
Role of Structural Hydrogen in the Reduction and Reoxidation of Iron in Nontronite

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Abstract: The effects of reduction and reoxidation of octahedral Fe^{3+} on the exchange of structural hydrogen in nontronite were determined using tritium (^3H) as a label element. The uptake of H from the surrounding solution of nontronite suspensions increased as the reduction of structural Fe^{3+} increased. Similarly, the loss of H from the structure increased as the reduction increased. The results are generally consistent with a reduction mechanism involving the loss of structural OH, leaving the affected Fe sites with less than six-fold coordination. The attenuation of increased negative charge on the clay layer, however, was less than predicted by such a mechanism.

During the reoxidation of reduced nontronite in suspension, about one-third of the H remaining as part of the structure following reduction was lost, whereas twice that amount of H was incorporated into the structure from the surrounding solution. A reoxidation mechanism is proposed whereby H_2O from the surrounding solution is incorporated into the mineral structure followed by the elimination of a hydrogen ion, returning the Fe to six-fold coordination. This mechanism implies the reversibility of Fe reduction in nontronite.

Key Words: Hydrogen • Nontronite • Oxidation • Reduction • Structural sites • Tritium

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