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IMPLICATIONS OF TEM DATA FOR THE CONCEPT OF FUNDAMENTAL PARTICLES

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Abstract

There are two critical aspects of the concept of fundamental particles (fp): (1) in the original "working test" proposed, "separates" should demonstrate a single-crystal *hk*0 pattern in TEM (*i.e.*, coherent interlayers), and (2) if "separation" of particles occurs, it should occur only along smectite-like interfaces, which are incoherent. Unfortunately, "separates" are, with rare exceptions, referred to as fp without any test, whereas data imply that some separates of smectite-rich clays (especially) give *hk*0 SAED patterns reflecting a polycrystalline (turbostratically stacked) array. More importantly, coherency across even smectite interlayers is commonly demonstrated in original rocks by: (1) TEM lattice-fringe images with "cross-fringes", (2) euhedral crystals of I/S, smectite, and illite, and (3) ubiquitous SAED patterns with discrete, but non-periodic *hkl* reflections. Layer sequences of S, I/S, and I have both incoherent and coherent interfaces, the proportion of the former decreasing with increasing grade; *i.e.*, they are neither entirely translationally periodic nor turbostratically stacked. During separation, cleavage may occur along coherent interfaces in smectite (likely) as well as illite, so that even where the working test for fp is valid, such crystallites may be only portions of more extended coherent sequences as they occurred in original rocks. The term fp should therefore be used only where appropriate tests have been carried out. Even in those rare cases, such confirmation implies only a minimum measure of thicknesses (in separates) of coherent sequences in equivalent original rocks. Because separation affects the thickness of crystallites, layer sequences and coherency, separates reconstituted for XRD analysis may be inappropriate measures of relations in original rocks, and therefore may lead to erroneous conclusions regarding genetic (crystal growth) relations. Before such features in separates are used in any predictive fashion, it is therefore essential to determine the ways in which layer relations in original rocks are modified by disarticulation and subsequent reconstitution for powder XRD analysis. Only where tests confirm consistency with the original definition of fp, and where the relation of such units has been verified as directly reflecting units in original rocks as originally implied, should the term "fundamental particle" be used. These criteria have only rarely been met. The term "separate" confers, on the other hand, an unbiased, unadorned and accurate description of the state of samples prepared for XRD analysis. It should be used in most cases, at least until additional tests of sufficient numbers and kinds of samples have been obtained that confirm the occurrence of fp.

Keywords: fundamental particles, coherent interlayers, smectite, illite/smectite, illite.

Sommaire

Il y a deux aspects du concept de "particules fondamentales" qui sont critiques: (1) selon le test proposé originalement pour établir l'existence de ces particules, les "séparés" devraient démontrer un spectre *hk*0 en microscopie électronique par transmission (TEM) indicatif d'un cristal unique, c'est-à-dire, avec des interfeuillets cohérents, et (2) où il y a séparation de particules, cette séparation devrait se produire le long d'interfaces à caractère smectitique, qui seraient incohérentes. A quelques rares exceptions près, on considère ces "séparés" comme amas de particules fondamentales sans aucun test, tandis que selon certaines données au moins, les argiles riches en composante smectitique, particulièrement, font preuve d'un spectre de diffraction électronique *hk*0 indiquant un empilement polycristallin à caractère turbostratique. Fait plus important encore, il y a une cohérence dans la direction transversale aux feuillets de smectite, dans les roches originales, selon les observations de (1) des franges réticulaires transversales, (2) des cristaux idiomorphes de illite-smectite, smectite, et illite, et (3) le développement ubiquiste de spectres de diffraction électronique montrant des réflexions *hkl* définies, mais non-périodiques. Dans la séquence de feuillets d'une smectite, d'un agencement illite/smectite, et d'une illite, les interfaces peuvent être incohérentes ou cohérentes, et la proportion des interfaces incohérentes diminue à mesure qu'augmente le degré de recristallisation. C'est donc dire que ces interfaces ne sont ni complètement à translation périodique, ni à empilement turbostratique. Pendant la séparation, le clivage pourrait profiter d'interfaces cohérentes dans le cas d'une smectite (probablement), de même que dans une illite, de sorte que même si le critère nécessaire pour établir la présence de particules fondamentales est valide, de tels cristallites pourraient bien être des sections de séquences cohérentes plus étendues dans les roches originales. On devrait donc limiter l'utilisation du terme "particule fondamentale" aux seuls cas où les tests appropriés ont été faits. Même dans ces cas isolés, un résultat positif implique seulement une mesure de l'épaisseur minimum de séquences cohérentes des séparés représentatifs des roches équivalentes. Parce que la séparation affecte l'épaisseur des cristallites, la séquence des feuillets et leur degré de cohérence, les fractions séparées reconstituées pour une

