



Interface-coupled dissolution-precipitation processes during acidic weathering of multicomponent minerals

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The chemical weathering of carbonate and silicate minerals on the Earth's surface controls important geochemical processes such as erosion rates and soil formation, ore genesis or climate evolution. The dissolution of most of these minerals is typically incongruent, and results in the formation of surface coatings (altered layers, also known as leached layers). These coatings may significantly affect mineral dissolution rates over geological timescales, and therefore a great deal of research has been conducted on them. However, the mechanism of leached layer formation is a matter of vigorous debate. Here we report on an in situ atomic force microscopy (AFM) and real-time Mach-Zehnder phase-shift interferometry (PSI) study of the dissolution of wollastonite, CaSiO_3 , and dolomite, $\text{CaMg}(\text{CO}_3)_2$, as an example of surface coating formation during acidic weathering of multicomponent minerals. Our in situ results provide clear direct experimental evidence that leached layers are formed in a tight interface-coupled two-step process: stoichiometric dissolution of the pristine mineral surfaces and subsequent precipitation of a secondary phase (silica in the case of wollastonite, or hydrated magnesium carbonate in the case of dolomite) from a supersaturated boundary layer of fluid in contact with the mineral surface. This occurs despite the bulk solution remaining undersaturated with respect to the secondary phase. The validation of such a mechanism given by the results reported here completely changes the conceptual framework concerning the mechanism of chemical weathering, and differs significantly from the concept of preferential leaching of cations postulated by most currently accepted incongruent dissolution models.