Supplementary Material

Effect of Hydrothermal Treatment on Plutonium Retention in Deep Liquid Radioactive Waste Disposal

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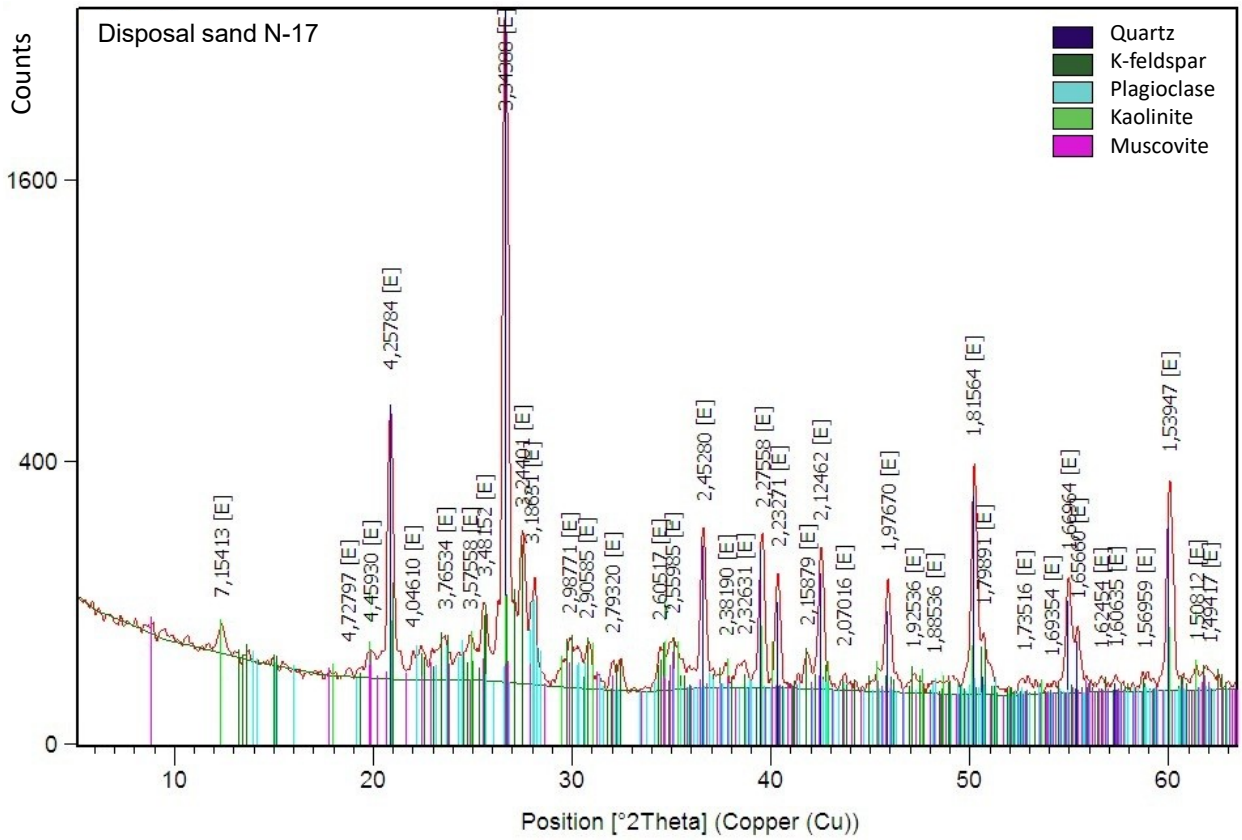


Fig. S1. The PXRD pattern of the disposal sand used for the experiments.

Table S1. The results of PXRD analysis of disposal sand

Mineral name	Chemical formula	Mass fraction, %
Quartz	SiO_2	65
K-feldspar	$KAlSi_3O_8$	14
Plagioclase	$(Na,Ca)[Al(Al,Si)Si_2O_8]$	5
Chlorite	$(Mg,Fe)_3(Si,Al)_4O_{10}(OH)_2 \cdot (Mg,Fe)_3(OH)_6$	1.5
Muscovite	$KAl2(AlSi_3O_{10})(OH)_2$	0.5
Kaolinite	$Al_2(Si_2O_5)(OH)_4$	12
Sum of crystalline phases		98

The sample may contain goethite, siderite, aragonite and pyroxene at the level of the detection limit

