Supporting Information

Ultrasonically assisted conversion of uranium trioxide into uranium(VI) intrinsic colloids

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Figure S1

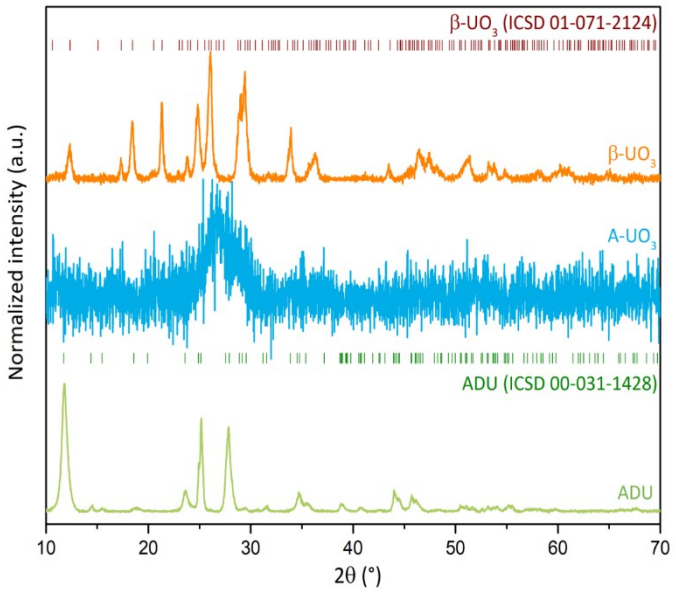

Fig. S1: Normalized PXRD diagrams (background corrected) acquired on ADU precursor (green), A-UO_3 powder (blue) and β-UO_3 powder (orange).

Figure S2

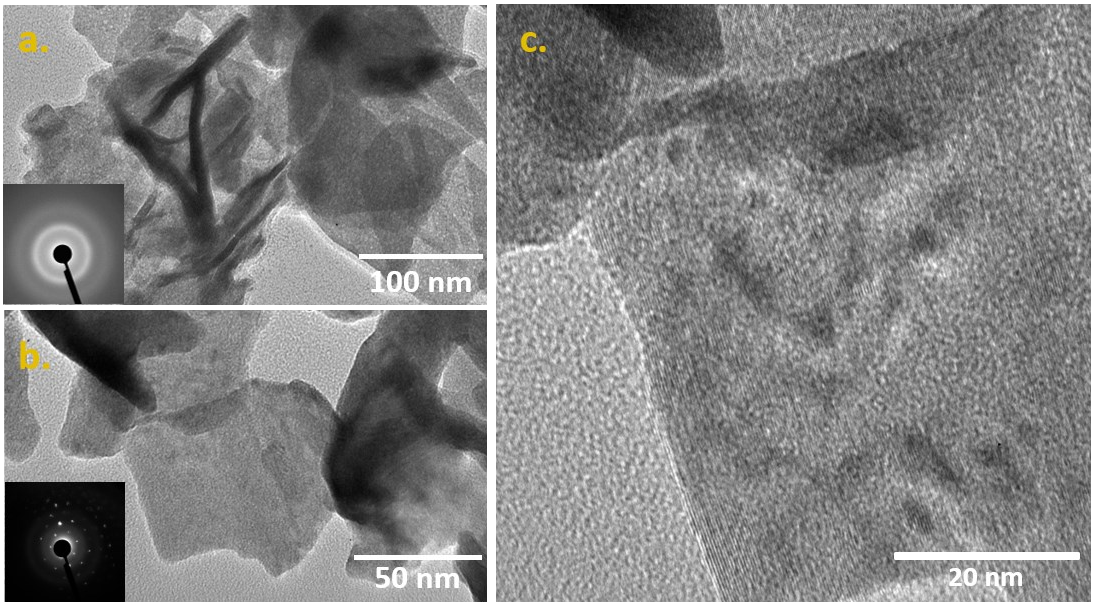

Fig. S2: HR-TEM analyses of U(VI) oxide powders obtained after calcination of the ADU precursor. Insets are the selected-area electron diffraction (SAED) patterns collected for each powder. (a) A-UO_3 powder, (b) - (c) β-UO_3 powder at different magnifications.