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analyse par diffraction X pourraient s'avérer inappropriées comme témoins des relations dans les roches originelles, et ainsi pourraient mener à des conclusions erronées à propos de relations génétiques au cours de la croissance cristalline. Avant d'utiliser de telles caractéristiques dans les séparés à des fins de prédiction, il sera donc essentiel de déterminer les façons selon lesquelles les agencements des feuillets dans une roche originale ont été modifiés au cours de la désarticulation et de la reconstitution d'un échantillon pour analyse par diffraction X sur poudre. Seuls dans les cas de concordance avec la définition originale de particule fondamentale, et de vérification de la présence de tels agencements dans la roche originale, pourra-t-on utiliser le terme "particule fondamentale". Les cas où ces critères ont été satisfaits sont rares. En revanche, le terme "séparé" décrit sans préjugé et avec précision l'état des échantillons tels que préparés pour une analyse par diffraction X. On devrait s'en tenir à ce terme dans la plupart des cas, au moins jusqu'à l'application de tests additionnels à un nombre et une variété suffisants d'échantillons pour confirmer la présence de particules fondamentales.

(Traduit par la Rédaction)

Mots-clés: particules fondamentales, interfeuillet cohérent, smectite, illite/smectite, illite.

Introduction

Ever since the pioneering work of Nadeau and colleagues (Nadeau *et al.* 1984a, b, c, 1985, Nadeau 1985, Nadeau & Bain 1986), the notion of "fundamental particles" (hereafter referred to as fp) has occupied a central place in the field of clay mineralogy. On the other hand, the relations implied by fp are not universally accepted, as reviewed, in part, by Reynolds (1992). Some authors (*e.g.*, Dong & Peacor 1996) have implied that the concept is flawed or otherwise subject to revision; the problems are here identified as follows: (1) In recent communications with a number of clay mineralogists with respect to the meaning of the term "fp", the author has encountered confusion or ambiguity regarding the definition and, more importantly, the significance of the concept. Such lack of agreement over such a basic and important concept can lead to misunderstandings, rather than to common solutions of problems (see below, TEM data *versus* XRD data). (2) The significance of fp in relation to clay crystallites as they exist in original rocks, *i.e.*, before being subjected to separation in preparation for X-ray diffraction (XRD) analysis, is not well established. The term "fundamental" implies a relation which, unfortunately, is not clear to everyone. Since it is the ultimate goal of much of the field of clay mineralogy to use data from clays to provide information on the nature and genesis of clay minerals in rocks as they exist in nature and not in "separates", the relation between the state of clays in original rocks, separates, and separates as reconstituted for XRD analysis is a question critical and crucial to many aspects of clay mineralogy. (3) The data in support of the notion of fp come almost exclusively from studies of "separates", as prepared largely for XRD analysis, but also in part for atomic force microscopy (AFM) and low-resolution TEM studies. The data in disagreement with aspects of the fp notion are largely, but not entirely, derived from relatively high-resolution transmission electron microscopy (TEM) data of unseparated (usually ion-milled) samples of original rocks. Insofar as the two types of data have been inferred to be inconsistent, and indeed, to be incompatible, the author is concerned that this apparent inconsistency has led to an unfortunate lack of a balanced and integrated assessment of all kinds of data, and to perceived inconsistencies that have exacerbated disagreement. Because all kinds of data relating to the same materials must ultimately lead to at least complementary unified conclusions, such relations can result in problems in arriving at uniformly accepted conclusions regarding the state and genesis of clays in the geological environment.

