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Induced Goethite Mineral Dissolution to Probe the Chemistry of Radiolytic Water in Liquid-Phase Transmission Electron Microscopy

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Liquid-phase transmission electron microscopy (LP-TEM) allows for *in situ* observations of the dynamic behavior of materials in liquids at high spatial and temporal resolution. As a result, LP-TEM is a very promising technique for mechanistic investigations of solid/water interface reactions under highly controlled conditions. However, one of the main drawbacks of LP-TEM comes from the interaction of the incident electron beam with water molecules within aqueous solutions. This drawback is directly dependent upon the number of electrons (electron flux density) scattering throughout the liquid cell and changing the chemical environment of the reaction to be observed. When the beam is interacting with the aqueous media, the water molecules are initially decomposed into primary species such as hydrated electrons e_h^-, hydrogen radicals $H^\bullet$, hydroxyl radicals $OH^\bullet$, or gaseous hydrogen H_2. All of these species distribute in the surrounding liquid via diffusion and lead to further reactions with both other water molecules and/or with the target solid sample. Therefore, the acidity as well as the redox chemistry of the aqueous solution may be significantly changed, largely influencing the phenomena under observation. In this study, we assessed the electron beam induced aqueous dissolution kinetics of nano-goethite *in situ* and evaluated the radiation effects within the liquid cell. Our results show that, on one hand, morphological changes in the monitored goethite during its interaction with low electron flux densities were associated with acidic dissolution, although the dissolution rates were ten times larger than those measured under bulk dissolution conditions. On the other hand, at higher electron flux densities, dissolution was enhanced, and the morphological evolution of the nano-particulate goethite structure did not anymore match solely acidic dissolution. Together with kinetic simulations of the steady state concentrations of generated reactive species in the aqueous medium, our results provide a unique insight into the interplay between redox and acidity and effects induced by radiation chemistry in LP-TEM studies.