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# Reaction Mechanisms of Smectite Illitization Associated with Hydrothermal Alteration from Ponza Island, Italy

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**Abstract:** A hydrothermally altered rhyolitic hyaloclastite from Ponza island, Italy, has four alteration zones with unique clay assemblages: (1) a non-pervasive argillic zone characterized by smectite; (2) a propylitic zone with interstratified illite-smectite (I-S) containing 10– 85% illite (I); (3) a silicic zone composed of I-S with  $\geq 90\%$  I and pure illite; and (4) a sericitic zone with I-S ranging from 66% I to pure illite. Atomic force microscopy reveals abrupt changes in particle morphology with illitization, including initial changes from anhedral plates to laths and then to euhedral plates and hexagonal plates. I-S particles progressively thicken with illitization and mean particle area (basal plane) remains constant from pure smectite to I-S with 80% I. However, particle area increases from 90 to 100% illite. Computer modeling of I-S structural forms indicates octahedral cation ordering progressively changes from *cis*-vacant smectite to interstratified *cis*- and *trans*-vacant I-S, and then to *trans*-vacant illite. In addition, polytypes progressively change from  $1M_d$  to  $1M$ , and then to  $2M_1$  illite. Electron-microprobe and X-ray fluorescence analyses show that I-S chemistry progressively changes during illitization, evolving toward a phengitic composition with  $\sim 0.89$  fixed interlayer  $K^+$  per  $O_{10}(OH)_2$ . Octahedral  $Mg^{2+}$  shows little change with illitization, varying from 0.3 to 0.5 cations per  $O_{10}(OH)_2$ . The layer charge of smectite is  $\sim 0.38$  equivalents per  $O_{10}(OH)_2$ .

On the basis of abrupt changes in morphology and progressive changes in polytype and chemistry, smectite illitization on Ponza involved a dissolution and recrystallization mechanism with multiple stages of nucleation and crystal growth. In this multi-step model, temperature of alteration provided the major control for the layer composition, polytype, and morphology of I-S crystallites. Other factors that may play a secondary role include:  $K^+$  availability, water-rock ratio, and permeability. Alternatively, the mechanism of I-S and illite formation at Ponza and other hydrothermal environments may occur by direct precipitation of I-S crystallites from rhyolite glass and may not involve progressive reactions of smectite precursors.

**Key Words:** Atomic Force Microscopy • Chemical Analysis • Illite • Polytype • Smectite • XRD

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