It is the author's belief that both XRD and TEM data largely lead to compatible conclusions regarding the state of clays in both separated samples prepared for XRD (and other) analysis. On the other hand, the concept of fp, as useful as it has been when only XRD and related data were available for the separates, has been found to be at least partially incompatible with data obtained from newer TEM techniques. Such progress and adjustment in theory are marks of normal and healthy advances in science, and are expected where new observations of various kinds become available. This paper therefore represents an attempt to bring an overview to the notion of fp, to ensure that nomenclature is uniformly used with common understanding, and to integrate some recent results to clarify some ideas with respect to the relation between XRD and TEM data. It is the author's hope that even if the results of this discussion are judged to be problematic by some, they will at least lead to further discussion and clarification of ideas and a more unified approach.

Test for Fundamental Particles

Any discussion of fp must be based on common agreement as to what they are and how to identify them. A "working" test for a fp was defined by Nadeau *et al.* (1984b) and again by Nadeau (1985), and recently reaffirmed by Nadeau (1998). That test is based simply on selected-area electron diffraction (SAED) patterns as obtained for the individual particles of separates. A separated grain is defined as a fundamental particle if the two-dimensional *hk*0 SAED pattern is that of a single crystal, *i.e.*, if it has the form of a hexanet. Such a hexanet reciprocal lattice is obtained when any sequence of layers in a given particle, from as little as one to any number, consists entirely of layers that form a coherent three-dimensional array, *i.e.*, with coherent interfaces related by rotations of 60° or multiples thereof around the **c*** axis. Although the term "coherent" was not used in the original definitions, the crystallographic implication of a single-crystal *hk*0 pattern is that the layers be related by three-dimensional periodicity (rotations of multiples of 60°, which occur periodically to define a given polytype), or by stacking faults that are random variations in the sequence of 60° rotations. Such a coherent arrangement of layers is differentiated from interfaces that are random with respect to rotation around the **c*** axis, a relation that is usually referred to as "turbostratic" in the field of clay mineralogy. Where layers of a sequence are randomly related, the integrated *hk*0 diffraction pattern consists of two or more hexanets related by random rotation about **c***; an infinite number of such layers gives an *hk*0 pattern that consists of powder-sample-like rings, each ring corresponding to an individual reflection in a single-crystal pattern or to more than one such reflection having identical or similar *d*-values (Freed & Peacor 1992). Care must be taken with TEM measurements to establish that layers giving rise to multiple hexanets are indeed from the same original layer sequence, and not the result of overlap of separate grains spread on the substrate. Furthermore, coherency is not implied by an SAED pattern of discrete 001 reflections with d = 10 Å and submultiples thereof, as it only indicates that 001 layers are parallel, as consistent with both turbostratic stacking of parallel layers and three-dimensional coherency. Although the "working definition" given above requires SAED data, presumably any method used to verify coherency of interlayers in a separated grain would suffice.

It is the author's concern that the term fp is used increasingly to describe the separated grains that are analyzed by XRD, but without any test for their single-crystal nature. The only examples of such tests that have come to the author's attention are those of Nadeau and colleagues as noted above, Środoń *et al.* (1992), and Freed & Peacor (1992). There are also studies in which low-resolution TEM observations show that separated crystallites are euhedral (*e.g.*, Lanson & Champion 1991), in which case they may be assumed to be three-dimensionally and translationally periodic. Although it is certainly far from possible to so analyze the separates used in all studies of clays, the number of cited examples is markedly insufficient to verify that the term fp can be confidently used with respect to separated particles in general or with any specific study. The separates of Gulf Coast mudstones studied by Freed & Peacor (1992) gave multiple patterns, which were the rule. Such separates therefore did not contain fp as determined by the required test. It is now abundantly clear that the textures of smectite, illite, and I/S are very different in sandstones, bentonites and other rock types. Since particle separation is in part dependent on textural relations that are different in various rock types (see below), it is necessary to carry out tests for single-crystal *hk*0 patterns, not only on a large number of samples, but a large number of samples of each geological type before a general claim can be made that separates from a given rock type are fp. The severity of the separation procedures (physical, chemical; see below) is obviously also a determining factor, and constancy of separation techniques is necessary if tests for the presence of fp in one sample are to be even implied to be applicable to others.

