

A nanoscale investigation of the K-feldspar-solution interface after hydrothermal alteration in a concentrated alkaline solution: evidence for a CIDR mechanism of dissolution

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Because very little is known about how feldspars are chemically altered in aqueous fluids at high pH, we carried out a series of experiments exploring the dissolution behavior of an alkali feldspar (orthoclase) at hydrothermal conditions (190 °C) in a concentrated alkaline solution ($\text{Ca}(\text{OH})_2$, $\text{pH}_{(25^\circ\text{C})} = 12.3\text{--}12.5$) over a 24 h period*. At the end of the alteration experiments, the hydrothermal products were quantified by XRD-Rietveld refinement: aside from remnant orthoclase grain cores (11-21%), the most important crystalline authigenic phases were calcite (35-50%) and hydrated silicate minerals (CASH+CSH, 43-54%). The hydrothermal product was also enrobed in resin. Polished grain mounts, examined by FESEM, revealed intact orthoclase grains surrounded by a 1-2 μm -thick rim of secondary authigenic crystalline phases. Overall, the rims appear to be porous, and thus probably did not impede solvent access to the orthoclase grains. Ultrathin FIB-prepared TEM foils comprising both the outer edges of orthoclase grains and the precipitate rim allowed us to investigate at the nm-scale the near-surface chemistry and structure of the interfacial region. Even though the orthoclase grains diminished in volume due to dissolution, they remained chemically and structurally intact. There was no evidence that Ca from the solvent penetrated into the orthoclase structure. Quite surprisingly, we also discovered the existence of a variably thick (10-200 nm) but continuous amorphous surface altered layer (SAL) abutting the orthoclase grain edges. The structural and chemical boundary between the two phases is both sharp (at the nm-scale) and spatially coincident. The lack of a structural gap between the two phases suggests it formed in situ. STEM-EDXS measurements show that it is enriched in K and Ca, and somewhat depleted in Si, and significantly depleted in Al. This amorphous SAL most likely formed continuously during chemical alteration via a coupled interfacial dissolution-reprecipitation process (CIDR). Given that this mechanism is also suggested to control acid and neutral pH weathering of feldspars, our data let us infer that CIDR should be considered a universal mechanism of feldspar dissolution over a broad range of pH and temperature conditions.

*Hellmann et al., 2021, Chem. Geology; Zhai et al., 2021, Appl. Geochem.

