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# A reinvestigation of smectite illitization in experimental hydrothermal conditions: Results from X-ray diffraction and transmission electron microscopy

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## Abstract

The hydrothermal reactivity of the <1 μm fraction, K-saturated SWy-2 Wyoming low-charge montmorillonite was studied in the 250–400 °C temperature range with reactions lasting between 5 and 120 days, with a solid/solution mass ratio of 1:10, and in 1 mol/L KCl solution. From X-ray diffraction (XRD) profile modeling results on K-saturated and ethylene-glycol solvated samples the illitization process appears to occur as a progressive replacement of expandable layers by layers with illitic behavior, in a single illite-smectite phase. However this treatment overestimates the amount of illite layers because of the presence of smectitic non-expandable layers. This was revealed by calcium exchange of the products, which causes re-expansion of the apparent illite layers. The illitization model then obtained consists of four phases with increasing illite content: discrete smectite, a randomly interstratified mixed-layered mineral (MLM) and two ordered MLMs that progressively replace the phases with less illite content. This polyphase model is consistent with results from transmission electron microscopy and chemical microanalysis, which show several types of particle morphology with different interlayer K content and Al-for-Si substitution that seem to correspond to the several phases detected by XRD. Thus, the actual illitization mechanism of smectite in our experiments is not a progressive reaction but a dissolution-precipitation process following the Ostwald step rule in which metastable smectite transforms into illite through a series of metastable illite-smectite phases.

**Keywords:** Smectite, illitization, X-ray diffraction, transmission electron microscopy