## Characterization of mixed-layer illite-smectite from bentonites using microscopic, chemical, and X-ray methods: Constraints on the smectite-to-illite transformation mechanism

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## ABSTRACT

We studied mixed-layer illite-smectite (I/S) (<1 or 0.5  $\mu$ m size fractions) in 28 diagenetic bentonite samples, Pliocene to Ordovician in age, from different locations in North America and Europe to investigate the smectite-to-illite transformation mechanism. XRDmeasured illite contents ranged from 3 to 100%, and layer ordering varied continuously from R = 0 to  $R \ge 3$ . XRD polytype analysis showed that with progressive illitization, I/S changes from a predominantly cis-vacant to a trans-vacant structure, although considerable scatter is present in the data. There is no evolution of illite in the I/S from  $1M_d$  to 1Mpolytypes with progressive illitization. Microprobe chemical analysis and combustion analysis for N showed that  $NH_4^+$  comprised 9–22% of the total fixed cation content. We calculated an interlayer charge for the illite end-member of 1.5 per  $O_{20}(OH_4)$ . Sample morphology was studied by atomic force microscopy, which showed a constant aspect ratio and particle diameter over the whole I/S series. Particle thickness increased with percent illite. Oxygen isotope compositions of the I/S clays did not display any systematic trends. Our results suggest that illitization of bentonite I/S proceeds via a solid-state mechanism, with local lattice rearrangements that permit structural (octahedral cis/trans occupancy) and chemical changes in 2:1 layers and cause interlayer coalescence.