Supplementary information

Gladstone-Dale relationship

Equation 1 The Gladstone-Dale relationship between the refractive energy K; refractive index n; density ρ of a mineral and its respective constituents. Where k_1, k_2 and w_1, w_2 represent the constituent refractive energies and weight ratios respectively.

$$K = \frac{(n-1)}{\rho} = k_1 w_1 + k_2 w_2 + \cdots k_n w_n \ (2)$$

Particle settling under centrifugation

Particle settling in centrifugal field is acted upon by two opposing forces, a centrifugal force and a drag force. Under laminar flow conditions (small particle sizes):

Inertial centrifugal force acting on a spherical particle:

$$F_{IC} = m\omega^2 R = \frac{4}{3}\pi r^3 \rho_{particle}\omega^2 R$$

Buoyancy force acting on particle, where α is angular acceleration:

$$F_B = \frac{4}{3}\pi r^3 \rho_{fluid}\alpha = \frac{4}{3}\pi r^3 \rho_{fluid}\omega^2 R$$

Viscous drag force acting on particle:

$$F_D = 6\pi r \mu V$$

Relative centrifugal force:

$$RCF = \frac{\omega^2 R}{g}$$

Force balance acting on a spherical particle at terminal velocity falling through a viscous fluid:

$$F_{IC} = F_B + F_D$$

$$\frac{4}{3}\pi r^3 \rho_{particle}\omega^2 R - \frac{4}{3}\pi r^3 \rho_{fluid}\omega^2 R = 6\pi r \mu V$$

$$\frac{2}{9}\frac{r^2 \Delta\rho \omega^2 R}{\mu} = V_s$$

$$\boxed{\frac{2}{9}\frac{r^2 \Delta\rho \omega^2 R}{\mu} = t_s}$$

Where:

$$\rho_{particle} = particle\ density$$

$$\rho_{fluid} = fluid\ density$$

$$\omega = angular\ velocity$$

$$\alpha = angular\ acceleration = \omega^2 R$$

$$R = radius\ of\ centrifugation$$

$$r = spherical\ particle\ radius$$

$$V_s = terminal\ particle\ velocity$$

$$t_s = terminal\ particle\ settling\ time$$

For a centrifugation time of 3minutes (180s), supernatant travel distance of 2cm; a graph may be constructed (Figure 1) to approximate terminal particle settling time:

