

The properties of interfacial layers revealed by *in situ* X-ray reflectivity and vertical scanning interferometry

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Growing evidence point towards a control of silicate mineral dissolution rates by interfacial processes, especially the formation of amorphous silica surface layers (ASSLs), which have been demonstrated to control the dissolution rate for a wide range of silicate minerals. To date, however, the mechanisms underlying these processes at the nanometer scale remain elusive. Our study aims at providing new constraints based on a multi-technique investigation of the early stages of the dissolution of labradorite feldspar, a silicate representative of the continental crust.

Our approach combines state-of-the-art experimental and computational tools that aims at identifying key parameters driving the *in situ* evolution of ASSLs. Both their thickness and density were simultaneously probed at the nanoscale by synchrotron-based X-ray reflectivity (XRR) in the course of labradorite dissolution conducted in a home-made cell at 80°C for bulk pH values ranging from 1.5 to 4. The reactivity of ASSLs could be independently assessed by measuring the temporal evolution of the retreat of the external fluid-layer interface by vertical scanning interferometry (VSI).

These data were used in combination to computational methods including reactive transport modeling to back calculate the local physico-chemical conditions prevailing at the inner layer-mineral interface, which cannot be probed directly.

Overall, our study provides new insights into the mechanisms underlying the formation ASSLs and their consequences in the control of reactions at the mineral-fluid interface.