

Influence of solution pH on the formation and migration of fines in deep subsurface energy systems

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Deep subsurface energy systems rely on optimized fluid transport during controlled stimulation of the host rock in order to exchange energy, CO₂, and/or hydrocarbons. However, these stimulation processes often promote fluid-rock interactions that may lead to the formation of small colloidal particles, or 'fines', that are suspected to migrate through the rock matrix, partially or fully clog pores and microfractures, and promote the mobilization of potential contaminants. Thus, the goal of this work is to understand the geochemical changes of the host rock in response to reservoir stimulation that promote the formation and migration of fines.

Two different carbonate-rich shales with varying pyrite content were exposed by through-diffusion to different solution pHs (acidic and neutral). Fines that are able to migrate from the shale into the bulk fluid were characterized by ICP-MS, synchrotron-based EXAFS, TEM, and single-particle ICP-MS. Iron and other mineral transformations at the shale-fluid interface were characterized by synchrotron-based XRF mapping.

When exposed to low pH solution, extensive mineral dissolution and secondary precipitation was observed. Additionally, iron (oxy)hydroxide particles collocated with silicates were observed by SEM. At low pH, single-particle ICP-MS shows the mobilization of chromium and other metals with these iron particles. Iron EXAFS spectra of the solution at the shale-fluid interface suggests the rapid formation of ferrihydrite nanoparticles.

Thus, we demonstrate that increasing the fluid pH promotes the mobilization of primary silicate minerals and the rapid formation of new iron precipitates. These iron fines have the potential to migrate through the shale and mobilize other heavy metals.