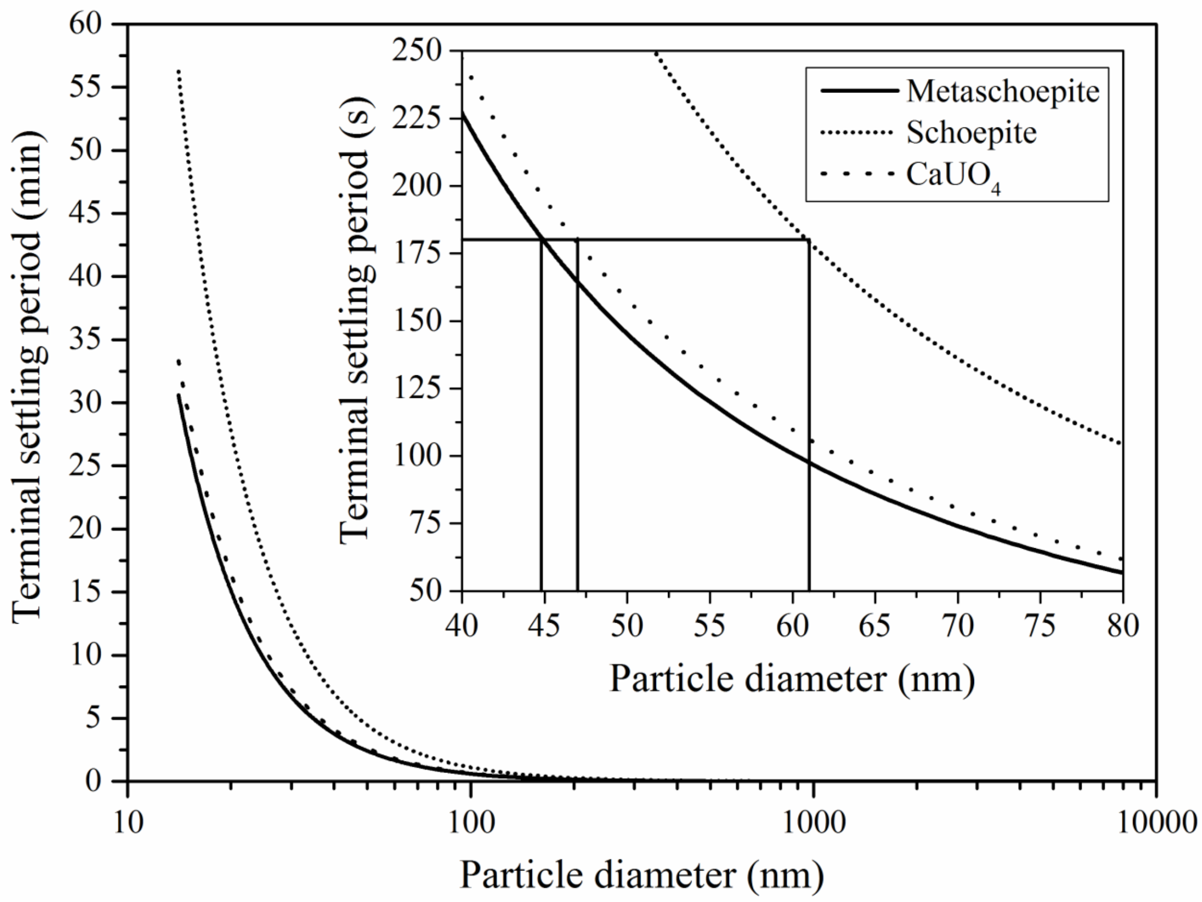
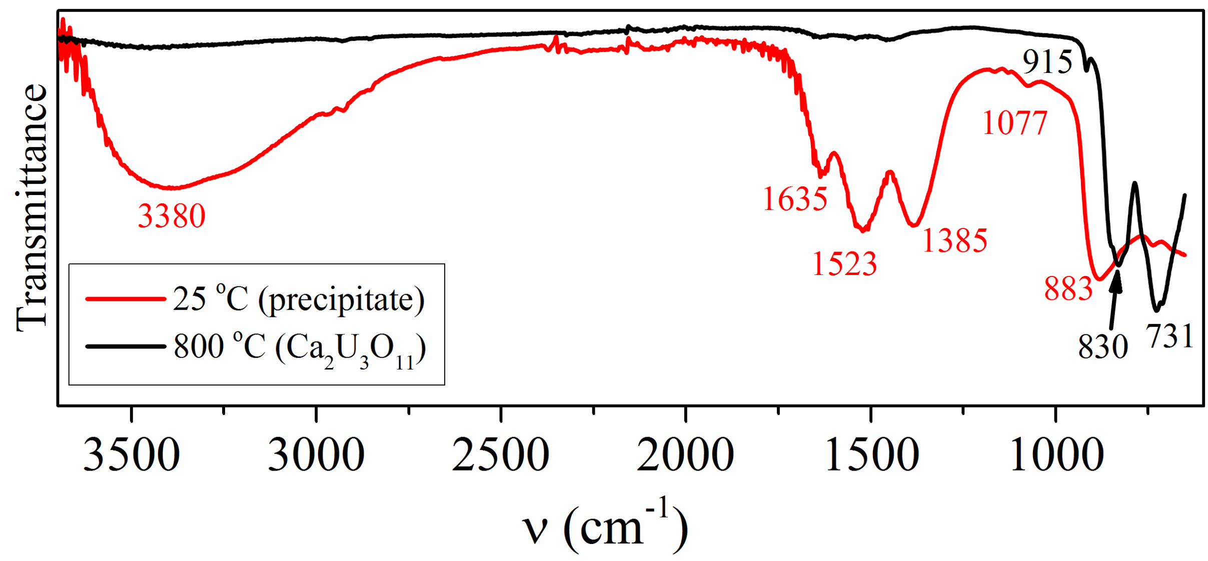


