

## Can 1:1 type clay minerals transform into 2:1 type swelling smectite?

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Clay minerals are ubiquitous in the Earth's crust and they may transform from one to another under natural and anthropogenic conditions. Thus, understanding clay transformation is of fundamental importance to unraveling geological processes and to synthesizing clay-based materials. To date, two pathways have been identified, i.e., the transformation among 2:1 type clay minerals (e.g., illitization of smectite) and from 2:1 type to 1:1 type (e.g., kaolinization of smectite). However, the transformation of 1:1 type to 2:1 type has not been reported except the case that smectite might form from serpentine and/or chlorite, but the transformation mechanism was unidentified.

Our results demonstrate a transformation mechanism from 1:1 to 2:1 clay type under hydrothermal condition, where individual or polymers of Si-O tetrahedra attach to the octahedral sheet of precursor 1:1 clay minerals to form 2:1 smectite. The transformation from dioctahedral kaolinite and halloysite to smectite was much easier than the one from trioctahedral serpentine to smectite. The presence of Al<sup>3+</sup> in solution accelerated the growth of smectite and improved its crystallinity, implying that the size matching between tetrahedra and octahedra was a key factor in controlling the transformation. Such a transformation mechanism can well explain a "polar layer" structure in 2:1 type phyllosilicates, i.e., the two tetrahedral sheets across the octahedral sheet have different composition and layer charge. Furthermore, our finding provides a mechanism for synthesizing valuable swelling smectite from inexpensive 1:1 clays. By optimizing hydrothermal conditions, different types of smectites may be synthesized to meet these diverse demands.