

Supplementary information

Table S1. Conditions of Pu precipitation experiments

Experiment	Carbonate source	[CO ₃ ²⁻], M	pH	pH buffer	Volume, mL
Experiment A	Dissolved CO ₂ (g)	~10 ⁻⁴ –10 ⁻³ *	8.4 ± 0.2	TRIS	10
Experiment B	Dissolved CO ₂ (g)	~10 ⁻⁴ –10 ⁻³ *	8.4 ± 0.1	TRIS	10
Experiment C	Dissolved CO ₂ (g)	~10 ⁻⁴ –10 ⁻³ *	8.2 ± 0.1	TRIS	45
Experiment D	Na ₂ CO ₃ /NaHCO ₃	0.04	8.6 ± 0.5	Na ₂ CO ₃ /NaHCO ₃	10
Experiment E	Dissolved CO ₂ (g)	~10 ⁻⁴ –10 ⁻³ *	pH range from 2 to 10 in increments of 2	—	10

* — maximum dissolved carbonates, based on a theoretical calculation by HYDRA/MEDUSA software, Fig. S1.

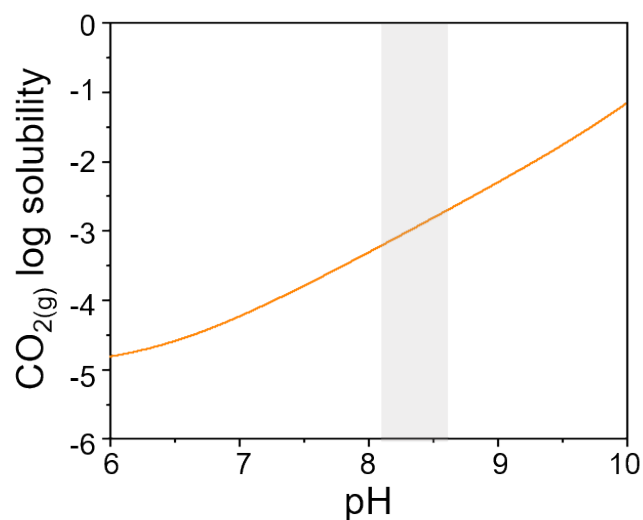


Figure S1. Solubility graph of CO₂(g) calculated by HYDRA/MEDUSA software. The gray area shows the pH conditions corresponding to Experiments A, B, C, and E.

