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Measuring Dissolution of Calcium in Natural Limestone via Microscopy and ICP-MS

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Understanding how chemical reactions impact the dissolution rate of rock surfaces over time is critical in subsurface energy activities such as geothermal energy recovery, oil and gas extraction, and carbon storage. Mineral dissolution can increase reservoir permeability as the dissolution can generate permeable pathways, enhancing energy recovery, while other coupled processes such as weakening of mechanical properties may have an opposite effect. To investigate the effects of chemical reactions on dissolution of a rock surface, experimental studies are conducted using solutions with three different pH levels of 4, 3, and 2 on the surface of an 18 mm x 25 mm polished limestone. This is achieved using a 3D-printed fluidic device that maintains a constant flow rate of 100 μL/hr. Effluent is collected about weekly to monitor the calcium concentration (Ca2+) using ICP-MS for assessing the changes of dissolution rate. To gain a clearer understanding of the effects of dissolution along the flow path of the limestone sample, topographic images are captured weekly —one directed towards the inlet and another towards the outlet—at a horizontal resolution of 0.45 μm and a vertical resolution of 0.83 μm using a Zeiss LSM 900 confocal microscope. These images are then compared with the Ca2+ concentration to better understand how dissolution occurs both across time and along the length of the flow path. The results indicate that at pH=2, the dissolution is 5x that at pH=3 when measuring Ca2+. Then, within the imaging results, the inlet is shown to have a roughly 2.5x larger dissolution rate than the outlet of the sample for both pH=3 and pH=2. Knowing both the Ca2+ concentration and the changes in topography across time, we can further analyze the impacts of pH on dissolution rates over the rock surface roughness and how it may alter surface asperities and change permeability within a reservoir. The next step of this research involves conducting experiments at pH=4 and comparing the results to those obtained at pH=2 and pH=3. Additionally, we will create channels along the limestone surface to investigate how a defined channel affects dissolution across the rock surface. SNL is managed and operated by NTESS under DOE NNSA contract DE-NA0003525.

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References

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