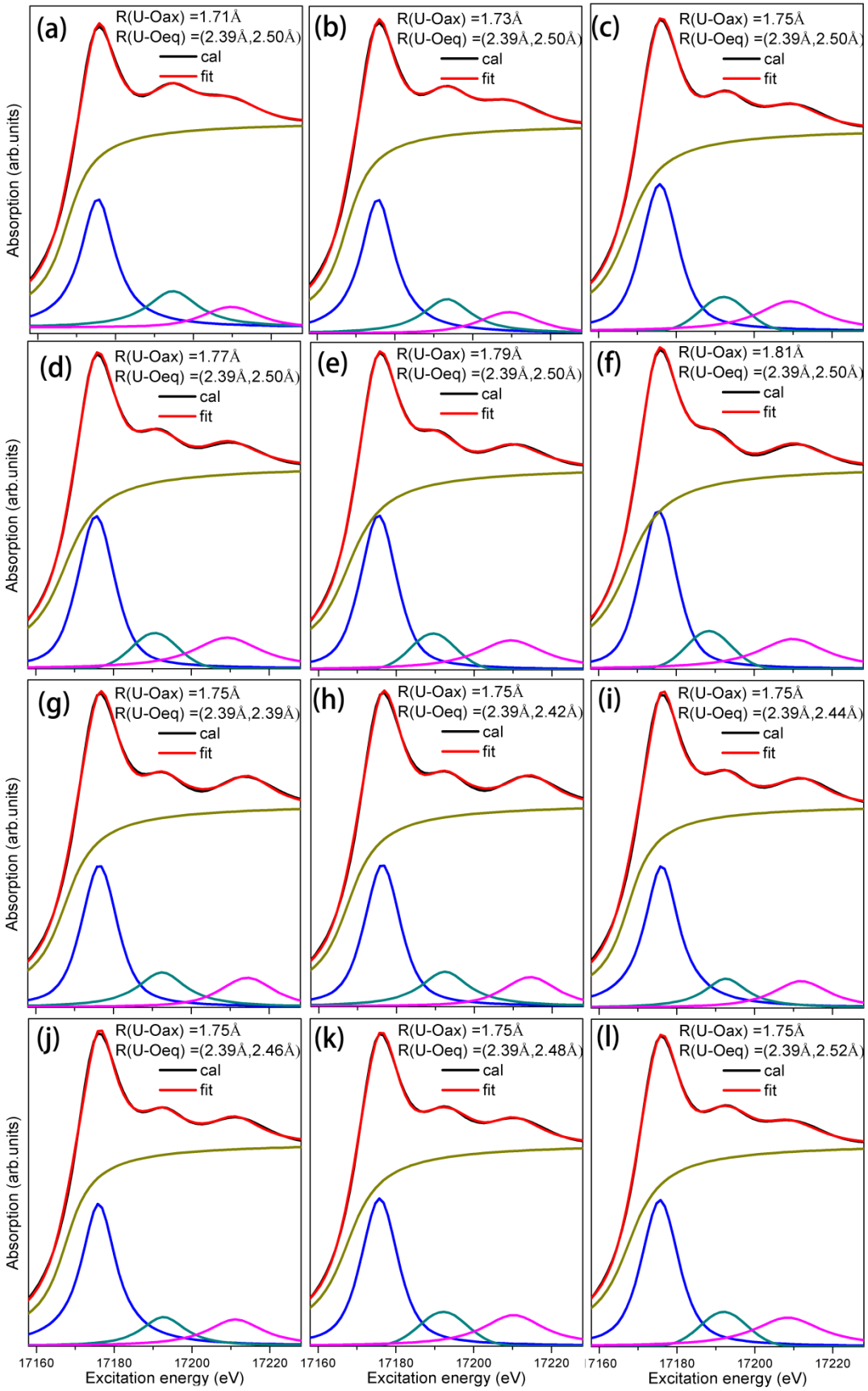

(a)
R(U-Oax) =1.71Å
R(U-Oeq) =(2.39Å,2.50Å)
cal
fit
(b)
R(U-Oax) =1.73Å
R(U-Oeq) =(2.39Å,2.50Å)
cal
fit
(c)
R(U-Oax) =1.75Å
R(U-Oeq) =(2.39Å,2.50Å)
cal
fit
(d)
R(U-Oax) =1.77Å
R(U-Oeq) =(2.39Å,2.50Å)
cal
fit
(e)
R(U-Oax) =1.79Å
R(U-Oeq) =(2.39Å,2.50Å)
cal
fit
(f)
R(U-Oax) =1.81Å
R(U-Oeq) =(2.39Å,2.50Å)
cal
fit
(g)
R(U-Oax) =1.75Å
R(U-Oeq) =(2.39Å,2.39Å)
cal
fit
(h)
R(U-Oax) =1.75Å
R(U-Oeq) =(2.39Å,2.42Å)
cal
fit
(i)
R(U-Oax) =1.75Å
R(U-Oeq) =(2.39Å,2.44Å)
cal
fit
(j)
R(U-Oax) =1.75Å
R(U-Oeq) =(2.39Å,2.46Å)
cal
fit
(k)
R(U-Oax) =1.75Å
R(U-Oeq) =(2.39Å,2.48Å)
cal
fit
(l)
R(U-Oax) =1.75Å
R(U-Oeq) =(2.39Å,2.52Å)
cal
fit
Absorption (arb.units)
17160
17180
17200
17220
Excitation energy (eV)

