

Conversion of rock to saprolite: A study of weathering rinds

ALEXIS K. NAVARRE¹, CARL I. STEEFEL²
AND SUSAN L. BRANTLEY³

¹Department of Geosciences, Pennsylvania State University,
University Park, PA 16801, USA

(anavarre@geosc.psu.edu)

²Earth Sciences Division, Lawrence Berkeley National
Laboratory, 1 Cyclotron Road, Berkeley, CA 94720, USA
(CISteefel@lbl.gov)

³Department of Geosciences, Pennsylvania State University,
University Park, PA 16801, USA (brantley@essc.psu.edu)

Weathering rinds can be defined as small-scale saprolites that develop on clasts. Developed in a controlled environment and protected from physical weathering by surrounding material, these clasts provide a unique setting to examine chemical weathering processes without additional complications due to the removal of weathered material by physical erosion. Basalt clasts with 3 cm weathering rinds collected from 125 ka alluvial terraces along the Pacific coast of Costa Rica have been analyzed with SEM backscatter and EDS images, EMP, and XRD. These data, combined with reactive transport simulations, have been used to characterize the reaction front and develop a conceptual model of the chemical processes that occur during saprolite formation.

Due to low parent porosity (<1%), initial transport of reactants and products to and from the reaction front occurs primarily by diffusion. Small amounts of iron oxide have been observed precipitated around augite crystals and in fractures coreward of the reaction front, but the dissolution of labradorite and precipitation of kaolinite is the first chemical change to occur at the reaction front that results in increased porosity. As weathering proceeds and porosity increases, fluid is able to flow through the rind (~50% porosity). This change in the dominant transport mechanism results in an influx of water with low silica concentrations and the replacement of kaolinite by gibbsite, an effect which is not possible with diffusive transport alone. Chemical data suggests that the diffusion dominant zone is <0.5 mm thick and separates unaltered rock from high porosity rind.

The rate of rind advance in this system is slow (2.4×10^{-4} mm/yr) compared to basalt weathering at the landscape scale (0.1-1.2 mm/yr) for the same climate. Most landscape scale rates are from large basaltic provinces. These basalts are extrusive with high porosity. Porosity has been indicated to have a primary control on weathering rind thickness and therefore weathering rate. Other possible causes for faster rates at larger scales could be the coupled effect of physical and chemical weathering.

Microscopic reactive diffusion of U(VI) in subsurface sediments: Characterization and modeling

C. LIU, P.D. MAJORS, J.M. ZACHARA
AND J.P. MCKINLEY

Pacific Northwest National Laboratory, Richland, WA 99352,
USA

Uranium in the US Department of Energy (DOE) Hanford 200 Area sediments was found to be distributed as uranyl silicate precipitates (Na-boltwoodite) predominantly within interiors of sediment granitic clasts and preferentially in a minor plagioclase feldspar component. The precipitates were minute, generally 1-3 μm across in either radiating or parallel arrays in microfractures of a few microns width and variable connectivity to particle surfaces (Liu et al, 2004). The uranyl precipitates dissolved and released from intraparticle regions to undersaturated aqueous solutions ($\text{Na-NO}_3\text{-HCO}_3$) at a rate of over two orders of magnitude slower than the intrinsic dissolution rate of Na-boltwoodite. The results indicated that diffusion limited the dissolution release of the uranyl precipitates. X-ray microprobing the precipitates before and after dissolution confirmed this conclusion.

A pulse-field gradient nuclear magnetic resonance (PFG-NMR) approach was developed to characterize microscopic diffusion. The approach uses water (H_2O) itself as a tracer and avoids mass displacement procedures. Measurements with PFG-NMR identified at least two intraparticle diffusion regions with H_2O diffusivities of over 2 orders of magnitude difference. The fast diffusion region has an apparent tortuosity of about 1.5 and appears associated with the waters on particle surfaces and in large intraparticle fractures, while the slow one has an apparent tortuosity of about 350 and is associated with secondary fractures. The two diffusion regions were generally confirmed by the NMR imaging of H_2O distribution as a function of water content.

A model was assembled to simulate reactive diffusion of radioactive waste into the intraparticle microfractures. Simulations indicated that waste U(VI) preferentially precipitated and concentrated within the feldspar microfractures where the saturation of Na-boltwoodite was maintained by the diffusive supply of U(VI) and Na from waste, and dissolution of silicates for Si.

Reference

Liu C., Zachara J.M., Qafoku O., McKinley J.P., Heald S.M., and Wang Z., *Geochim Cosmochim Acta* **68**, 4519-4537.