The necessary variety of tests of fp character have yet to be made to demonstrate the general applicability of the fp concept. Some tests even reveal non-fp character. Those relations are part of the basis of the author's concerns for the possible misinterpretation of relations where the term fp is applied to separates without appropriate tests.

Relation of FP to the State of Clays in Original Rocks

The term fp has implications for the state of clays in the original rocks from which separates used in XRD or AFM analysis are derived, as implied by the word "fundamental", but a crisp definition of that relation apparently does not exist. What, therefore, is the implication of fp for clays in unseparated rocks? Nadeau (1985) specifically discussed the relations regarding coherent and incoherent interfaces with reference to fundamental particles. He noted the following: "The concept of interparticle diffraction of fundamental particles within aggregates, however, is consistent with the XRD data and rationalizes their turbostratic disorder... Precession XRD photographs of an undisturbed flake of rectorite... have also shown it to be turbostratic (Kodama 1966). This suggests that, prior to dispersion, these clays are composed of aggregates of fundamental particles... The majority of smectites are disordered turbostratic arrangements of layers (Brindley 1980), and therefore consistent with their being aggregates of fundamental particles." That is, the fp observed in separates are assumed to have been related by incoherent interfaces in original rocks. That is a critical distinction, as *it implies that the sites of coherent and incoherent interfaces in separates prepared for XRD are identical to those in original rocks.* The implications of the word "fundamental" for layer interfaces in I/S of rocks are thus crystal clear (although the specific sequencing of such fp must be different in separates and rocks, the average over large numbers of crystallites would be the same, as measureable by XRD of separates). A further implication is provided by the statement of Środoń *et al.* (1992) to the effect that fundamental particles contain no smectite layers, but are bounded by smectite interlayers; *i.e.*, assuming that no cleavage occurs across illite interlayers of original rocks during separation, the collection of fp has the same distribution as that of coherent packets of illite in the original rocks.

A key aspect of the above relations is that it is "assumed" that the interfaces between fp in original rocks are incoherent. The author is aware of only one test of that relation using XRD (see below), by Reynolds (1992). A test of that relation is admittedly difficult, could not be made even a decade ago, but nevertheless can be made now. It requires that the layer sequences in a variety of original rocks be clearly identified with respect to their degree of coherency or incoherency, and that the separates obtained therefrom be similarly characterized to determine if they are identical. A serious difficulty is in the uncertainty-like principle in such relations, in that specific sequences of layers cannot be correlated because once identified in original rocks, they must be affected by specimen preparation for separates. That requires that large-enough numbers of layer relations be defined in order to obtain a statistically relevant sample. Results should be obtained on a variety of rock types, including bentonites, shales, and rocks of hydrothermal origin, and of a variety of grades of each. But such studies are rare, that by Šucha *et al.* (1996) being a notable exception. The assumption regarding the relation between interfaces in separates and equivalent rocks is therefore almost entirely just that at present, an assumption. Without the appropriate test(s), it cannot be regarded as definitive, especially as there is an increasing volume of contrary evidence (see below), and the conclusion that it is definitive must therefore be based on a leap of faith.

