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The effect of porosity clogging on the diffusivity of porous media: Novel microfluidic experiments and pore-scale modelling approaches

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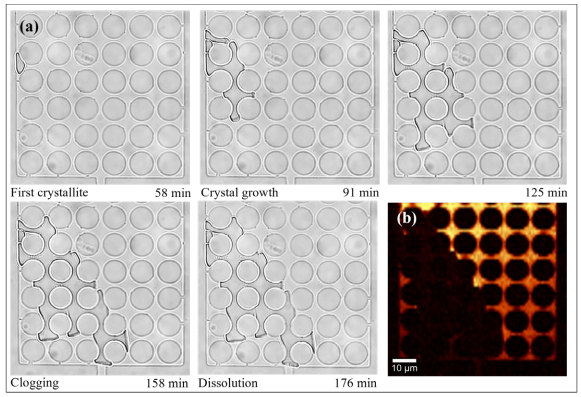

Figure 1 (a) Bright field images of celestine growing in the microfluidic chip. **(b)** Raman image of D_2O distribution during porosity clogging.

Understanding geochemical processes and their impact on macroscopic transport properties of porous media is essential for describing the long-term evolution of various subsurface systems[1]. Chemical and thermal gradients promote mineral precipitation reactions in porous media, resulting in a reduction of porosity and potentially clogging transport pathways of solutes. Commonly applied porosity-diffusivity relationships in continuum-scale reactive transport modelling based on Archie's law and extended versions thereof describe the case of clogging as a final state, setting the effective diffusivity to a negligible low value[2]. However, recent experiments and pore-scale modelling investigations demonstrated the limitations of empirical laws in predicting effective transport properties in response to a precipitation induced porosity reduction and pore clogging, suggesting a non-negligible inherent diffusivity of newly-formed precipitates[3-4]. To verify this hypothesis, we developed a microfluidic reactor design that combines time-lapse optical microscopy and confocal Raman spectroscopy, providing real-time insights into mineral precipitation induced porosity clogging under purely diffusive transport conditions, using the precipitation of celestine ($SrSO_4$) as a model system (Figure 1a). As the pore network became clogged, isotopic tracer diffusion experiments were conducted and monitored by Raman spectroscopy to visualize the transport of deuterium through the evolving microporosity of the precipitates, demonstrating the non-final state of clogging (Figure 1b). The evolution of the porosity-diffusivity relation in response to precipitation reactions shows an increasingly deviating behavior to Archie's law. The application of an extended power law improved the description of the evolving porosity-diffusivity relation, but still neglected post-clogging features. Currently, we develop microfluidic setups to answer the question how clogging-related processes depend on initial pore geometries. The combination of microfluidic experiments and pore-scale modelling opens new possibilities to identify and validate relevant pore-scale processes, providing data for upscaling approaches and to derive key relationships for continuum-scale reactive transport simulations.

1. Masoudi et al. 2021 Int. J. Greenh. Gas Control.
2. Xie et al. 2022 Appl. Geochem.
3. Deng et al. 2021 Water Resour.
4. Deng et al. 2022 Appl. Geochem.