Figure S3

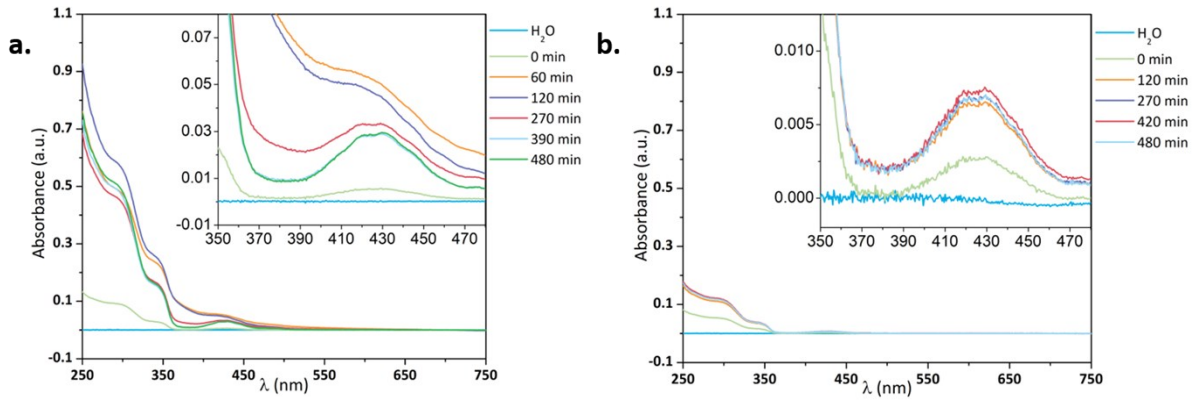

Fig. S3: UV-Vis absorption spectra observed during the treatment of β-UO_3 in pure water under (a) sonication at 20 kHz ultrasound (20 °C, Ar/(10%)CO, 0.35 $W.mL^{-1}$) and (b) mechanical stirring (silent conditions).

Figure S4

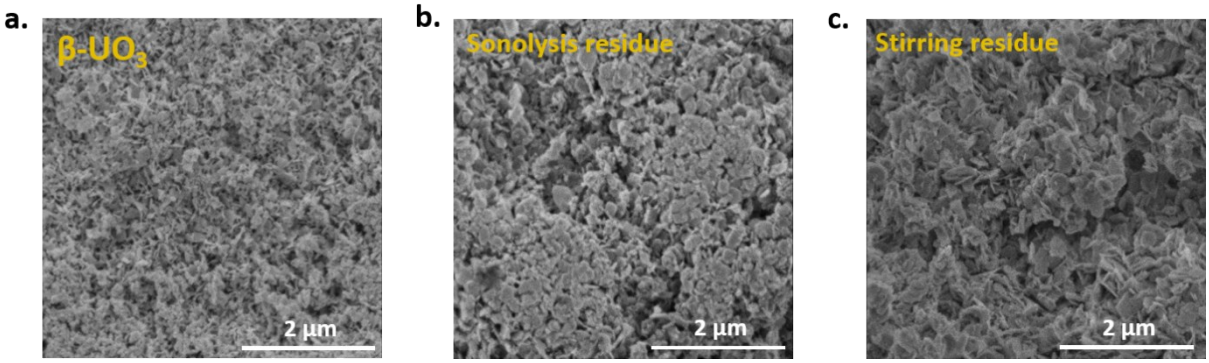

Fig. S4: SEM images of β-UO_3 (a) before treatment, (b) after sonication (20 kHz, 20 °C, Ar/(10%)CO, 0.35 W.mL^{-1}) and (c) after mechanical stirring in silent conditions.