Figure S5 The calculated U L_3 edge XANES spectra of $UO_2(NO_3)_2(H_2O)_2$ model and their pseudo-Voigt line shapes by least-squares fitting in the linear combination analyses. (a-f) the axial bonds have been elongated from 1.71 to 1.81 Å, the equatorial bonds from the contribution of H_2O and NO_3 ligands were kept fixed at 2.39 and 2.50 Å, respectively. (g-l) the equatorial bonds of U-O_{eq}(NO_3) have been elongated from 2.39 to 2.52 Å, the bond lengths of U-O_{eq}(H_2O) and U-O_{ax} were kept fixed at 2.39 and 1.75 Å, respectively.

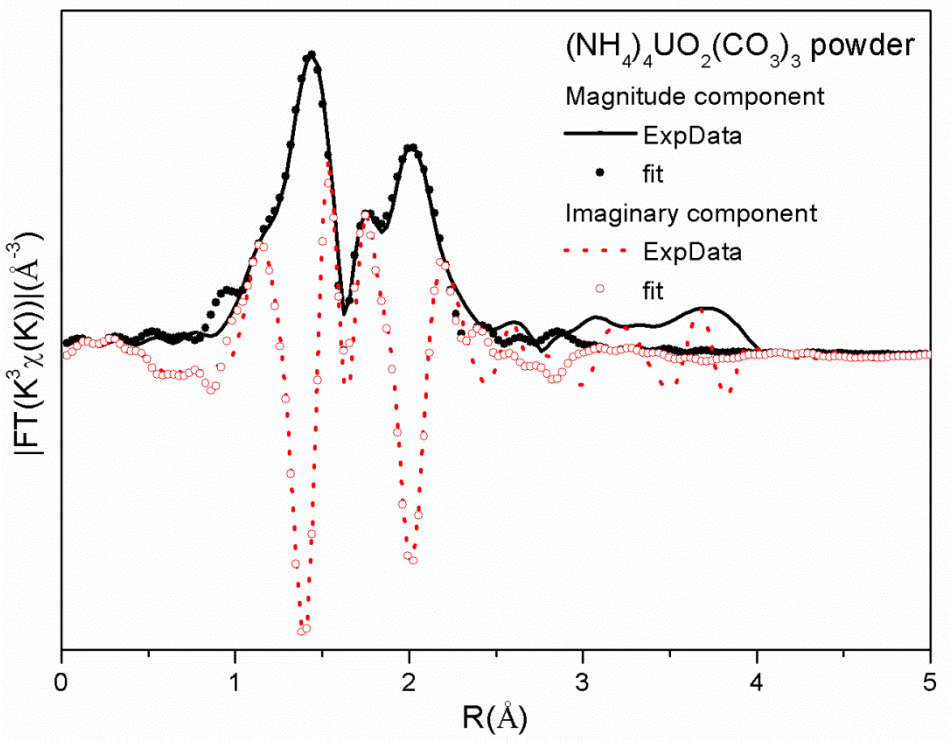


Figure S6 Experimental magnitude and imaginary components of the Fourier Transform at the U L_3-edge for $(NH_4)_4UO_2(CO_3)_3$ powder and its corresponding fits after subtracting the double-electron excitation.

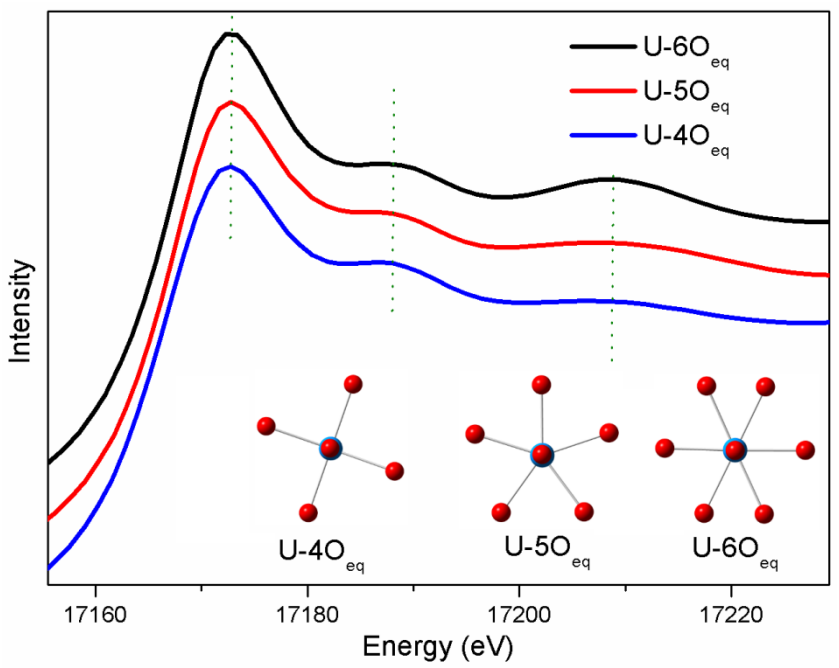


Figure S7 The comparison of the position of peak A, B and C affected by all three possible symmetries with same coordination distance ($R_{U\text{-}Oax}$=1.77Å, $R_{U\text{-}Oeq}$=2.42Å) and different coordination number.

