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Atomic force microscopy imaging of coupled dissolution-precipitation on a calcite surface when exposed to a chromium-rich fluid

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Dissolution and precipitation of minerals in the presence of aqueous fluids is controlled by a boundary fluid layer in which the atoms attach/detach from the mineral surface and diffuse towards the bulk fluid. However, when dissolution at the mineral surface is faster than diffusion of dissolved species away from the surface, a concentration gradient develops at the reaction interface. In the last years, several studies of nanoscale imaging have been performed to look at precipitation and dissolution of nanoparticles on different carbonate surfaces and how these nanoparticles could trap various pollutants such as Se, As, and Sb. In this study, we have performed a series of time-lapse atomic force microscopy (AFM) experiments and a series of experiments in a stirred flow-through reactor. The aim of these experiments was to study how solid nanoparticles form and attach to the surface of calcite in the presence of chromate ions, a toxic element, as well as quantify the amount of trapped Cr on calcite. In both the experiments, we injected distilled water before injecting a solution with a constant Cr(VI) concentration. In the flow-through reactor the solution had a concentration of either 10 or 100 ppm Cr while in the AFM, it was 1, 10, 50 or 100 ppm Cr. All experiments were performed at room temperature and under a constant initial pH in the range 6-12. During the AFM experiments, we observed calcite dissolution along etch pits. In the presence of chromate, a new mineral nucleated and grew at the calcite surface by a process of coupled dissolution-precipitation. The amount of precipitates varied with experimental conditions. In experiments with high concentrations of Cr or at high pH values, we observed a lot of precipitates, while we rarely observed any precipitates at lower concentrations of Cr. At pH 10.5 the precipitates covered the calcite surface that became passivated. The dissolution rates measured from the growth of etch pits showed that they were slightly dependent on the Cr concentration. We imaged the calcite surface with scanning electron microscopy and observed some Cr in the precipitates, indicating that chromium could be captured from solution and trapped in solid nanoparticles in the presence of calcite. The observation of Cr in calcite may have implications for the leaching of concrete or the fate of waters leaking from landfills, where, in both cases, the release of chromate ions into the environment could have harmful consequences.