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COMMENT

ON A DEFINITION OF ILLITE/SMECTITE MIXED-LAYER

A recent contribution by Veblen *et al.* (1990) on the high-resolution transmission electron microscopy (HRTEM) of illite/smectite mixed-layers (I/S) has been extremely valuable in illuminating the structural state of I/S minerals. These investigators gave a definition of I/S that may deserve comment. A regularly interstratified illite/smectite crystallite was defined as a coherent stack of 2:1 layers having *illitic* and *smectitic interlayers.* This is probably the most appropriate definition of I/S mixed-layers and has far-reaching implications regarding their structure and formation. Obviously, the emphasis is on the nature of the interlayers, for both X-ray powder diffraction and HRTEM reveal the differences in interlayer configurations of these minerals.

Expandable (smectitic) interlayers are typically found in a stack (–S=S=S–) of smectite layers whereas nonexpandable (illitic) interlayers are observed in a coherent stack (.I:I:I:I.) of illite layers. The single layers of both illite and smectite are presently assumed to be "nonpolar" having a *C2/m* symmetry. The tetrahedral sheets within such a single layer of illite or smectite are therefore expected to be similar in charge and in composition. "Polar" 2:1 layers were suspected to be present in some I/S phases by Sudo *et al.* (1962), Weiss *et al.* (1970), Lagaly (1979), and Brown (1984), among others. Within a polar 2:1 silicate layer one of the tetrahedral sheets carries low or no charges as do the tetrahedral sheets in smectite, whereas the other tetrahedral sheet is highly charged as those in an illite layer. The polar character of such a layer can be simply presented by the notation (–P.) with dot surface being the illitic basal surface and dash surface representing smectitic basal surface. A stack of P-layers such as

–P:P = P:P = P:P = P:P–

creates a regular sequence (R1) of smectitic (=) and illitic (:) interlayers as observed in a perfectly R1-ordered rectorite by Ahn and Peacor (1986). P-layers were used by Ahn and Buseck (1990) to simulate HRTEM images to match the experimental images of R1-ordered I/S. Implicitly, the P-layers are often used in structural models of I/S for simulating their XRD patterns.

A random sequence of illite and smectite layers generates three different interlayers (=), (:), and (∸), as in the following example:

–S∸I:I∸S = S∸I:I∸S = S–

and one of them (S∸I) may be incompatible, therefore incoherent. A random sequence of smectitic (=) and illitic (:) interlayers without the incoherent S∸I junction can be simply generated by a stack of S and P layers:

–S = P:P = S = S = P:P = S–

R≥1-ordered I/S may therefore have incoherent I/S interfaces (∸), but R≥1-ordered I/P should have coherent I/P interfaces (: or =), as represented below:

–P:I:I:P = P:I:I:P = P:I:I:P–

Any sequence of I layers can be "coherently" coupled with a sequence of S-layers through P-layers:

.I:I:I:P = S = S = S–

HRTEM studies (Guthrie and Veblen 1989; Veblen *et al.,* 1990) have, in fact, demonstrated the coherency between the component layers in R≥1-ordered I/S.

A stack of I/P layers can be easily broken at P = P junctions rather than at I:I junctions and generate particles similar to "fundamental illite particles." In fact, a (–P:P–) pair of layers is functionally similar to the fundamental 20-Å illite particle described by Nadeau *et al.* (1984a, 1984b, 1984c). The –P:P– pair may account for the reactivity of external smectitic surfaces of fundamental illite particles. Such smectitic surfaces were suggested by Altaner *et al.* (1988) for fundamental illite particles. Thus, the P-layer may be the link unifying two concepts ("fundamental particle" and "interstratification of I and S layers") that have made great contributions to our understanding of the smectite-to-illite conversion.

The rectorite-illite series of regularly interstratified I/S can be properly represented as I/P mixed-layers. The morphological variations in this series may then be expected to range from laths of rectorite to pseudohexagonal platelets of mica. Such morphological features in R≥1 I/S series were described in natural samples by Inoue *et al.* (1987, 1988) and in synthetic products by Güven and Huang (1991). Ironically, the P-layer concept implies that some of the so-called I/S mixed-layers may not have illite or smectite as component layers, and perfectly R1-ordered rectorite may not contain either of them, but just a coherent stack of P-layers.

Now that the path has been paved by Veblen and his co-investigators, future studies on the structural state of I/S minerals may consider the P-layer concept. The hypothesis certainly deserves such a test.

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