Transformation of Ferrihydrite in High-Salinity Solutions with Various Sulfate Ratios: Implications for Formation of Goethite in Gale Crater on Mars

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Iron oxides formed through water-rock interactions on early Mars. The rover Curiosity has reported ferrihydrite and hematite as the dominant iron oxides in Gale crater [1]. Ferrihydrite is a poorly crystalline iron oxide, while hematite and goethite are crystalline iron oxides produced by the transformation of ferrihydrite in aqueous solutions [2]. Although ferrihydrite transforms to goethite under acidic conditions [3], it has been reported that hematite forms in highly saline solutions even under acidic conditions [4]. This explains the scarcity of goethite on Mars. However, in addition to hematite, substantial amounts of goethite were identified for the first time in several sediments in Gale crater [5]. The presence of collapsed phyllosilicates and highly soluble Mg sulfates, and the lack of carbonates in these sediments indicate acidic and highly saline water containing sulfate ions [6]. Such water resembles acid mine drainage on Earth, where goethite precipitation dominates. Thus, we hypothesized that goethite formed in sulfate-rich water. However, since Fukaya et al. (2025) investigated the ferrihydrite transformation in chloride solutions, the influence of sulfate under high-salinity conditions was not considered. Therefore, to clarify why significant goethite is present in Gale crater sediments even under high-salinity conditions, this study examined the transformation of ferrihydrite in acidic solutions with a constant ionic strength of 1 mol/kg while systematically varying the SO_4^{2-} ratio (= $SO_4^{2-}/(Cl^-+SO_4^{2-})$), using X-ray absorption spectroscopy.

When the SO_4^{2-} ratio was 0%, ferrihydrite transformed solely into hematite. In contrast, the proportion of goethite in the transformation products increased with SO_4^{2-} ratio. At an SO_4^{2-} ratio of 100%, ferrihydrite transformed solely into goethite. These results suggest that goethite in Gale crater may have formed from acidic and high-salinity waters containing sufficient sulfate ions. These findings imply that hematite formed in chloride-rich water rather than by thermal dehydration of goethite on the Martian surface.

[1] Blake et al. (2024), *Minerals* 14, 568. [2] Cornell and Schwertmann (2003), *The Iron Oxides*. [3] Schwertmann and Murad (1983), *CCM* 31, 277-284. [4] Fukaya et al. (2025), *ACS Earth Space Chem* 9, 1654-1666. [5] Chipera et al. (2023), *JGR*