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Pre-nucleation clusters as molecular precursors to nanoscopic liquid-liquid demixing

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Contrary to the notion of classical nucleation theory, the occurrence of stable clusters in aqueous solution prior to nucleation has been demonstrated for various minerals. While it is classically assumed that ion-by-ion growth of un- and metastable species is central to phase separation events, nucleation via stable pre-nucleation clusters is based upon aggregation of larger nanoscopic entities. This process initially yields amorphous nanoparticles, and represents a crystallization pathway, which has been studied in the most abundant biominerals, calcium carbonate and calcium phosphate, in particular. Utilizing computer simulations, it has been proposed that pre-nucleation clusters are highly dynamic and liquid-like chains of alternating kations and anions, which lay the foundation to an initial nanoscopic liquid-liquid separation. While various experimental observations may be explained by this mechanism, the locus of the proposed liquid-liquid miscibility gap remains unknown. We present an overview of the pre-nucleation cluster pathway, and demonstrate that a combination of experimental techniques may be utilized to localize the binodal and spinodal curves characterizing a nanoscopic liquid-liquid miscibility gap of aqueous calcium carbonate.