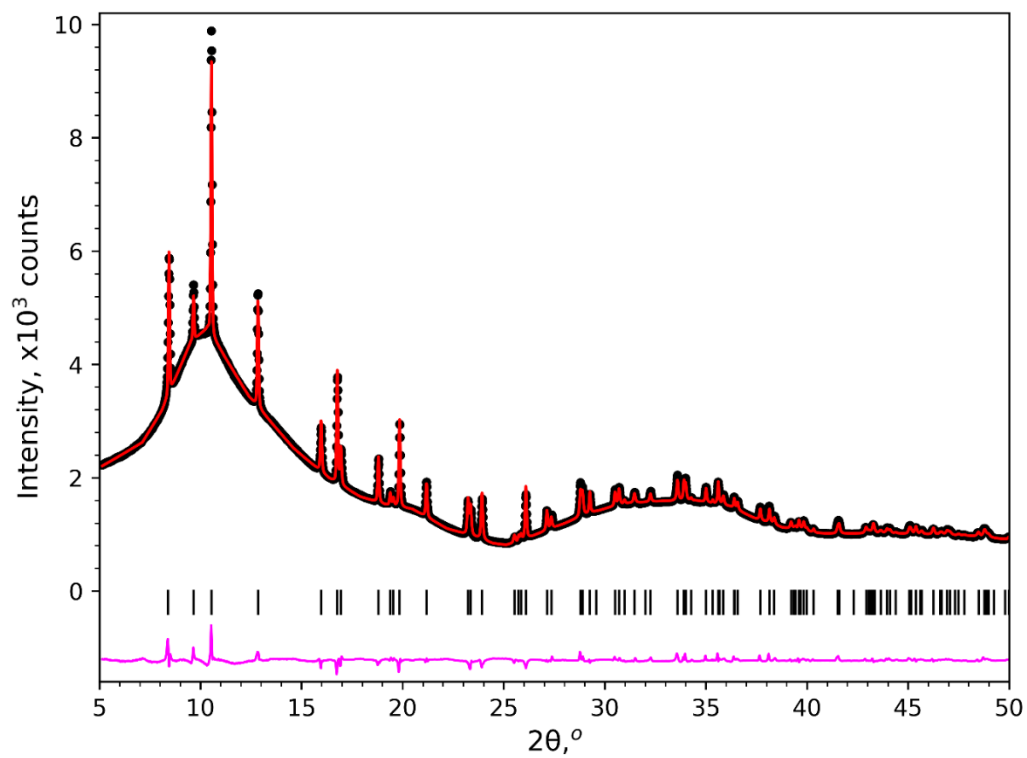


Figure S2. Rietveld refinement plot ($\lambda = 0.74 \text{ \AA}$) of the powder XRD pattern of KPuO_2CO_3 . Experimental data (black markers), refined fit (red solid line), difference profile (offset magenta line), and positions of Bragg peaks (vertical bars).

Table S2. Crystal and refinement data for the structure of KPuO_2CO_3 .

Formula	KPuO_2CO_3
Formula weight	375.1
Diffractometer	Belok/XSA BL
Wavelength (Å)	0.74
Data collection method	Debye–Scherrer
Temperature (K)	293(2)
Crystal system	hexagonal
Space group	$P6_3/mmc$
Temperature (K)	293
a (Å)	5.0746(3)
b (Å)	5.0746(3)
c (Å)	10.0472(11)
α (°)	90
β (°)	90
γ (°)	120
V (Å ³)	224.07(3)
Z	2
D_{calc} (g·cm ⁻³)	5.5598
μ (mm ⁻¹)	20.334
2 θ range, °	5.108–49.999
Number of reflections	110
Parameters, restraints	29, 1
R_p , ωR_p	0.0107, 0.0167
R_{Bragg}	0.0644
$\Delta\rho_{\text{min}}$, $\Delta\rho_{\text{max}}$ (eÅ ⁻³)	-3.35, 5.41

Table S3. Selected bond distances (Å) in the structure of KPuO_2CO_3 . Symmetry codes: (i) $1 - y, 1 - x, 0.5 - z$; (ii) $-1 + x, x - y, 0.5 - z$; (iii) $-x + y, y, 0.5 - z$; (iv) $1 - y, 1 + x - y, z$; (v) $x, 1 + x - y, 0.5 - z$; (vi) $1 - y, x - y, z$.

Parameter	Bond distance (Å)
Pu1–O2 Pu1–O2 ⁱ	1.799(2)
Pu1–O1 Pu1–O1 ⁱⁱ Pu1–O1 ⁱⁱⁱ Pu1–O1 ^{iv} Pu1–O1 ^v Pu1–O1 ^{vi}	2.54(3)
O1–C1	1.36(3)
K1–O2 ⁱ x6	3.0152(8)
K1–O1 ⁱ x6	2.964(17)

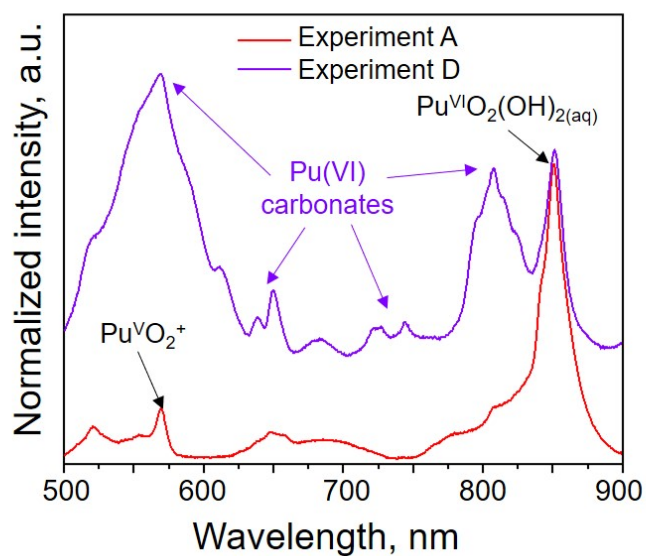


Figure S3. UV-Vis spectra of solutions from Experiments A and D after 5 days.

