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A lab on a chip concept for rationalizing hydro-geochemical processes at the pore scale

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Hydro-geochemical processes such as transport-induced mineralization are important processes governing the evolution of many systems in the context of energy-related exploitation of the sub-surface. These processes can lead to an alteration of permeability, diffusivity and other physical characteristics of the rock matrix that can have significant effects on subsurface solute and gas transport. The understanding of these phenomena at the pore scale is a prerequisite for the development of predictive conceptual approaches to describe the evolution of the subsurface. Our lab on a chip concept which combines microfluidic experiments and reactive transport modelling [1, 2] has proven to be a powerful tool to (i) evaluate the impact of hydrological heterogeneity on nucleation mechanisms [3], (ii) decode oscillatory zoning exhibited by solid solutions crystallizing in porous media [4], (iii) unravel the dynamic nature of porosity clogging at perturbed interfaces [5], and (iv) parameterize porosity-diffusivity relationships [6] with respect to coupled mineral dissolution-precipitation reactions. At present, we resolve current controversies on crystallization in confinement addressing specifically whether nucleation is pore size dependent.

Our investigation focuses on one of the most problematic minerals, barite, encountered in energy-related sub-surface exploitation [7]. Microfluidics experiments conducted in nano-confined volumes of solution, i.e., in droplets ranging between 0.3 and 3 nL, showed that nucleation is a probabilistic event that scales with the volume of fluid. While our statistical analysis shows that inhibition of barite nucleation will start at pore sizes $< 1 \mu\text{m}$, theoretical calculations show that the pore size-controlled solubility [8] effect (a thermodynamic effect) becomes effective only in pores of sizes less than $< 0.1 \mu\text{m}$. In a second step, the influence of diffusive transport was also investigated by fostering the crystallization of barite in a pore network consisting of large and small micrometer-sized pores interconnected by fine squared capillaries of $1 \mu\text{m}^2$ cross-sectional area. Although preferential mineralization was observed in larger pores, at low supersaturation crystallization was observed only in the fine capillaries. This unexpected behaviour can be explained by the high reactive surface area (and defects) per unit volume resulting from an increase in nucleation sites in the capillaries. The general conclusions of our study are that in porous media the nucleation kinetics of barite is more and more affected by the surface energy of the substrate as the pore size decreases from micrometric to nanometric scale. Consequently, it can be expected that mineralization occurs preferentially in larger pores in rock matrices but other parameters such as (i) exchange of the fluids w.r.t reaction time, and (ii) the shape, roughness, and surface functional properties of the pore should also be considered as they might reverse this trend.

Participation

In-Person

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