Comparing reactivity of glasses with their crystalline equivalents: the case study of albite

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Understanding the weathering processes of silicates is a crucial issue for several geological (C cycle and CO_2 budget) and environmental (CO_2 sequestration, nuclear waste disposal) concerns. While the dissolution reactions of numerous silicate minerals and natural glasses have been extensively investigated during the last decades, the parallel between glasses and crystals is rarely drawn. Moreover, for both substrates, the formation of amorphous Si-rich surface layers (ASSLs) during weathering and their passivating properties is not fully understood.

To evaluate the effect of crystallintity on the elementary reactions that govern dissolution rates of silicates, the dissolution of amorphous and crystalline albite was investigated as a function of pH at 90°C. Regarding albite crystal, the (001), (010), (10-1) and (1-11) faces were submitted to dissolution experiments in order to tackle the influence of crystal anisotropy on reaction rates. Experiments were also performed on powdered albite (all faces exposed) to evaluate the representativness of the face-resolved results regarding the global dissolution process. Two pH values at which ASSLs are either present (pH 1.5) or absent (pH 10), were studied. To constrain the effect of ASSLs on dissolution rates, experiments were performed in solutions saturated, or not, with respect to amorphous silica. In parallel, experiments were performed using solution initially doped with ^{29}Si to resolve small chemical changes and enligthen the mechanisms leading to glass/crystal dissolution and ASSL formation.

Dissolution rates at the interface between the ASSL and the pristine glass/mineral were determined from element and isotope budget in the solution. Reactions rates at the interface between the ASSL and the bulk solution were calculated on the basis of surface retreat data obtained with a vertical scanning interferometer (see [1] for details). This strategy allowed to reveal the chemical exchanges between glass/mineral and solution as a function of pH and to clarify the mechansisms of formation and the physico-chemical properties of ASSLs developing at this interface.

[1] Wild et al. (2016) *Chem Geol* **442**, 148-159.