

## Flow rate and ionic strength effects on calcite reactivity in microchannels

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Reactions at the mineral – water interface play a central role in environmental processes. Calcite, in particular, occurs widely, regulates the pH of natural waters, is a major global CO<sub>2</sub> sink, and is effective at metal sequestration. Most studies on the reactivity of calcite have been conducted using calcite powders or freshly cleaved surfaces in contact with bulk solution. In natural environments, however, chemical reactions may occur in interconnected pore space, where the mineral-water interactions are controlled by advective-diffusive transport under fluid saturated conditions. Effect of fluid flow on reactive transport is a well known phenomenon. However, reactivity of minerals under advection dominated conditions is not well studied. Here we present the effects of different fluid flow conditions and pathways on chemical and structural changes at the calcite (104) surface as a function of reaction progress using a novel microfluidic channel apparatus.

A series of experiments were conducted using a microfluidic cell attached directly to the calcite (104) surface to assess surface reactivity under dynamic flow conditions. Calcite was reacted at room temperature with a pH 3.5, 5 mM Pb(NO<sub>3</sub>)<sub>2</sub> solution over a range of ionic strength adjusted by addition of NaClO<sub>4</sub>. These confined microfluidic environments enable us to precisely control fluid composition and fluid velocity within the microfluidic channels. We observed that variations in flow rate and ionic strength affect the calcite dissolution rate and morphology of secondary Pb-bearing precipitates within the flow channels. We also observed that the angle between the fluid flow direction and the crystallographic orientation of the calcite (104) surface has an effect on the resulting topography and distribution of the secondary phases.

Understanding these effects will enable us to predict emergent phenomena in geochemical processes involving metal-calcite interactions and long-term metal sequestration, pore-scale dissolution and nucleation during reactive transport in calcite-bearing mineral-water systems.

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