High-Resolution Transmission Electron Microscopy and Electron Diffraction of Mixed-Layer Illite/Smectite: Experimental Results

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Abstract: High-resolution transmission electron microscopy (HRTEM) and electron diffraction experiments have been performed on R1 and R> 1 illite/smectite (I/S) samples that from X-ray powder diffraction (XRD) experiments appear to contain well-ordered layer sequences. The HRTEM images confirmed earlier computer image simulations, which suggested that periodicities due to I/S ordering can be imaged in TEM instruments of moderate resolution. The experiments also confirmed that in instruments of this sort, the strongest contrast arising from the compositional difference between I and S layers occurs under rather unusual imaging conditions of strong oveffocus. Some selected-area electron diffraction (SAD) patterns showed additional diffraction spots consistent with R1 and R3 ordering. SAD patterns and cross-fringes arising in HRTEM images from non-00*l* reciprocal lattice rows indicated that the stacking vectors of most adjacent 2:1 layers were not randomly oriented with respect to each other. Thus, the I/S was not fully turbostratic, but instead consisted of very thin, coherently stacked crystallites that extended across the fundamental particles postulated by Nadeau and coworkers.

S/(I + S) ratios were determined for about seventy HRTEM images obtained and interpreted by three different TEM operators. These ratios were consistent with those obtained from standard XRD procedures, suggesting that results obtained by XRD can be used to infer the initial structural state of mixed-layer I/S prior to treatment of samples for XRD experiments. The HRTEM experiments thus demonstrated that the two specimens examined consisted of ordered I/S existing as small crystals, most of which contained more layers than the fundamental particles of Nadeau and coworkers. The non-turbostratic stacking suggests an energetic interaction between the individual fundamental particles, leading to at least two alternative thermodynamic descriptions of these materials. Although the I/S crystals in the present experiments probably were disaggregated into fundamental particles during sample preparation for XRD, the I/S crystals appear to have separated only along the smectite interlayers. If the term “ fundamental particle ” is to be used for primary, untreated I/S, its original definition should be modified to include not only free particles, but also those that occur as layers within small crystals. It further should be recognized that these particles can interact thermodynamically and crystallographically with their neighbors.

Key Words: Electron diffraction • Fundamental particle • High-resolution transmission electron microscopy • Illite/smectite • Interstratification • Smectite

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