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A lab on a chip approach for deciphering porosity clogging at barrier interfaces in deep geological repositories for radioactive wastes

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Geological repositories for nuclear waste are based on multi-barrier concepts, combining engineered materials with a suitable host rock. Chemical and thermal gradients at barrier interfaces can lead to mineral precipitation, resulting in a reduction of porosity and potentially leading clogging, which significantly affects the diffusivity of the porous media [1]. A process understanding of porosity clogging and its effect on macroscopic transport properties is thus essential for a reliable assessment of the evolution of the repository. Although in the context of nuclear waste disposal, a porosity reduction appears desirable to inhibit radionuclide migration, it can be detrimental, particularly in the case of corrosion-induced gas generation [2]. So far, mechanisms and kinetics of clogging are poorly understood, challenging the predictive capabilities of reactive transport models.

Recent experiments conducted to benchmark continuum-scale models revealed the inadequacy of conventional porosity-diffusivity relationships (Archie's law) to account for changes in effective diffusivity of evolving porous media [1]. Further pore-scale modelling investigations suggested an inherent diffusivity of newly formed precipitates [3]. To verify this hypothesis, we developed a “lab-on-a-chip” approach [4] to decipher clogging phenomena, combining time-lapse optical microscopy and confocal Raman spectroscopy. The microfluidic device consisted of a 2D pore network linked to two supply channels which enabled the diffusive mixing of Sr^{2+}_{aq} and $SO_4{}^{2-}_{aq}$ ions, triggering the precipitation of celestine. As the pore network became clogged, the transport of deuterium through the evolving microporosity of the celestine crystals was visualized by *in-operando* Raman imaging, demonstrating the dynamic nature of porosity clogging. The clogging inhibited further mixing of reactants, leading to dissolution and recrystallization of celestine in a cyclic manner. Numerical tracer experiments using pore scale modelling were conducted on the 2D images of the evolving pore network to determine the effective diffusivity and to derive advanced porosity-diffusivity relationships. The microfluidic experiments in combination with pore-scale modelling opens new possibilities to decipher pore-scale processes and provides data for upscaling parameters and deriving key relationships for continuum-scale reactive transport simulations.

[1] Chagneau et al. (2015), *Geochem.Trans.*16,13.

[2] Xu et al. (2008), *App.Geochem*.23,3423-3433.

[3] Deng et al. (2022), *WaterResour.Res.*137,105207.

[4] Poonoosamy et al. (2019), *Chem.Geol.*528,119264.