Figure S5

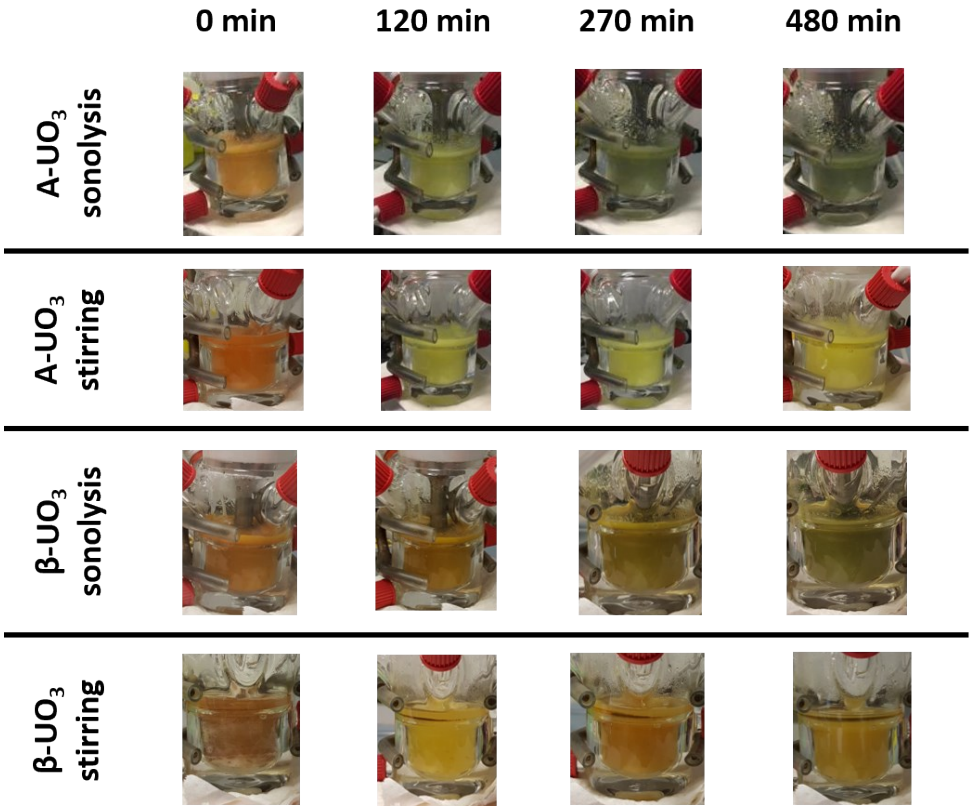

Fig. S5: Photos of the glass reactor during UO_3 powder treatments by sonication at 20 kHz ultrasound (20 °C, Ar/(10%)CO, 0.35 W.mL^{-1}) and mechanical stirring (silent conditions).