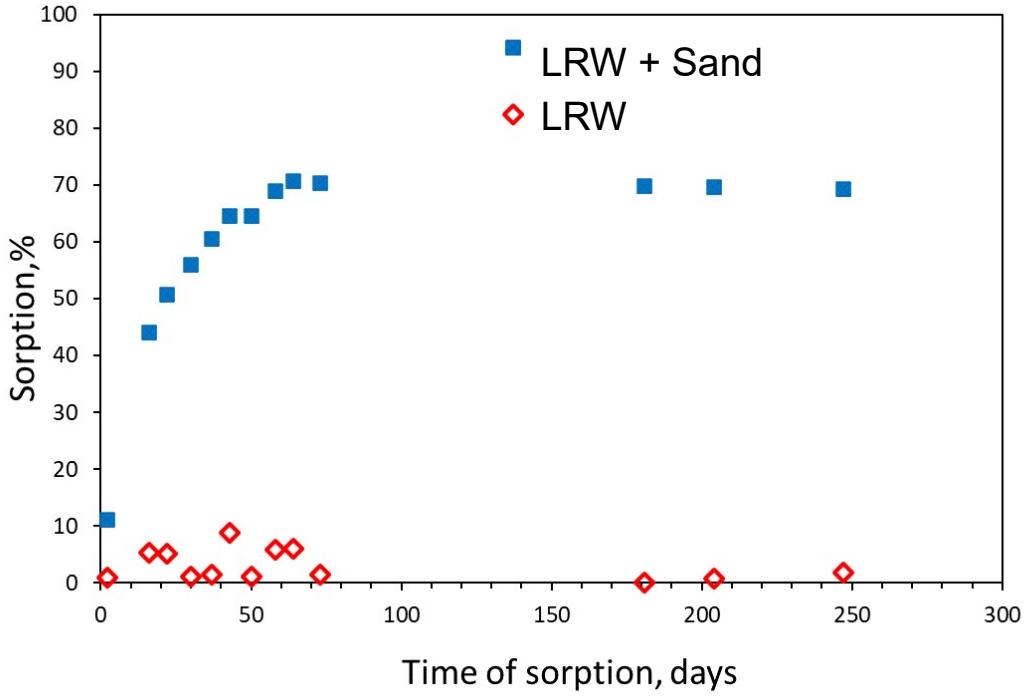


Fig. S2. Room-temperature sorption of Pu from model LRW solutions in the presence and absence of disposal sand (T = 22°C, $[Pu]_{tot}=10^{-7}$ M, [Sand] = 100 g/L, $pH_i = 2.4$).

