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## The initial stage of mica weathering: Coupling in situ AFM observation to solution chemistry

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Although understanding mica alteration is crucial to the dynamics of water–rock interaction, studies on mica dissolution mechanisms at the nanometer scale are quite rare. In this study, we investigated the early stage of muscovite alteration combining chemical analysis of the interacting solution with in situ Atomic Force Microscopy determinations of the reacting mineral surface. Experiments were carried out using a liquid cell of an Atomic Force Microscopy determining simultaneously the evolution of the reacting fluids and the nanometers size details of the mineral surface. We selected the (001) basal face of a large monocrystal of muscovite from South India for this study.

The state of the muscovite monocrystal's pristine surface after cleavage was observed to be perfectly flat, with no apparent evidence of defect points or dislocations. The surface's molecular periodicity resolution showed the hexagonal-like formations typical of the nanoscale occurrence of Si-centered tetrahedral arrangements along muscovite sheets. However, when a solution was introduced in the liquid cell by way of a peristaltic pump, pits appeared rapidly displaying a stair-shape pattern consistent with the mineral structure of a T-O-T periodicity. This early stage evolution is responsible for the stoichiometric Al/Si ratio in solutions.

Our nanometer scale determinations allowed us to estimate the anisotropic loss of matter generated by the step movement. We found the directional lateral velocity V(hk0) to be 3 fold higher than the basal velocity V(00l), demonstrating the higher reactivity of the lateral compared to basal surfaces during the dissolution process. The directional velocity of dissolution at the lateral face decreased by more then one order of magnitude for a 5-fold increase in pH, while velocity at the basal face did not change significantly as a function of pH. We observed also the formation of new phases of nanometer size that coating the dissolving mineral surface inhibits the step movement with consequence loss in congruency. Coatings that initially form at the lateral higher reactive faces control the overall dissolution process. Coat formation and growth depends on both aqueous solution composition and dissolving mineral structure.