

Conversion of serpentine to smectite under hydrothermal condition: Implication for a solid-state transformation

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Understanding clay mineral transformation is of fundamental importance to grasping phyllosilicate crystal chemistry and unraveling geochemical processes. In this study, hydrothermal experiments were conducted on lizardite and antigorite, to investigate the possibility of the transformation from serpentine to smectite, the effect of precursor minerals' structure on the transformation and the transformation mechanism involved. The obtained products were characterized by XRD, TG, AFM, HRTEM and ²⁷Al MAS NMR. The characterization results show that both lizardite and antigorite can be converted to smectite, but such conversion is much more difficult than that of kaolinite group minerals. The successful transformation is mainly evidenced by the occurrence of the characteristic (001) reflection of smectite at 1.2 - 1.3 nm in the XRD patterns and smectite layers with a thickness of 1.2 - 1.3 nm in HRTEM images of hydrothermal products as well as the dehydroxylation of the newly formed smectite at a higher temperature in comparison to that of precursor minerals. The difficulty for the transformation of serpentine to smectite may be due to the lack of enough available Al in the reaction system, in which the substitution of Al³⁺ for Si⁴⁺ in the neo-formed tetrahedral sheet is critical to control the size matching between the neo-formed tetrahedral sheet and octahedral sheet in precursor minerals. Since the neighboring layers in antigorite are linked by the strong Si-O covalent bond, the transformation only takes place at the edges of an antigorite layer rather than the whole layer, hindered by Si-O covalent bonds between two adjacent layers, and the resulting smectite is non-swelling. This reflects that the Si-O covalent bonds linking the adjacent layers in precursor antigorite are inherited by the transformation products, suggesting a solid-state transformation mechanism involved. AFM observations of the hydrothermal products from lizardite demonstrate that the transformation starts from the edge of precursor mineral and extends towards the inner part with extension of hydrothermal treatment time. The edges of the hydrothermal products is composed of neo-formed smectite layers while the central part contains identical number of lizardite layers. This strongly suggests that a solid-state transformation mechanism is involved in the conversion of serpentine to smectite.