

## Supplementary Material for:

### Evidence for liquid-liquid phase separation during the early stages of Mg-struvite formation

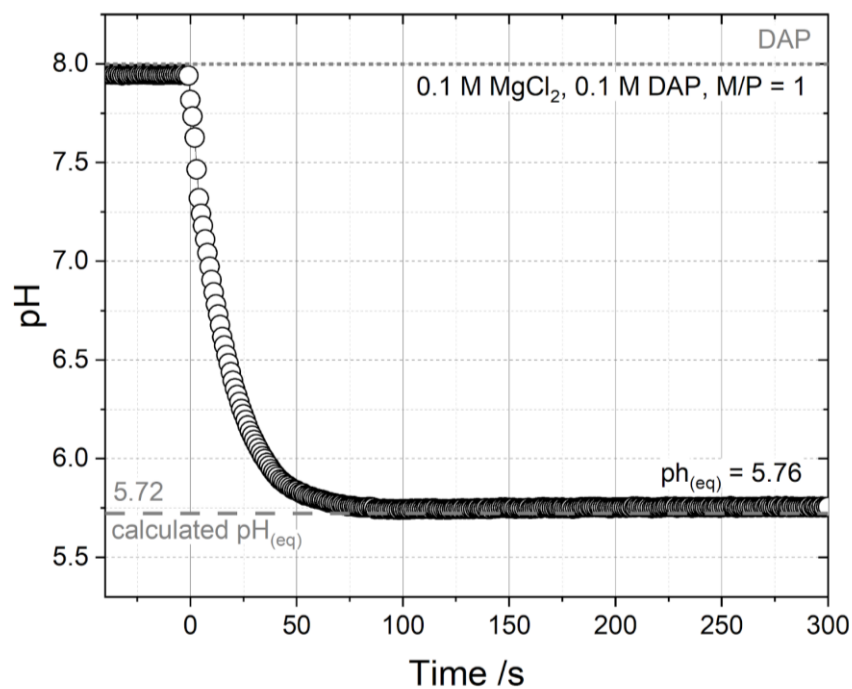
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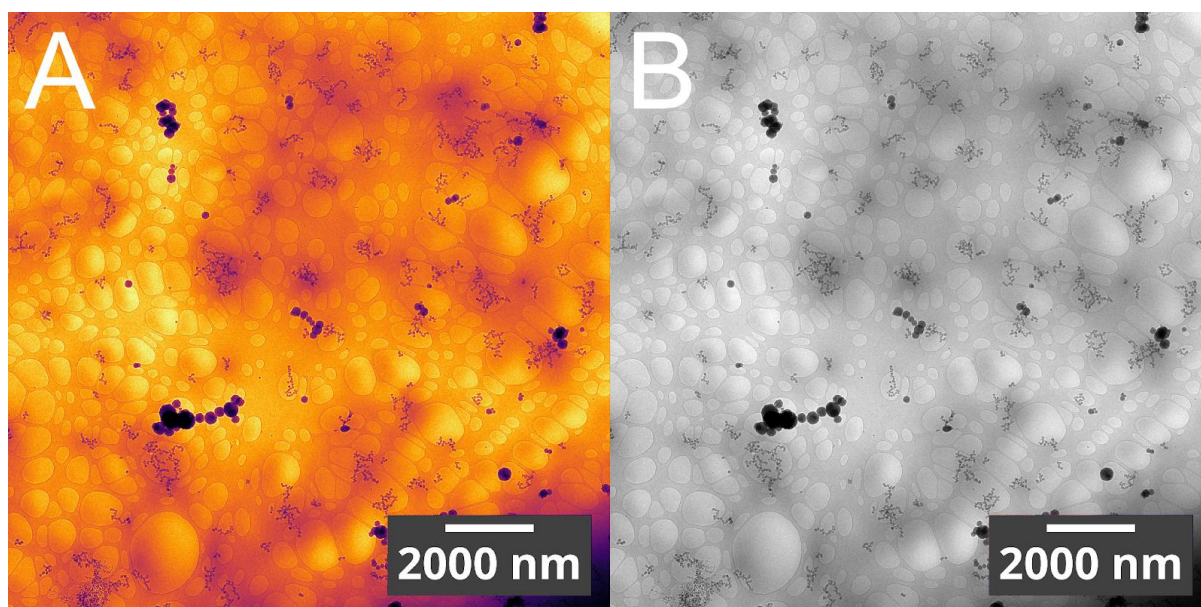
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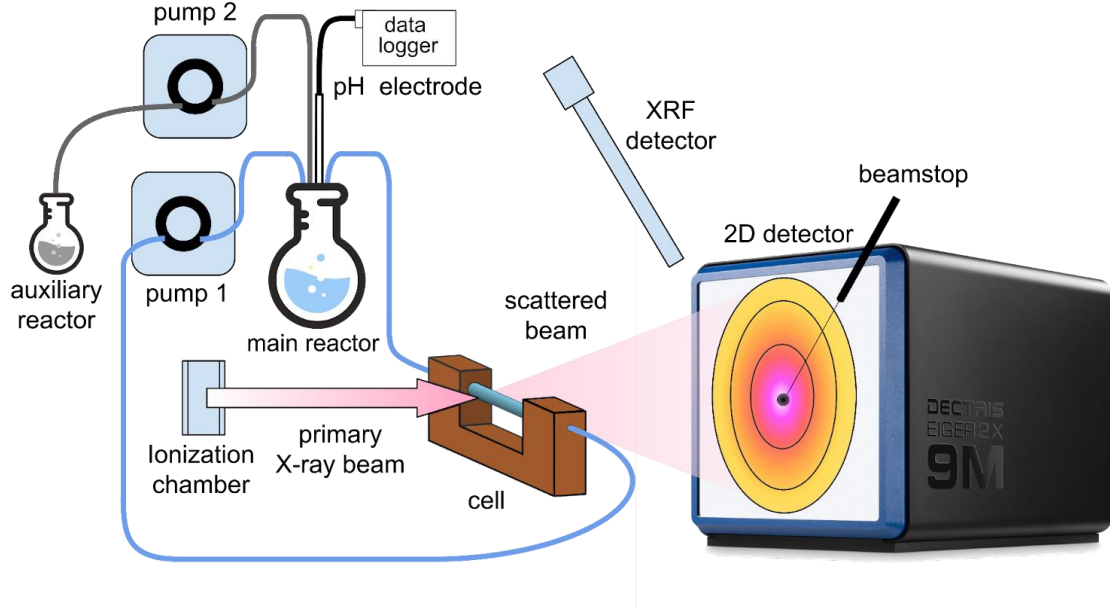
**Keywords:** struvite, phosphates, scattering, nucleation, amorphous phases, non-classical, liquid-liquid phase separation



