Liquid precursors in mineral crystallization : an experimental test of nucleation theories

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Crystallization from ionic solutions often proceeds via a multi-step route, where dense liquid or amorphous nanometer-sized precursors form before the crystalline phase emerges [1]. The nature and behaviour of these precursors play a crucial role in crystallization, ultimately shaping the formation and properties of the resulting nanocrystals [2]. Yet, they are absent from the “classical” single-step theory (CNT), which is still the main framework for modelling and designing nanomaterials, underscoring the need for new theoretical tools aligned with experimental reality.

Among these precursors, reactant-rich droplets remain the most challenging to investigate due to their liquid and transient nature, which hinders access to their properties or formation mechanism, and therefore complicates experimental discrimination between competing crystallization theories [3]. In mineral systems, such nanometer-sized liquid precursors are often suspected but rarely confirmed, as their lifetime is typically below a minute [4] , thereby requiring advanced and in situ experimental approaches.

Here I will show how we elucidated the formation mechanism and properties of reactant-rich droplets involved in the crystallization of cerium oxalate, a well-established surrogate of actinide oxalates in the nuclear industry, through multiscale, quantitative in situ analysis. By combining synchrotron X-ray methods, microfluidics, and electron microscopy [5] , we tracked these liquid precursors down to microsecond timescales and nanometer lengths, and determined their nature, physicochemical properties, and transition to the final crystals to critically assess existing theoretical models. This work provides new insights into mineral crystallization, how it can be accurately modelled, and offers experimental tools to probe such processes.

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