Table S1 Comparison of local structure information between ICSD-23825 standard model from XRD and $UO_2(NO_3)_2(H_2O)_2$ solid powder by EXAFS fitting, as well as the parameters of the fit of $(NH_4)_4UO_2(CO_3)_3$ powder. (CN is the coordination number. *R* is the distance between the uranium absorber and surrounding coordination atoms. σ^2 is the mean square disorder. R-factor reflects the quality of the fit.)

Sample	Bond Type	CN	*R*(Å)	$\sigma^2\times10^{-3}$(Å^2)	R-factor
ICSD-23825	$U\text{-}O_{ax}$	2	1.75, 1.77	-	-
	$U\text{-}O_{eq}(H_2O)$	2	2.40		
	$U\text{-}O_{eq}(NO_3)$	2	2.50		
		2	2.54		
$UO_2(NO_3)_2(H_2O)_2$	$U\text{-}O_{ax}$	2	1.75±0.02	1.7±0.1	0.01
	$U\text{-}O_{eq}(H_2O)$	2.0±0.2	2.39±0.02	4.6±0.3	
	$U\text{-}O_{eq}(NO_3)$	3.7±0.3	2.50±0.02	5.6±0.7	
$(NH_4)_4UO_2(CO_3)_3$	$U\text{-}O_{ax}$	2	1.80±0.02	2.1±0.2	0.01
	$U\text{-}O_{eq}(CO_3)$	5.8±0.3	2.45±0.02	5.9±0.4	

Table S2 Energy position values determined for the resonant feature (A, B, C) and the arctangent (atan) function to model the edge jump, as well as the energy separation (ΔE) between the WL and the continuum resonance by using the linear combination least-squares fit modeling (WINXAS program).

	Position of Peak A	Position of Peak B	Position of Peak C	ΔE_1 (±0.02) (peakB-peakA)	ΔE_2 (±0.02) (peakC-peakA)	Position of Atan
$UO_2(NO_3)_2(H_2O)_2$ - solid	17175.21	17191.63	17208.56	**16.42**	**33.35**	17172.37
UO_2^{2+} in 1M-$HClO_4$	17174.77	17189.66	17211.60	**14.89**	**36.83**	17172.37
UO_2^{2+} in 4M-HNO_3	17175.25	17190.23	17211.49	**14.98**	**36.24**	17172.37
UO_2^{2+} in 7M-HNO_3	17175.30	17190.73	17209.45	**15.43**	**34.15**	17172.37
UO_2^{2+} in 10M-HNO_3	17175.14	17191.07	17208.09	**15.93**	**32.95**	17172.37
$(NH_4)_4UO_2(CO_3)_3$ powder	17174.89	17188.45	17209.8	**13.56**	**34.91**	17172.37
UO_2^{2+}-AO complex	17174.54	17189.67	17212.25	**15.13**	**37.71**	17172.37
Cal-R(U-O_{ax}) =1.71Å	17175.41	17194.74	17209.62	**19.33**	34.21	17167.5
Cal-R(U-O_{ax}) =1.73Å	17175.59	17193.36	17209.30	**17.77**	33.71	17167.5
Cal-R(U-O_{ax}) =1.75Å	17175.72	17192.12	17209.21	**16.40**	33.49	17167.5
Cal-R(U-O_{ax}) =1.77Å	17175.53	17190.61	17209.18	**15.08**	33.65	17167.5
Cal-R(U-O_{ax}) =1.79Å	17175.44	17189.48	17209.46	**14.04**	34.02	17167.5
Cal-R(U-O_{ax}) =1.81Å	17175.35	17188.40	17209.80	**13.05**	34.45	17167.5
Cal-R(U-O_{eq}) =(2.39, 2.52Å)	17175.62	17192.00	17208.44	16.38	**32.82**	17167.5
Cal-R(U-O_{eq}) =(2.39, 2.50Å)	17175.72	17192.12	17209.21	16.40	**33.49**	17167.5
Cal-R(U-O_{eq}) =(2.39, 2.48Å)	17175.81	17192.20	17210.20	16.39	**34.39**	17167.5
Cal-R(U-Oeq) =(2.39, 2.46Å)	17175.98	17192.66	17211.13	16.68	**35.15**	17167.5
Cal-R(U-Oeq) =(2.39, 2.44Å)	17176.10	17192.58	17211.93	16.48	**35.83**	17167.5

Cal-R(U-Oeq) =(2.39, 2.42Å)	17176.21	17192.44	17212.90	16.23	**36.69**	17167.5
Cal-R(U-Oeq) =(2.39, 2.39Å)	17176.36	17192.40	17214.30	16.04	**37.94**	17167.5