Figure S6

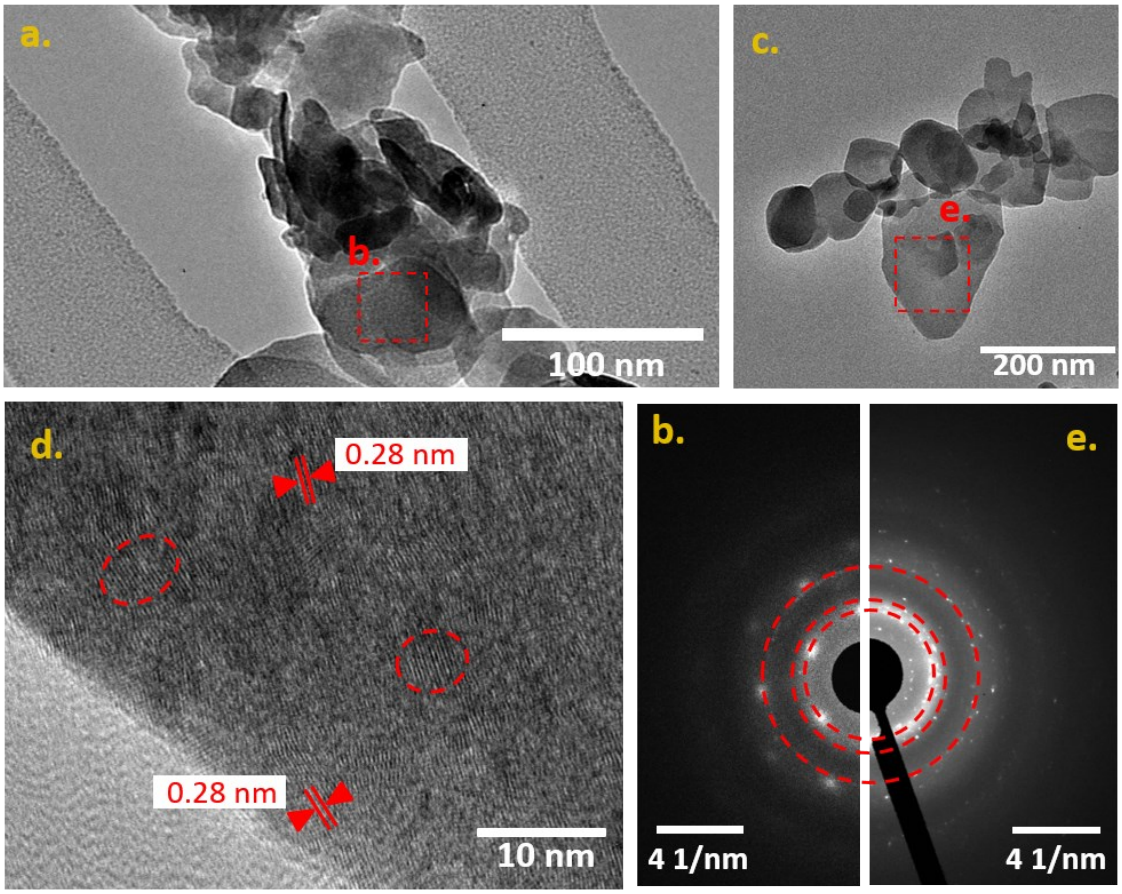

Fig. S6: HR-TEM analyses of the neoformed solids obtained after treatment of β-UO_3 powder. Dashed squares represent the selected areas corresponding to an analysis by electron diffraction (SAED). Dashed circles denote individual nanoparticles. (a) Sonolysis residue, (b) SAED pattern of an aggregate formed during sonolysis, (c) - (d) stirring residue at different magnifications and (e) SAED pattern of a polycrystalline nanoparticle aggregate formed during mechanical stirring. In panels (b) and (e), dashed circles correspond to UO_2 diffraction rings.[1]

Table S1

Table S1: d_n spacing values (for $1 \leq n \leq 4$) of theoretical crystals (UO_2, schoepite, *meta*-schoepite), neoformed solids obtained after UO_3 treatments and A-UO_3 sonolysis supernatant.[1] For schoepite and *meta*-schoepite, d_n values are calculated from corresponding ICSD patterns (ICSD 00-013-0241 and ICSD 01-070-4765, respectively). For experimental samples, d_n values are measured on HR-TEM SAED patterns. The error is estimated equal to ± 0.2 Å.

	UO_2	d_n (Å)	3.15	2.73	1.93	1.65
	Schoepite	d_n (Å)	7.21	4.38	3.59	3.55
	Meta-schoepite	d_n (Å)	8.48	8.17	7.19	6.32
HR-TEM	A-UO_3 sonolysis residue	d_n (Å)	3.41	2.95	2.09	1.78
		d_1/d_n (Å)	-	1.16	1.63	1.92
	A-UO_3 sonolysis supernatant	d_n (Å)	3.14	2.74	1.89	1.66
		d_1/d_n (Å)	-	1.15	1.66	1.90
	A-UO_3 stirring residue	d_n (Å)	3.33	2.90	2.06	1.75
		d_1/d_n (Å)	-	1.15	1.62	1.91
	β-UO_3 sonolysis residue	d_n (Å)	3.36	2.90	2.03	1.74
		d_1/d_n (Å)	-	1.16	1.66	1.93
	β-UO_3 stirring residue	d_n (Å)	3.31	2.90	2.00	1.71
		d_1/d_n (Å)	-	1.14	1.65	1.94

Figure S7

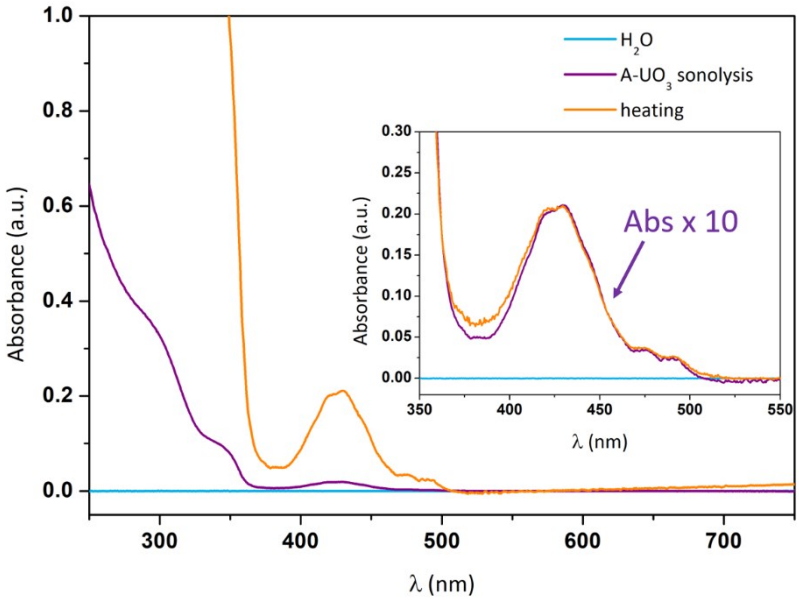

Fig. S7: UV-Vis absorption spectra of supernatant obtained after sonolysis of A-UO_3 in pure water. The spectra are measured before and after concentration by heating. The inset shows the initial spectrum (purple line) multiplied by 10 for comparison.

