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Barite nucleation from atomic-scale simulation: Contrasting classical and non-classical reaction pathways

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Understanding mineral nucleation and growth is critical to develop a predictive capability for multiple subsurface processes, including scale formation, enhanced geothermal energy, CO2 injection, etc. However, our ability to predict nucleation rates is limited, including for how rates are affected by additives, substrate properties/roughness, aqueous solution compositions, etc. The reaction pathways by which nucleation occurs has been of interest for decades, with accumulating evidence for non-classical or classical mechanisms of crystallization. Despite the intensity of research, information about which mechanisms are active under a given condition (and why) is still elusive with contradictory evidence found in the literature.

Here, I will present the results of atomic-scale, rare event simulations at constant chemical potential to understand the nucleation process of barite (BaSO4) at two different supersaturations. We probe nucleation occurring by non-classical and classical pathways, including the dynmically ordered liquid like polymer (DOLLOP) structure versus more crystalline forms. We compare the structures of the incipient nuclei and energy barriers to their formation by biasing the simulation in two different ways to promote more DOLLOP versus higher coordination numbers more indicative of crystalline barite. We also compare the structures of nuclei produced in the simulations to experimental pair distribution function measurements of barite nucleation in confined amorphous silica nanopores. From this, some general and insightful conclusions are drawn about non-classical versus classical nucleation pathways and their likelihood.