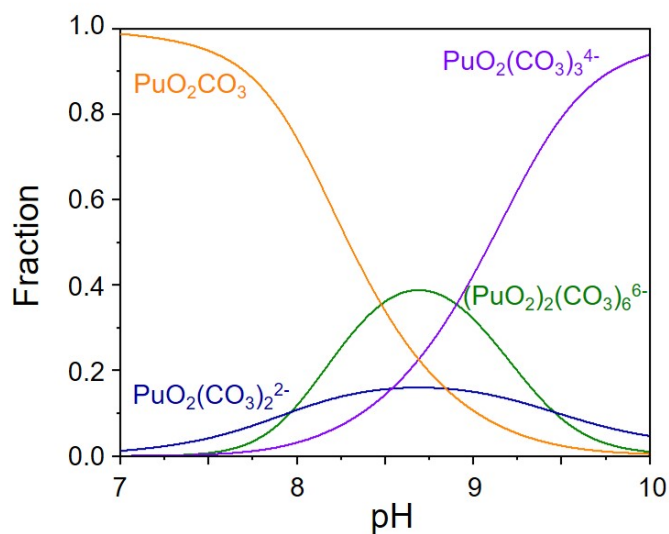


Figure S4. Predominant Pu(VI) carbonate species in solution with $[\text{CO}_3^{2-}] = 0.04 \text{ M}$, $[\text{PuO}_2^{2+}] = 1 \times 10^{-4} \text{ M}$, calculated by HYDRA/MEDUSA software.

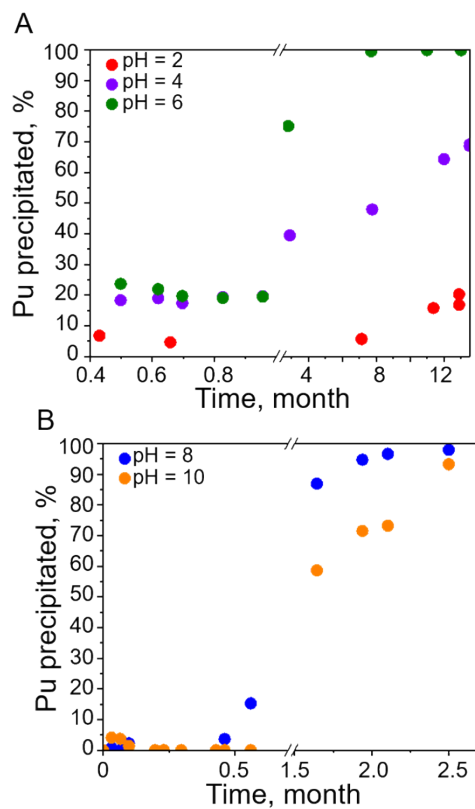


Figure S5. Dependence of Pu precipitated percentage on time in Experiment E with (A) pH = 2, 4, 6, and 12; (B) pH = 8, 10.