**Fig. S1.** The time-resolved pH curve from the struvite precipitation reaction (Eq. 1 in the main text) combined with PHREEQC calculated equilibrated pH (grey dotted line) is displayed. Starting with a slightly basic solution of 0.1 M DAP ( $\text{pH} \approx 8.0$ ), the pH drops quickly within a hundred seconds to 5.76 due to mixing with a slightly acidic 0.1 M  $\text{MgCl}_2$  solution and the release of protons during precipitation. Theoretically calculated and experimentally determined pH values were in good agreement with each other.



**Fig. S2.** Cryo-TEM bright-field imaging from the reactant solution sampled after 5 s after mixing: (A) with an applied false-colour scale 'inferno' to emphasize potential electron density contrasts and (B) in grey-scale as a reference to avoid misinterpretations resulting from the colourations.



**Fig. S3.** Sketch of a flow-through-setup, which shows the general experimental configuration in our measurements: a main reactor (main cycle) attached with tubing to a borosilicate glass capillary, pH electrode with data logger and an auxiliary reactor (injection cycle); The beam can penetrate the filled capillary and measures the scattering signal on a 2D detector.

### Supporting Note 1:

We derive how the OZ correlation length,  $\xi_{OZ}$ , from Eq. 2 in the main text, can be compared with an equivalent radius of gyration  $R_g$ , or a radius of an equivalent sphere. The Taylor expansion of the OZ normalised intensity expression is given by Eq. S1, following the same notation as in Eq. 2.

$$I(q, \xi_{OZ}) = \frac{1}{1 + q^2 \xi_{OZ}^2} \simeq 1 - q^2 \xi_{OZ}^2 + q^4 \xi_{OZ}^4 + \dots \quad \text{Eq. S1}$$

Similarly, we calculate the Taylor expansion of the Guinier form factor, which is dependent on  $R_g$  (Eq. S2).

$$I(q, R_g) = \exp\left(-\frac{q^2 R_g^2}{3}\right) \simeq 1 - \frac{q^2 R_g^2}{3} + \frac{q^4 R_g^4}{18} + \dots \quad \text{Eq. S2}$$

By comparing the first terms of the expansions in Eqs. S1 and S2 we obtain Eq. S3.

$$\frac{q^2 R_g^2}{3} = q^2 \xi_{OZ}^2 \Rightarrow R_g = \sqrt{3} \xi_{OZ}$$

For a sphere of radius  $R$ , the relationship to its radius gyration is  $R_g = \frac{3}{5}R$ , and consequently, we obtain  $R \approx 2.89 \xi_{OZ}$ .