## Formation of saponite by hydrothermal alteration of metal oxides: Implication for the rarity of hydrotalcite

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## ABSTRACT

Conversion of hydrotalcite (Ht) to saponite was observed by hydrothermal alkaline alteration of metal oxides. The conversion was through a pathway of hydration-dissolution-precipitation. It involved several critical steps, including the construction of Ht from metal oxides, dissolution of  $Al^{3+}$  from Ht, condensation of metasilicate anions with Ht, and finally crystallization of saponite. The condensation was favored by relatively low Mg/Al ratios of Ht, along with high concentrations of  $Al^{3+}$  and silicate oligomers in the environment, resulting in highly crystalline saponite. The latter conversion was greatly accelerated by the isomorphous substitution of  $Al^{3+}$  for  $Si^{4+}$  in silicate oligomers. The substitution generated the extra negative charge and led to the aforementioned condensation with Ht surface, thereby promoting the formation of saponite TOT layers. During the process,  $CO_2$  is an indispensable component. Initially intercalated as  $CO_3^{2-}$  to form Ht,  $CO_2$  was subsequently eliminated from the solid phase, and saponite form dwhen the layer charge was reversed. Thus, this study presents a novel formation mechanism of saponite from metal oxides via hydrotalcite and contributes to a better understanding of the crystallization, chemical stability, and transformation of Ht to saponite. The results are also relevant to evaluating metal availability and carbon cycling on the surface of the Earth.

Keywords: Smectite, carbonate, hydrotalcite, mineral transformation, saponite