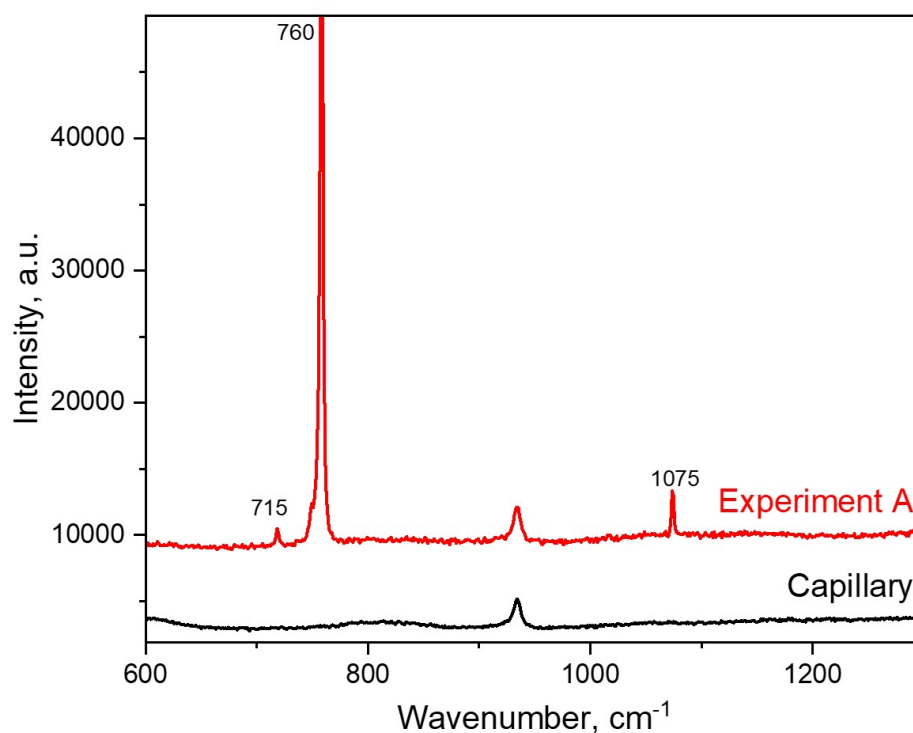


Figure S6. Raman spectra of the solid phase from Experiment A placed into quartz capillary in comparison to empty capillary

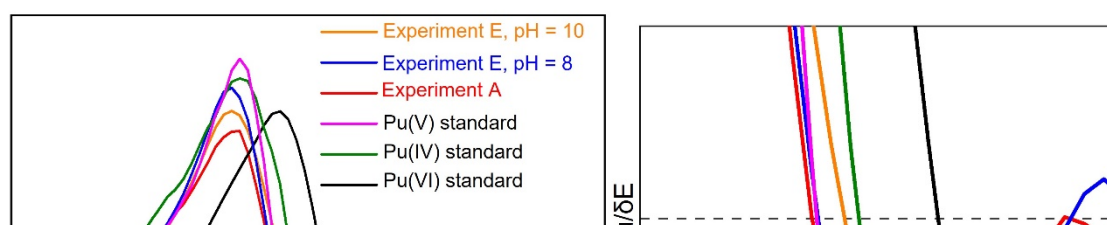


Figure S7. XANES first derivative spectra of the experimental results compared to Pu(VI, V and IV) standards with magnification of zero-crossing (right).

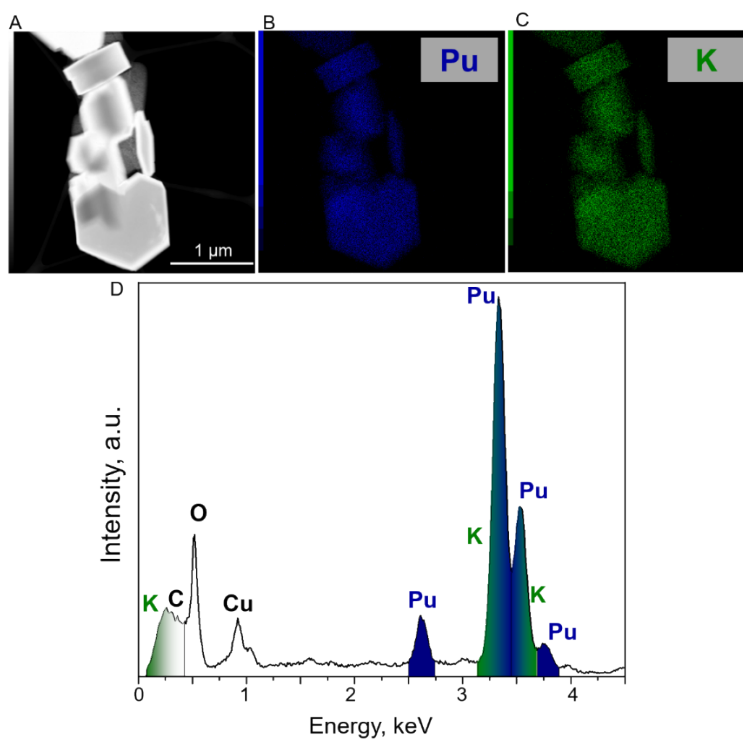


Figure S8. (A) HRTEM image; (B, C) Maps of Pu and K components; (D) EDX spectrum of solid phase precipitated in Experiment A.