Figure S8

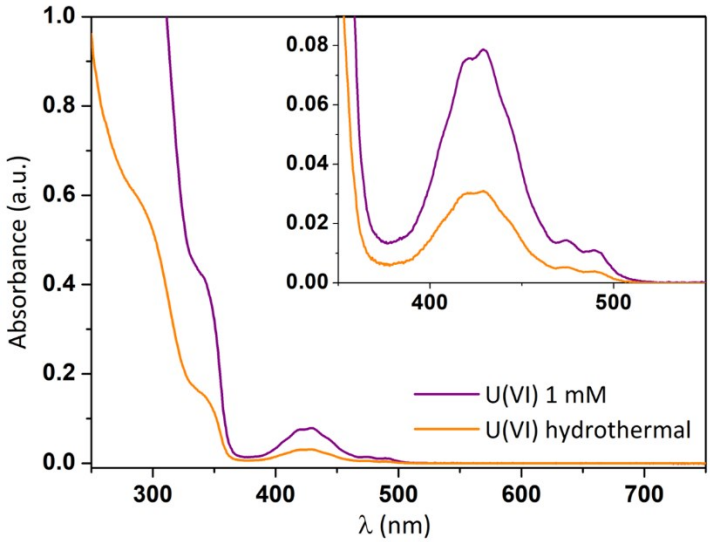

Fig. S8: UV-Vis absorption spectra of a 1 mM aqueous U(VI) solution before (purple line) and after (orange line) hydrothermal treatment (200 °C, 3 h).

Figure S9

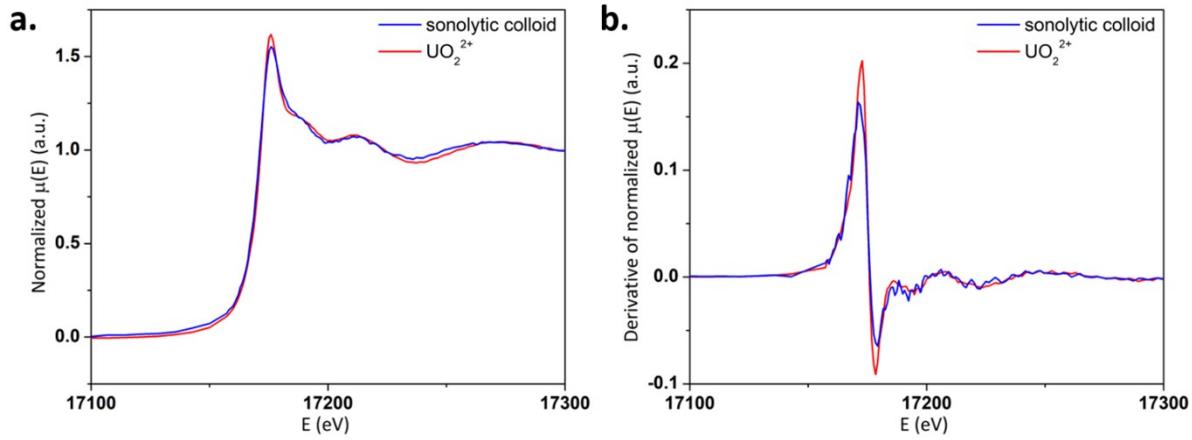

Fig. S9: (a) U L_{III} XANES experimental spectrum acquired on sonolytic colloids of U(VI) obtained after A-UO_3 treatment (blue line). In addition, UO_2^{2+} (aquo form in pure water) XANES spectrum is provided for comparison (red line)[2]. (b) The corresponding derivatives of XANES spectra for sonolytic colloids (blue line) and UO_2^{2+} (red line).