Figure 1 Graph of predicted nanometric spherical particles settling times. (Water at 25°C μ= 0.89 mN s m^{-2} [1]); ρ_p is particle density (Schoepite: ICSD 82477, ρ=4818.64 kg m^{-3}, Metaschoepite: ICSD 23647, ρ=8017.66 kg m^{-3}, $CaUO_4$: ICSD 31631, ρ=7450 kg m^{-3}); ρ_f is fluid density (Water at 25°C, ρ= 997.1kg m^{-3} [1]); R is 0.06m.

Fourier Transform Infra-Red (FTIR) spectroscopy

Powdered samples (~20mg) were analysed using an A2 Microlab Portable mid-IR spectrometer with a Diamond Internal reflection cell (DATR). 10 measurements were completed for each sample and merged.



Literature data				This study		
$Na_2U_2O_7.6H_2O$ [2]	**Becquerelite [3]**	**CaU_2O_7 [4]**	**$Ba_2U_3O_{11}$ [4]**	**25 °C**	**800 °C**	**Assignment**
3379-3578	3504			2500-3700		ν H_2O, OH
1645	1625			1635		δ H2O
				1385-1523		ν_3 NO3
	1250			1077		δ UOH in-plane
	997					
936	927					
882	840			883		ν_3 UO_2^{2+}
	812					
		835	830		830	U=O
		730	750		731	U-O

Figure 2 FTIR spectra of poorly crystalline hydrous Ca-uranate (25 °C) formed at pH 12 and crystalline Ca-uranate ($Ca_2U_3O_{11}$) after dehydration at 800 °C with summarised tentative band assignments based on literature data for analogous compounds.

Summary of derived molar [Ca/U] stoichiometry and formulae from analyses

Method	Molar [Ca/U] ratio	Stoichiometric formula
SEM-EDS	0.63 ± 0.02	$Ca_2U_{3.18}O_{11.5}$
pXRD-Rietveld	0.60 ± 0.03	$Ca_2U_{3.32}O_{12}$
ICP-OES	0.68 ± 0.04	$Ca_2U_{2.92}O_{10.77}$
TG	-	(I) $Ca_2(UO_2)_3O_{3.75}(OH)_{2.5}.3.5H_2O$ (II) $Ca_2(UO_2)_3O_{3.75}(OH)_{2.5}$ (III) $Ca_2U_3O_{11}$ (IV) $CaUO_4$, UO_2
Average	0.64 ± 0.03	$Ca_2U_{3.1}O_{11.4}$

1. Kestin, J., M. Sokolov, and W.A. Wakeham, *Viscosity of liquid water in the range− 8 C to 150 C.* Journal of Physical and Chemical Reference Data, 1978. **7**(3): p. 941-948.

2. Chernorukov, N.G., O.V. Nipruk, and E.L. Kostrova, *Synthesis and study of sodium uranate Na2U2O7·6H2O and of products of its dehydration and thermal decomposition.* Radiochemistry, 2016. **58**(2): p. 124-127.

3. Cejka, J., *Infrared spectroscopy and thermal analysis of the uranyl minerals.* Reviews in Mineralogy and Geochemistry, 1999. **38**(1): p. 521-622.

4. Allen, G.C. and A.J. Griffiths, *Vibrational spectroscopy of alkaline-earth metal uranate compounds.* Journal of the Chemical Society, Dalton Transactions, 1979(2): p. 315-319.