

NEW INSIGHTS ON MICA REACTIVITY AT THE MINERAL-SOLUTION INTERFACE

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The weathering behaviour of mica, and phyllosilicates in general, plays a key role in a wide variety of natural and engineered environments. Consequently, the reactivity of micas (e.g. biotite, muscovite, phlogopite, etc.) has been studied for a broad range of physicochemical solution conditions, probing, among others, the influence of pH, temperature and salinity. These studies used both macroscopic [e.g. 1] and microscopic [e.g. 2] approaches. Despite the many-fold insights gained from these studies, a general relationship between the solution properties and the nanoscale processes occurring at the mineral-solution interface is still missing. Moreover, the influence of surface controlled processes on the measured bulk dissolution rates also needs to be sorted out.

Thus, in order to establish a general framework between the physicochemical solution properties and the interface processes controlling mica alteration, we have studied in detail the surface reactivity of biotite, phlogopite and muscovite in contact with aqueous solutions for a broad range of pH's (1 to 6.8) and temperatures (23- 120 °C). Dissolution, hydration and (re-)precipitation processes occurring at the mineral-solution interface were observed making use of a custom-build hydrothermal atomic force microscopy [3]. These short-term *in situ* experiments were complemented with *ex situ* characterization of long-term and high temperature (up to 200°C) experiments conducted in hydrothermal batch reactors. The obtained microscopic data were correlated with previously obtained macroscopic dissolution rates.

Based on these experimental data we propose a tentative mica reactivity diagram, linking the surface mechanism(s) to the bulk dissolution behaviour as a function of the chemical composition and the physicochemical properties of the solution.

[1] Rozalén et al. (2008) *Geochim. Cosmochim. Acta* **72**, 422.

[2] Cappelli et al. (2013) *Cryst. Growth Des.* **13**, 2880.

[3] Jordan et al. (1999) *Geochim. Cosmochim. Acta* **63**, 3183.