Supplementary information

Gladstone-Dale relationship

Equation 1 The Gladstone-Dale relationship between the refractive energy $K$, refractive index $n$, density $\rho$ 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.

$$K = \frac{(n - 1)}{\rho} = k_1w_1 + k_2w_2 + \cdots k_nw_n \quad (2)$$

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:

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

Buoyancy force acting on particle, where $\alpha$ is angular acceleration:

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

Viscous drag force acting on particle:

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

Relative centrifugal force:

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

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

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

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

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

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

Where:

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

$$\rho_{fluid} = \text{fluid density}$$
\[ \omega = \text{angular velocity} \]
\[ \alpha = \text{angular acceleration} = \omega^2 R \]
\[ R = \text{radius of centrifugation} \]
\[ r = \text{spherical particle radius} \]
\[ V_s = \text{terminal particle velocity} \]
\[ t_s = \text{terminal particle settling time} \]

For a centrifugation time of 3 minutes (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 \( \mu = 0.89 \text{ mN s m}^{-2} \) \cite{1}); \( \rho_{p} \) is particle density (Schoepite: ICSD 82477, \( \rho=4818.64 \text{ kg m}^{-3} \), Metaschoepite: ICSD 23647, \( \rho=8017.66 \text{ kg m}^{-3} \), CaUO\(_4\): ICSD 31631, \( \rho=7450 \text{ kg m}^{-3} \)); \( \rho_f \) is fluid density (Water at 25°C, \( \rho=997.1 \text{ kg m}^{-3} \) \cite{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.
Figure 2 FTIR spectra of poorly crystalline hydrous Ca-uranate (25 °C) formed at pH 12 and crystalline Ca-uranate (Ca₂U₃O₁₁) 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

<table>
<thead>
<tr>
<th>Method</th>
<th>Molar [Ca/U] ratio</th>
<th>Stoichiometric formula</th>
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<tbody>
<tr>
<td>SEM-EDS</td>
<td>0.63 ± 0.02</td>
<td>Ca₂U₂O₇(OH)₃.5</td>
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<tr>
<td>pXRD-Rietveld</td>
<td>0.60 ± 0.03</td>
<td>Ca₂U₃O₁₁</td>
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<td>ICP-OES</td>
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<td>Ca₂U₂O₁₂.6H₂O</td>
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<tr>
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<td>(II) Ca₃(UO₂)₂O₇.₃(H)₂.₅</td>
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<td>(III) Ca₃U₂O₁₁</td>
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<td>(IV) CaUO₄, UO₂</td>
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<tr>
<td></td>
<td>Average</td>
<td>0.64 ± 0.03</td>
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