Ordered Interstratification of Dehydrated and Hydrated Na-smectite

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Abstract: The 001 spacing of Na-smectite was found to vary from 9.6 Å at 0% relative humidity (RH) to 12.4 Å at 60–65% RH. The 9.6-Å spacing corresponds to dehydrated Na-smectite, and the 12.4-Å corresponds to Na-smectite with one water layer. A regular series of intermediate values resulted from ordered interstratification of the 9.6- and 12.4-Å units. Ordered interstratification was confirmed by the presence of a 001 spacing of 9.6 + 12.4 Å = 22 Å. This peak appeared under experimental conditions at about 35% RH. It appeared for calculated simulations of ordered stacking of 50/50 mixtures (± 10%) of 9.6- and 12.4-Å units. The 004 peak of this 22-Å spacing interacted with the 002 of the 9.6-Å spacing of ordered mixtures of more than 50% 9.6-Å units and with the 002 of the 12.4-Å spacing of ordered mixtures of more than 50% 12.4-Å units. The result of this interaction was a complex peak, the position of which was a function of the ratio of 9.6- and 12.4-Å units. This complex peak was noted for experimental and for calculated conditions. Calculated tracings assuming ordered stacking matched the experimental tracings closely, whereas those assuming random stacking did not.

Ordering was apparently due to the interaction of the positive charge of the interlayer cation repelling the positive charge of the hydrogens of the hydroxyl ions, one above and one below, closest to the interlayer space. The collapse of a single interlayer space (dehydration) brought the interlayer cation closer to the hydrogens of the hydroxyls causing the hydroxyls to rotate such that the hydrogens shifted toward the adjacent interlayer spaces. Collapse of these two interlayer spaces was therefore more difficult. This same mechanism helps explain ordering in illite/smectite. The difference is that hydration/dehydration is quick and reversible, whereas the change from smectite to illite is slow and irreversible.

Key Words: Hydration • Hydrogen positions • Illite • Interstratification • Smectite • Water • X-ray powder diffraction

Clays and Clay Minerals; August 1986 v. 34; no. 4; p. 379-384; DOI: 10.1346/CCMN.1986.0340404

Clay Minerals Society (www.clays.org)