

The evolution of saponite: An experimental study based on crystal chemistry and crystal growth

CHAOQUN ZHANG^{1,2,4}, HONGPING HE^{1,3,4,*}, SABINE PETIT^{2,*}, FABIEN BARON², QI TAO^{1,†},
BRIAN GREGOIRE², JIANXI ZHU^{1,3,4}, YIPING YANG^{1,4}, SHICHAO JI^{1,4}, AND SHANGYING LI^{1,4,‡}

¹CAS Key Laboratory of Mineralogy and Metallogeny/Guangdong Provincial Key Laboratory of Mineral Physics and Materials, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou, 510640, China

²Institut de Chimie des Milieux et Matériaux de Poitiers (IC2MP), UMR 7285 CNRS, Université de Poitiers, F-86073 Poitiers Cedex 9, France

³CAS Center for Excellence in Deep Earth Science, Guangzhou, 510640, China

⁴University of Chinese Academy of Sciences, Beijing, 100049, China

ABSTRACT

Element incorporation and partitioning during the evolution of clay minerals have significant implications for element cycling in geochemical processes. The main aim of this experimental study is to further our understanding of element redistribution and crystal growth during smectite evolution under different physicochemical conditions. The precursors (i.e., pure Mg- and Ni-saponite) were separately prepared by hydrothermal syntheses at the same set of temperatures (i.e., RT, 50, 150, 180, 200, and 220 °C) for one day. Then the starting materials were obtained from the mechanical mixtures of the identical molar weight of Mg- and Ni-smectite precursors prepared at the same temperature. Subsequently, Series I samples were obtained by hydrothermally treating different starting materials at 220 °C for two weeks while Series II samples were hydrothermally synthesized under various temperatures (220, 300, 400, and 500 °C) for one week using the starting materials prepared at 220 °C. Both the precursors and resultant saponites were characterized by XRD, FTIR, TEM, and STEM. The FTIR spectra of the precursors only exhibit the $\nu\text{Mg}_3\text{OH}$ and $\nu\text{Ni}_3\text{OH}$ bands, corresponding to Mg-saponite and Ni-saponite, respectively. However, the occurrence of $\nu\text{Mg}_2\text{NiOH}$ and $\nu\text{Ni}_2\text{MgOH}$ bands in the resultant saponite indicates the dissolution of the corresponding Mg- and Ni-saponite precursors and recrystallization of Mg-Ni mixed saponite. The dissolution extents of Mg- and Ni-saponite precursors, which affect the degrees of random distribution of octahedral Ni and Mg in resultant Mg,Ni-saponite, are significantly controlled by the temperature gap (ΔT) between the precursors prepared and the resultant Mg,Ni-saponite obtained. In general, a larger ΔT leads to a higher dissolution extent of saponite precursors and a higher degree of random distribution of octahedral Ni and Mg cations in the resultant Mg,Ni-saponite structures. Thus, the distribution mode of octahedral cations in saponite, which is not only relevant to a given hydrothermal temperature but also dependent on ΔT for final products, cannot be used as a geothermometer. TEM and STEM observations provide visual evidence that the particles of saponite coarsen when ΔT is higher than zero. Both the crystal-chemical and morphological features during saponite evolution suggest that saponite particles coarsen mainly via partial/complete dissolution of precursors followed by recrystallization and growth of Mg,Ni-saponite in which crystal growth by layer attachment cannot be excluded. This study presents an experimental approach to evaluate the evolution of clay minerals in terms of crystal chemistry and crystal growth and offers a better understanding of the contributions of clay mineral evolution to element cycling.

Keywords: Smectite, crystal growth, crystal chemistry, cation distribution, clay evolution