Observations of Coherent Layer Sequences in I/S of Rocks

Cross fringes

Only in recent years, and largely subsequent to the original definitions of fp relations, have TEMs become readily available and applied to studies of layer sequences in original rocks. Such studies are usually concerned with observations of ion-milled samples, which retain original rock textures. One of the first studies concerned with interlayer coherency was that of Ahn & Buseck (1990), who obtained high-resolution transmission electron images (HRTEM) of Zempleni I/S, showing that at least in the samples studied, illite- and smectite-like layers are coherently related. The HRTEM images were of much higher resolution than those of subsequent studies, which utilized 001 reflections to obtain lattice-fringe images in combination with "cross fringes" obtained from a single set of non-001 reflections. The studies of Guthrie & Veblen (1989a, b, 1990) and Veblen *et al.* (1990) were critical in defining appropriate experimental factors and in providing a firm structural basis for interpretation of images. Veblen *et al.* obtained images with cross fringes, *i.e.*, lattice fringes from non-001 reflections that cut across 001 fringes, using I/S of hydrothermal origin from two localities. Where cross fringes cut across two or more contiguous 001 interlayers, the alternate layers must be related by multiples of 60° of rotation around **c***; *i.e.*, they must be related by coherent interfaces. Banfield *et al.* (1991) reported that such coherency was observed even in smectite forming in a saline lake environment. Dong & Peacor (1996) subsequently carried out an extensive study of smectite, I/S and illite from a sequence of Gulf Coast mudstones, and observed coherent sequences averaging 3–4, 6–7, and 9–10 layers for smectite-, I/S- and illite-rich samples, respectively, and the interested reader is referred to that paper for images showing extended coherent sequences rich in smectite interlayers. They pointed out that it is difficult to image such coherent sequences through cross fringes, and therefore that the numbers of layers in such coherent sequences observed are minimum values, *i.e.*, the actual numbers of layers consecutively related by coherency must be larger than those observed. Their observations were based in part on a new technique for impregnation of samples that prevents collapse of smectite-like interlayers and allows identification of individual S, I/S and I layers in lattice-fringe images (Kim *et al.* 1995). Observations of significant coherency were also made by Huggett (1995) for I/S in North Sea mudstones.

Nadeau has noted (pers. commun.) that the particle thicknesses observed for elementary particles of illite by Nadeau *et al.* (1985) for bentonite and sandstone samples are not much different than the thicknesses over which cross-fringes were observed by Dong & Peacor (1996). The thicknesses observed by Nadeau *et al.* were directly correlated by those authors with the proportion of illite, which implies that the particles were illite particles without smectite interlayers, *i.e.*, fp. Nadeau *et al.* directly argued against cleavage along illite interlayers during sample preparation, further implying a direct relation between particles consisting only of illite layers and with their states in original rocks. The correlation between the thicknesses observed by Nadeau *et al.* (1985) and Dong & Peacor (1996) is only approximate and not meaningful for two reasons: (1) The thicknesses of coherent sequences observed by Dong & Peacor are smaller than the true values, owing to the difficulty in making such observations, and (2) the emphasis of Dong & Peacor was on cross fringes that included samples of smectite or I/S (largely R1 I/S), *i.e.*, smectite interfaces are a major component of sequences of coherently related layers in original rock samples. Insofar as they relate 2:1 layers, such coherently related smectite interfaces must be considered as components of crystallites in the original rocks. Conclusions about crystal sizes and therefore about growth mechanisms would be based on faulty data if only the coherently related illite interfaces were considered.

Nadeau (pers. commun.) has also pointed out that TEM images of ion-milled samples may be an artifact of sample treatment, which would affect all samples at ambient P and T because rocks at depth are subject to high fluid pressures, and therefore subject to change when brought to the surface and treated in the laboratory. Indeed, fine-grained pelitic rocks studied by TEM display no or almost no pore space, whereas in-well measurements indicate several % porosity (Nolen-Hoeksema, pers. commun.), and laboratory measures of porosity utilizing infiltration of fluids at equivalent pressures are compatible with values determined *in situ*. Expansion of smectite interlayers relative to illite interlayers most certainly must occur, and the effects of drying the sample under ambient P,T conditions may result in reconfiguration of smectite-like interfaces. Such an effect would, if it occurs, equally affect samples studied by all methods, however, and cannot be tested given our present technology. In any event, rotation of entire, extended layers during loss of fluid pressure with resulting change in relative positions must at the least be minimal, given the extent of such layers [*e.g.*, Ahn & Peacor (1986) with respect to "megacrystals of smectite"], and the restriction imposed by constraining textures. Nevertheless, such assumptions remain untested.

