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Elucidating Clast-Scale Transport-Reaction Dynamics during Shale Weathering

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Chemical reaction rates at the molecular level are intimately connected to the local environmental chemistry, such as solute concentrations at the fluid-mineral interface. However, in a range of geomaterials, these rates are predominantly determined by fluid transport properties at the clast scale. To elucidate the impact of pore structure on reaction fronts at this pivotal scale, we employed a suite of imaging, spectroscopy, and neutron scattering techniques to analyze rock fragments from weathering profiles of shale at Pennsylvania, USA.

In an outcrop of Marcellus Shale, characterized by high organic matter (OM) content and variable carbonate levels, weathering manifests as an almost complete depletion of sulfides, carbonates, plagioclase, and chlorite. This process is accompanied by an increase in the number, size, and connectivity of pores following carbonate dissolution and OM decomposition. Intriguingly, only approximately 60% of the original OM is depleted at the land surface, with larger OM particles being preferentially consumed, while smaller OM particulates, intimately associated with clays, remain largely intact during weathering. The weathering profiles are capped by a layer rich in carbonates (over 85 wt%), where porosity and pore connectivity decrease significantly compared to the weathered Marcellus shale, and no evidence of sulfide oxidation is present. Conversely, the weathered Rose Hill Shale, a gray shale with lower OM content and varying carbonate levels (0-20 wt%, scattered or vein-filled), exhibits smaller pore sizes and porosity than its Marcellus counterpart. Notably, in carbonate-rich clasts of Rose Hill Shale, sulfide oxidation and porosity generation are more pronounced than in carbonate-lean clasts from similar depths. These observations underscore the critical role of rock texture and the presence and distribution of rapidly dissolving minerals in dictating the extent of weathering through transport-reaction dynamics.