

An experimental and modeling scheme for nucleation under dynamic saturation: A case study of CaCO₃ precipitation with implications for energy production from and CO₂ sequestration in subsurface reservoirs

AMIT G REISS, YUQING YE, AMY T KAN AND MASON B TOMSON

Rice University

Presenting Author: ar166@rice.edu

Mineralization is key for water-rock interactions and the development of fluid chemistry in natural and engineered environments. Energy extraction from hydrocarbon and hydrothermal reservoirs, as well as CO₂ sequestration, causes perturbation to the equilibrium of subsurface brines that become increasingly supersaturated towards various minerals, resulting in nucleation and scale-formation that clogs pipes, damages machinery, and alters the porosity and permeability of reservoir rocks.

Currently, experimental studies of nucleation kinetics utilize setups where the saturation is constant before precipitation. Moreover, numerical models assume constant saturation before nucleation, whether the formulation is based on the classical theory [1] or empirical schemes [2]. However, the processes causing the increased supersaturation, such as degassing, evaporation, temperature change, etc., during energy production, and the increasing ion concentration following the dissolution of reservoir rocks during CO₂ sequestration are continuous. Subsequently, traditional experiments and models fail to address the dynamic nature of supersaturation driving mineralization.

We developed a novel experimental system for quantifying nucleation kinetics during increasing supersaturation over a wide range of environmental conditions and a modeling scheme describing the process. We conducted experiments at pH of ~6.8, 50-90 °C, and I=1m to determine the effect of environmental conditions and the presence of organic polymers on the nucleation kinetics of CaCO₃ mineral phases, the forming polymorph, and crystal habit under dynamic supersaturation. Furthermore, we tested the impact the rate of saturation change has on the kinetics and formed solids.

Here, we will present our experimental system and results showing, amongst other things, that: 1. For a given set of conditions, induction occurs at a similar SI but at a time that is inversely dependent on the rate of increase in supersaturation; 2. Temperature dependence of organic polymer impact on CaCO₃ polymorph and; 3. a numerical model accounting for the dynamics of supersaturation that provides excellent agreement between measured and calculated induction times with differences that scale with the analytical uncertainty and an R²=0.97 over the entire range of environmental conditions we tested.