SAED patterns

Very few attempts have been made to obtain cross fringes by TEM, and the argument may be made that even though every attempt has led to verification of at least limited coherency, the number of studies is far too small to permit generalizations to be made. However, more general tests for coherency exist. In particular, Grubb *et al.* (1991) noted that SAED patterns of smectite, I/S, and illite of diagenetic origin that occur in shales from a wide range of localities are invariably similar. That is, they display non-00*l* reflections, all of which are non-periodic, ill-defined, diffuse and weak in intensity. The interested reader may refer to Grubb *et al.* (1991) or Dong & Peacor (1996) for examples. Even though the non-00*l* reflections do not display the continuously diffuse character required of ideal $1M_d$ illite, the SAED patterns display considerable disorder to a greater extent than any other samples, and thus the term $1M_d$ was used for them. Dong & Peacor (1996) subsequently showed that smectite, I/S and illite from the Texas Gulf Coast and other localities gave SAED patterns of the same type. The lack of complete disorder (*i.e.*, the non-existence of ideal $1M_d$ stacking) was further demonstrated by the observation of cross-fringes obtained by including at least one of the non-periodic, diffuse, non-00*l* reflections in the objective aperture of the TEM. That is, the cross fringes are the Fourier transform of the ill-defined, non-periodic reflections in such SAED patterns. Both the cross fringes and ill-defined non-00*l* reflections are measures of the same local interlayer coherency.

The occurrence of the (largely) non-periodic SAED patterns that were originally described by Grubb *et al.* is therefore direct evidence for the presence of a significant degree of coherency in the stacks of layers that give rise to them. It is an easy task to obtain such SAED patterns, but difficult to obtain images with cross fringes from them. Although the number of observations of cross fringes is limited to those described above, thousands of SAED patterns of smectite, I/S and illite obtained at the University of Michigan have almost without exception exhibited the same characteristics as those which lead to cross fringes. A very limited list includes diagenetic through low-grade metamorphic sequences from New Zealand, the Gaspé Peninsula, and the Welsh sedimentary basin, but the same has been observed for a long list of other localities. It is important to add, however, that our sample of localities is heavily weighted to slates, shales and mudstones from a wide range of grade, and to a number of bentonites, but with limited hydrothermal examples (*e.g.*, sediments in the area of the Salton Sea) and clastic sediments coarser than siltstones. It is nevertheless clear that, especially for shales and mudstones, not only illite but (to a lesser degree) stacks of I/S and smectite ubiquitously have a significant degree of coherency, and this is a near-universal relation.

Euhedral crystals

One of the touchstones of crystallography is Steno's "law of interfacial angles", from which the notion of three-dimensional periodicity is derived. That is, the presence of euhedral crystals of I/S or illite provides unambiguous evidence for translation periodicity, or stacking faults related by rotations of multiples of 60° about **c***, *i.e.*, coherency. Where such crystals are observed, the coherency is inevitably validated by the presence of single-crystal *hk*0 diffraction patterns, often with the usual imperfect reflections that are due to strain, but as perfectly consistent with periodicity as interrupted only locally by defects of kinds typical of all crystalline materials; that is, the emphasis is on periodicity with local exceptions, rather than on turbostratic stacking with local exceptions due to coherency across so-called illite interlayers. The former corresponds simply to an ideally imperfect crystal. Because coherent layer relations are perfectly consistent with the fp hypothesis, it is important to verify that the euhedral crystals being studied are really I/S rather than pure illite without smectite interlayers. In other words, smectite-free illite may occur as euhedral crystals, for which the fp hypothesis would also predict complete coherency. It is only the presence of smectite layers that leads to the prediction of incoherency.