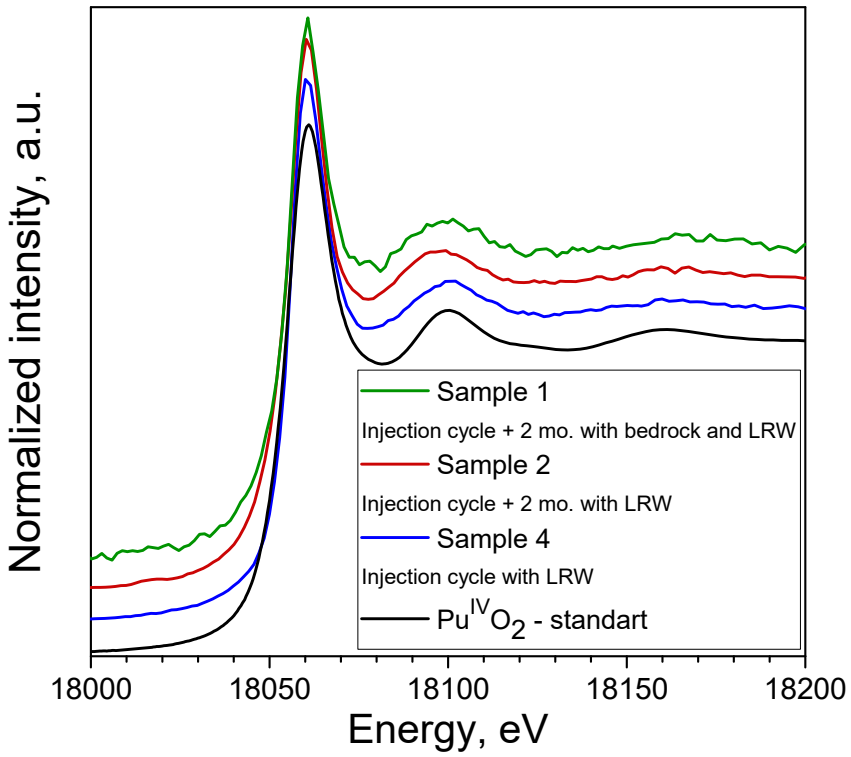


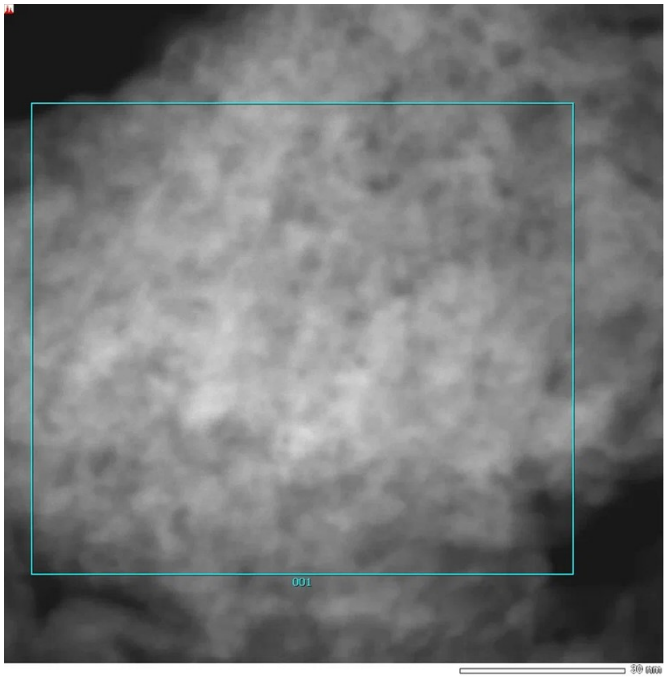
Fig. S3. Pu L_3-edge XANES spectra of studied samples in comparison with Pu(IV) standard.

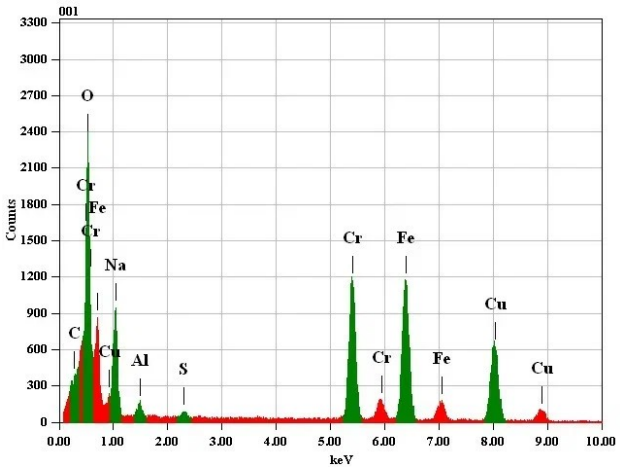
Table S2. Structural parameters obtained from the fitting of EXAFS spectra

Sample	Shell						R-factor
	Pu-O			Pu-Fe			
	CN	R, Å	σ, Å^2	N	R, Å	σ^2, Å^2	
Sample 2	8*	2.25±0.02	0.014±0.001	4.0±1.4	3.43±0.03	0.015*	0.026
Sample 4	8*	2.25±0.03	0.017±0.001	2.21±1.1	3.43±0.04	0.015*	0.019

CN – Coordination number, R – Interatomic distance, σ^2 – Debye− Waller factor, k-range = 3 – 9 Å^{-1}; R-range = 1.2 – 3.4 Å

* - fixed values





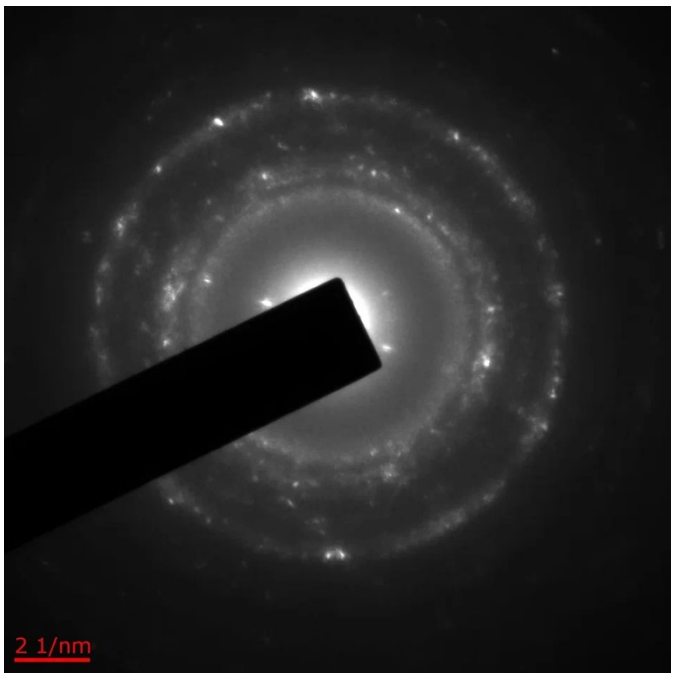


Fig. S4. EDX spectrum and SAED pattern of LRW sediments agglomerate in Sample 2 (One injection cycle, 2 months conditioning with LRW).

