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Preferential crystallization in larger pores: Myth or reality

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Crystallization of minerals in porous media is an important process governing the evolution of many subsurface systems. This process can have significant effects on subsurface contaminant transport, e.g., regarding the immobilisation of ^{226}Ra released from radioactive wastes through co-precipitation with sulphates in clay rocks[1]. Nucleation is often claimed to be inhibited as the pore size decreases[2], based on observations of mineral precipitation kinetics. The formation of supercritical nuclei and the following mineral growth rapidly depletes solute species from the pore solution. This significantly decreases the saturation ratio and thus the driving force for the formation of critical nuclei. Consequently, in porous media such as clays, where solute transport is slow and pore sizes vary over several orders of magnitude, co-precipitation processes immobilizing radionuclides is expected to occur in larger pores. To investigate pore-size dependency on nucleation, we designed a microfluidic experiment combined with time lapse optical microscopy addressing the crystallization of barium sulphate (the host phase for radium) in micro-confinement. Our micromodel consists of two supply channels interconnected by an inert pore network consisting of circular pores of various diameters and heights, interlinked by square capillaries with a cross-sectional area of 1 μm^2. The precipitation of barite was induced by counter-diffusion of SO_4^{2-} and Ba^{2+} into the pore network, with experiments conducted at saturation ratios between 1.1 to 4. At high saturation ratios, no differences between small and large pores were observed. In contrast, at lower saturation a preferential mineralization in the larger pores with shorter induction time was noted. Surprisingly, in these experiments the dynamics of nucleation were also accelerated in the capillaries. A possible explanation, which is still being investigated, is that the size of the critical nuclei is in the same order of magnitude as the dimension of the capillaries, which could favour nucleation[4]. Currently we investigate the nature (crystalline versus amorphous) and shape of the crystals, which are guided by the patterns of the pore network.

1 Curti et al. 2019 Eur. J. Mineral.
2 Prieto 2014 Mineral. Mag.
3 Poonoosamy et al. 2019 Chem. Geol.
4 Hedges and Whitelam 2013 Soft Matter