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The Effects of Chemomechanical Processes on Limestone Weathering Rates

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Dissolution within porous media is a critical process in many environmental and geological settings. The formation and evolution of soils, rocks, and landscapes (Buhmann and Dreybrodt, 1985; Brantley, 2008; Jin et al., 2010), efficiency of carbon capture in geological reservoirs (Matter and Kelemen, 2009), and the weathering of man-made structures are all highly dependent on the rates at which minerals dissolve. However, the factors governing dissolution in geological media are not yet well understood. In general, rock dissolution rates are governed by fluid composition, fluid flow rates, and the surface area of the mineral–fluid interface (Lasaga, 1998). As a surface becomes rougher, the surface area in contact with a reactive fluid increases, and it is therefore often assumed that rough surfaces dissolve more rapidly than smooth surfaces (Fischer and Lüttge, 2007). However, a number of studies have shown a complex relationship between roughness and reactivity (e.g., Anbeek, 1992; Gautier et al., 2001). Emmanuel and Levenson (2014) found that erosion rates in fine-grained micritic limestone blocks are as much as two orders of magnitude higher than rates estimated for coarse-grained limestones. AFM imaging suggested this is the result of rapid dissolution along micron-scale grain boundaries, followed by mechanical detachment of tiny particles from the surface. Such chemomechanical processes may be the dominant erosional mode for fine-grained carbonate rocks in many regions on Earth. This erosion extends to the micron scale, and grain detachment can be a crucial mechanism controlling denudation rates in carbonate terrains.

In order to better understand this weathering process we undertook a series of experiments looking at the weathering of carbonates. 5/8" x 5/8" cores of four limestones (Netzer, Shiuvta, Carthage Marble, Texas Cream) were exposed to flowing water at 30°C and several pH values to mimic the weathering process. Annular Cd masks of stepped sizes, and small beam stepped locations analyses, were used to analyze the weathering structure by (U)SANS and (U)SAXS as a function of distance from the edge. SEM analysis was also used to look at the pore structure and surface weathering. The results, both in terms of core/rim variations and pore size dependence, were found to be strongly dependent on initial permeability and rock structure, as well as time and pH.

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