Figure S10

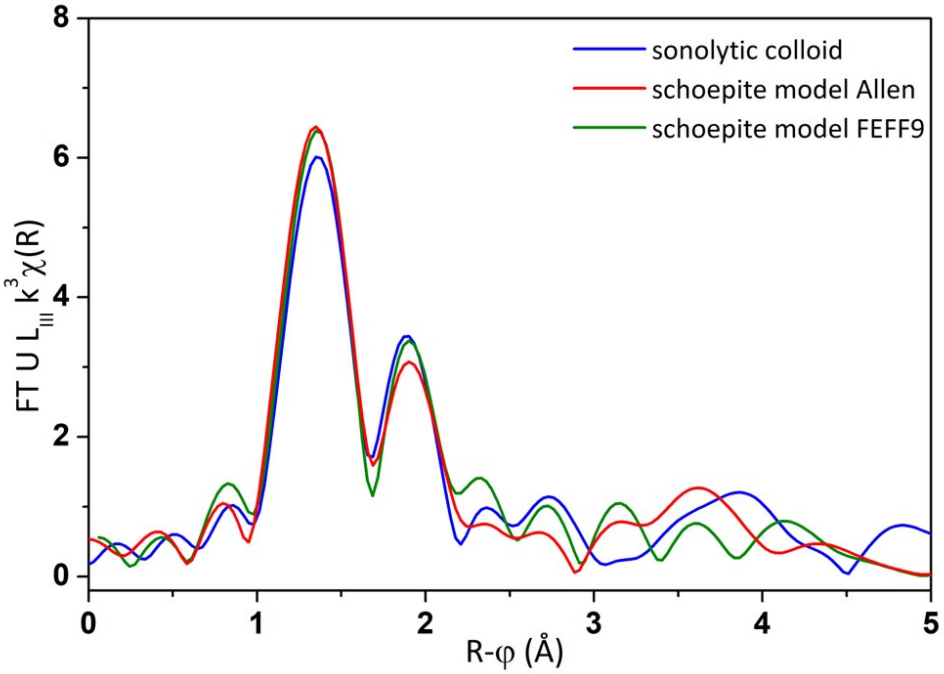

Fig. S10: Fourier transforms (FT) of the k^3-weighted 2.5-11.2 $Å^{-1}$ range for the sonolytic colloids obtained after A-UO_3 treatment (blue line) compared to the schoepite model (green line) obtained with FEFF9 full scattering calculation and the schoepite model (red line) reconstructed from Allen's fit parameters.[3]

Figure S11

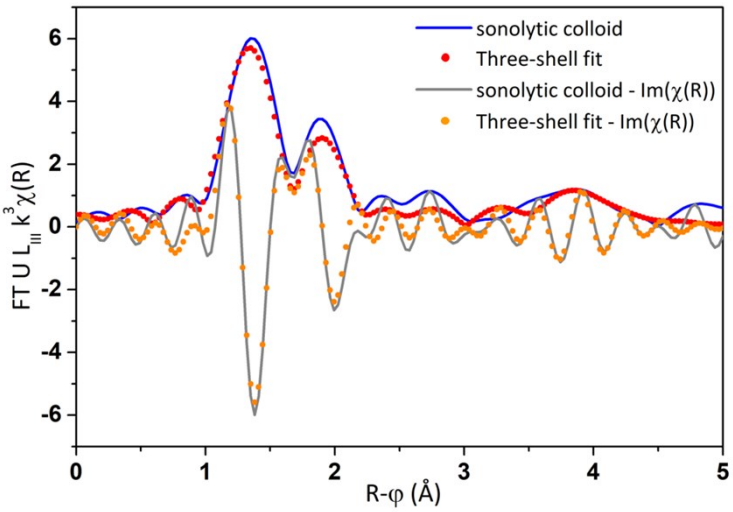

Fig. S11: Fourier transforms (FT) of the k^3-weighted 2.5-11.2 $Å^{-1}$ range for sonolytic colloids obtained after A-UO_3 treatment (blue line) and the corresponding fit using a three-shell model (red points). In addition, the imaginary parts are plot (in grey line and orange points, respectively).

Table S2

Table S2: Model and parameters considered for the F-test on EXAFS fits of U(VI) colloids sample for the four-sell fit with/without the U-U shell. R is equal to the square root of the IFFEFIT parameter R-factor. n is the number of free parameters used in the corresponding fit and m the number of independent data points.

Shells	R	n	k range ($Å^{-1}$)	R range (Å)	m*	F^4
O_{yl}, O_{eq1}, O_{eq2}, U	0.221	10	2.5-11.2	1.1-5	23.6	4.2
O_{yl}, O_{eq1}, O_{eq2}	0.307	7				

*calculated according to the Stern's rule[5]

References

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