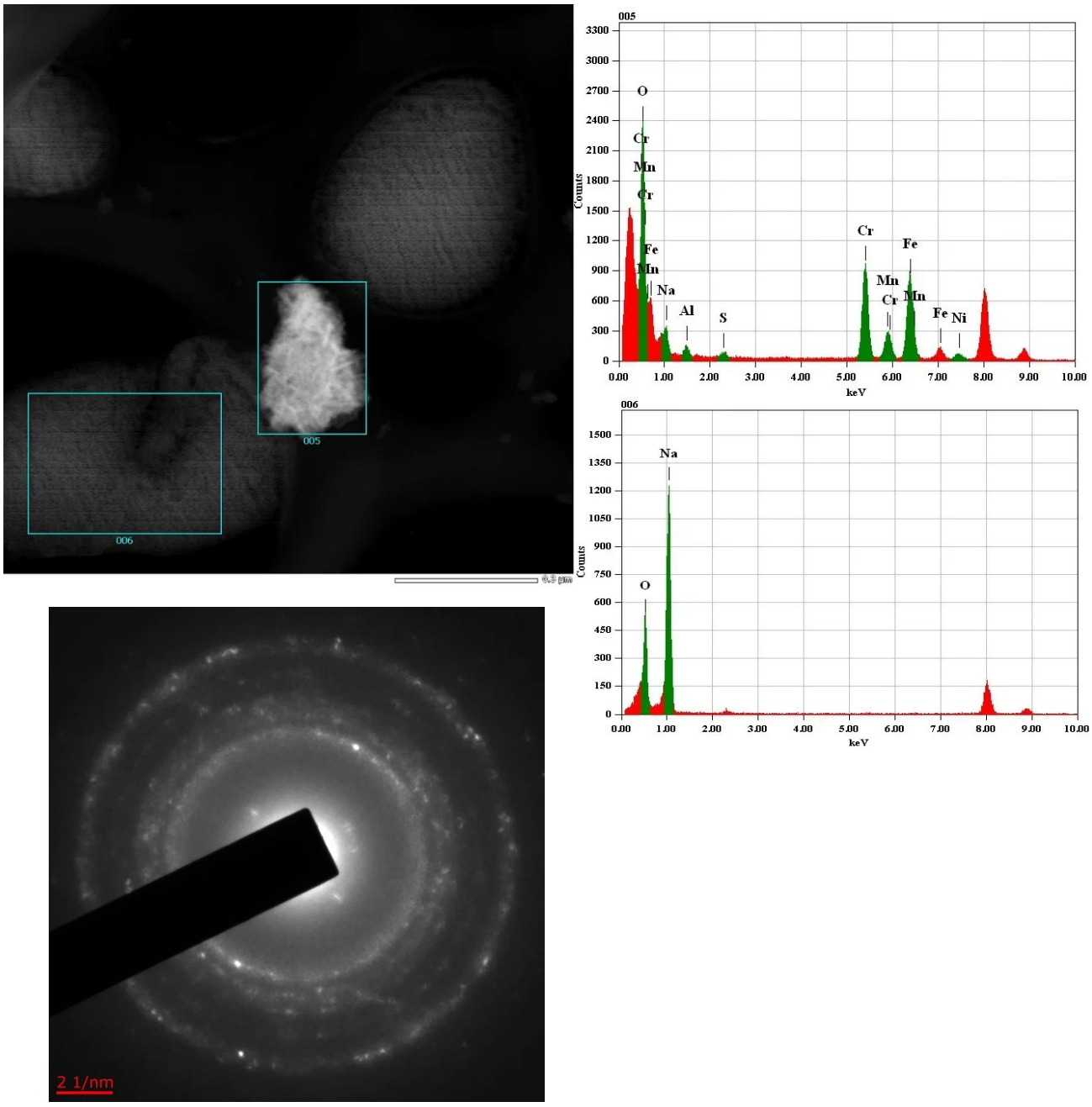


Fig. S5. EDX spectra and SAED pattern of LRW sediments agglomerate in Sample 2 (One injection cycle, 2 months conditioning with LRW).

Table S3. Plutonium oxidation states in acidic leachates assessed by HDEHP extraction

Conditions	Extraction, %	
	Aqueous phase (Pu(V))	Organic phase (Pu(IV), Pu(VI))
One injection cycle, 150°C, LRW	77	23
One injection cycle, 150°C, LRW+Sand	81	19
Three injection cycles, 150°C, LRW+Sand, 3 months conditioning	71	29