Microstructural modifications of dissolving silicate minerals: Why should we bother?

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Providing experimental constraints on mineral dissolution and precipitation kinetics has mobilized geoscientists for over 40 years. The early development some 30 years ago of reactive transport codes to model fluid-rock interactions has maintained a continous interest in this work, with the ultimate need for the formulation of so-called 'kinetic rate laws' that could be implemented in such codes. Although the importance of reactive transport simulations for a wide range of geochemical concerns is no longer to be proven, the poor agreement between laboratory- and field-derived weathering rates casts doubt on the reliability and relevance of numerical simulation outputs. While the reasons for such discrepancies are manifold, as a starting point, we hypothesize that they mainly rely on the long suggested -though poorly studied- spontaneous evolution of the microstructural properties of dissolving mineral surfaces, also referred as to mineral ageing. This presentation is intended to bring new insights on this process, based on detailed analyses of dissolving cleavages of a series of silicate minerals, using state-of-the-art microscopic and spectroscopic techniques of surface characterizations (e.g. FIB-TEM, AFM, VSI, and nanoSIMS). In particular, we will show that (1) the transport properties of altered surface layers dramatically differ from that of the bulk solution and evolve with reaction the formation of local chemical progress, promoting environments in the vicinity of the inner altered surface/crystal interface, and (2) mineral dissolution rate is linked to the bulk fluid composition through relations which are specific to the crystallographic orientation.

Taken together, our results confirm the conclusion recently reached by some authors [1] that the classical derivation of empirical 'rate laws' based on the dissolution of freshly crushed powders may be misleading, and alternate models of mineral reactivity will be presented.

[1] Lüttge et al. (2013) Elements, 9, 183-188.