

122 - Transformation of hexagonal birnessite into triclinic birnessite by aqueous Mn(II) and the formation of todorokite

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Birnessites, the most common family of manganese oxides, affect the fate and transport of toxic metal(loid)s, many nutrient elements and organic substances in the environment. However, their formation and transformation pathways are still poorly understood. Acid birnessite and vernadite, which are hexagonal birnessites and enriched with Mn(IV) vacant sites and lack of significant structural Mn(III) and Mn(II), are common analogues to naturally-occurring birnessites. Here we investigated the transformation products of acid birnessite and vernadite after being reacted with low concentrations of dissolved Mn(II) under alkaline conditions and the possibility of the transformation of the products into tunnel structure todorokite. The results showed that the hexagonal birnessites reacted with Mn(II) at pH \geq 8 transformed into triclinic birnessite. The transformation can be ascribed to the formation of Mn(III) via the conproportionation reaction between adsorbed Mn(II) on vacant sites and the surrounding Mn(IV) and the subsequent Mn(III) incorporation into the vacant sites. The Mn(III) had a high concentration with an ordered distribution within the structure of the transformed triclinic birnessites as the birnessites can be further converted into todorokite after Mg²⁺ exchanged and refluxed under the atmospheric pressure. The transformation was faster at higher pH, suggesting that alkaline conditions may favor each of the transformation steps. Compared with acid birnessite, the transformation of vernadite into triclinic birnessite was much faster, indicating that the crystallization and turbostratic degrees also affected the transformation.

This study for the first time directly observed the transformation from birnessites with hexagonal symmetry into those with triclinic symmetry, and systematically demonstrated the complete transformation sequence of the three naturally-abundant Mn oxides in the surface environment, i.e., hexagonal birnessite \rightarrow triclinic birnessite \rightarrow todorokite. The study also revealed the critical role of Mn(II) in mediating the transformation and accordingly controlling the Mn oxide mineralogy and redox activities.

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