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Direct Visualization of X-ray Enhanced Mineral Dissolution

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Complex dissolution and precipitation processes provide a challenge for the removal and remediation of nuclear waste. Of particular interest are aluminum bearing phases such as gibbsite and boehmite which constitute a large fraction of tank waste at legacy sites such as the Hanford Nuclear Reservation in Washington State. These minerals are formed under complex chemical and radiological conditions, far from equilibrium, which can be difficult to replicate in an experimental setting. Developing a fundamental understanding of radiation induced processes is of particular interest. The Interfacial Dynamics in Radioactive Environments and Materials (IDREAM) Energy Frontier Research Center is dedicated to the development of experimental and computational methods necessary for understanding interfacial processes in such environments to determine key factors that underpin crucial processes such as dissolution.

In this work the integration of atomic force microscopy and X-ray irradiation for the direct visualization of gibbsite dissolution is presented [1-2]. This initial ex situ experimental work compared dissolution rates in 0.1 M NaOH by directly measuring pre/post dissolution morphology of individual gibbsite particles. A nearly two-fold increase in dissolution rate was observed when dissolution was carried out under X-ray irradiation. Surprisingly, this enhancement was present even outside the X-ray irradiation region. This suggests radiolytic products, especially longer-lived species such as peroxide, play a primary role in dissolution. Irradiation was also carried out under inert dry conditions prior to dissolution to exclude the influence of these radiolytic products. In this case the dissolution rate was nearly identical, but only at the X-ray center suggesting direct X-ray damage also plays a significant role. This approach augments bulk measurement techniques, such as ICP by allowing for the comparison of particle dissolution based on individual particle morphology/crystallinity, defects, and aggregation. This work paves the way for in situ observations of radiation induced dissolution under a wide range of experimental conditions providing a platform for better understanding these complex systems.

References

[1] Riechers, et al. (2021), *Rev. Sci. Instrum. 92*, 113701

[2] Riechers, et al. (2021), *Commun. Chem.* 4, 1-7