

Formation pathway of calcium sulfate minerals in natural and engineered environments

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Calcium sulfate phases are abundant evaporitic minerals occurring both on Earth and Mars, including spectacular hydrothermal formations such as the giant gypsum crystals [1]. Despite its significance in both natural and engineered environments, the CaSO₄-H₂O system has received surprisingly little attention in the recent tsunami of studies addressing alternative mechanisms of solution-mediated mineral formation. Nevertheless, recent findings suggest a rather complex mineralization process for CaSO₄ phases, involving different precursor species [2,3].

The aim of this talk is to review the current state of our understanding of calcium sulfate precipitation from solution. Following a brief introduction on the mineralogy and relevance of calcium sulfates, the CaSO₄-H₂O phase diagram will be discussed, including a detailed account of the respective transition temperatures and the influence of salinity on relative stability fields. Subsequently, the formation mechanisms of CaSO₄ phases will be addressed, both from a classical perspective and in view of recently gained insights, considering the influence of different solution conditions. In particular, the effects of temperature, ionic strength, solvent polarity and additives on precipitation dynamics and phase stability will be highlighted [e.g. 4,5]. Based on all this evidence, a tentative unified model for calcium sulfate crystallization across its phase diagram is proposed [6], identifying water activity and corresponding changes in the hydration of CaSO₄ precursors as key aspects during phase selection.

Finally, the central questions that still need to be resolved before a holistic view of the nucleation, growth and transformation mechanisms of solid phases in the CaSO₄-H₂O system is attained will be briefly discussed.

[1] Van Driessche et al (2010) *PNAS*, 108, 15721.

[2] Van Driessche et al (2012) *Science*, 336, 69-72.

[3] Stwaski et al (2016) *Nat. Commun.* 7, 11177.

[4] Ossorio et al (2014) *Chem. Geol.* 386, 16.

[5] Tritschler et al (2015) *Angew. Chem.* 54, 1.

[6] Van Driessche et al. (2017) *New Perspective on Mineral Nucleation and Growth*, Springer-verlag.