The author is aware of only two examples of TEM observations of separated, euhedral crystals of smectite or I/S for which equivalent studies of ion-milled samples (which verify layer sequences) have been made. One involved Texas Gulf Coast mudstones, for which Freed & Peacor (1992) studied separates, and in which Dong & Peacor (1996) observed cross fringes. The latter observations confirmed the identification of euhedral crystals of separates as being dominantly R1 I/S. In addition, Hover *et al.* (1996) have shown that trioctahedral smectite crystallizes directly from saline fluids in sediments from near the water – sediment interface, giving rise to euhedral crystals of smectite as observed in separates. Identification was confirmed using ion-milled samples, although no attempt was made to observe cross-fringes. On the basis of studies of separates only, however, there are many observations of euhedral crystals where evidence is very strong that the layer sequences have high proportions of smectite interlayers (*e.g.*, Lanson & Champion 1991). These data provide an additional set of examples of coherency in I/S layer sequences which, as they involve euhedral crystals, are especially appropriate to coarse clastic sediments and materials subjected to hydrothermal fluids.

The state of coherency in stacks of layers in rocks

The fp concept *assumes* that particles separated for XRD analysis were either separate units (stacks) in original rocks, or they constituted MacEwan crystallites (thick stacks) with incoherent and coherent interfaces. Cleavage is assumed to occur on the incoherent interfaces, which are the smectite interlayers, thus separating the stack into fp, within each of which interfaces are coherent and illite-like. But such an assumption must be tested and shown to be true if the fp hypothesis is at all relevant. Although that was not possible at the time the hypothesis was introduced, it is possible now. The only extensive TEM data which purport to do this, however, are those of Dong & Peacor (1996), for which cross-fringes were observed. They emphasized the point that the observed coherent sequences had minimum numbers of layers, as observation of cross fringes is difficult. Where observed, they are meaningful, but where not observed, no conclusion can be reached regarding coherency or non-coherency. Nevertheless, the SAED data of Dong & Peacor (1996) and Freed & Peacor (1992) clearly show that for the Texas Gulf Coast samples studied, layer sequences of smectite, I/S and illite are neither entirely coherently nor incoherently related; that is, stacks consist of a complex assemblage of coherently and incoherently related interfaces. Where the sequences are coherent, the most common stacking sequence is that of the $2M_1$ polytype, although polytypic sequences are complex, being unique to any single grain.

Dong & Peacor (1996) noted that the numbers of adjacent coherently related layers increase with grade, from smectite to R1 I/S to illite in the passive burial sedimentary environment of Texas Gulf Coast sediments. Observations of many diagenetic to low-grade metamorphic sequences have shown that the density of defects decreases with increasing grade (Peacor 1992), presumably because high densities of defects are metastable with respect to states of stable equilibrium. Thus, illite (muscovite) that forms under epizonal conditions occurs in stacks of layers consisting of hundreds of layers within which layer terminations are rare, and layers are vitually entirely coherently related in $2M_1$ sequences.

Reynolds (1992) directly addressed the assumption, made in the fp concept, that interfaces between fp are only incoherent along smectite layers. He obtained XRD patterns of powdered and original rock samples. XRD patterns from corresponding rocks and powders were effectively identical. The critical observation, however, was that those reflections implying relative relations among layers were consistent with turbostratic stacking in both separates and original rocks, *i.e.*, the interfaces between separate crystallites and those of original rocks are turbostratic. This result has been taken by some as a general proof that smectite interlayers are always or almost always incoherent (turbostratic) in all occurrences of all rock types. Reynolds discussed the problems of size of coherent scattering domains for XRD *versus* TEM, but was unable to satisfactorily rationalize his XRD data with those of TEM. Reynolds's samples were all K-saturated bentonites with the exception of one Zempleni sample. Observations have not been made by TEM on the same specimens of K-saturated bentonite, and discrepancies between XRD and TEM results cannot therefore be directly compared for them. Indeed, the observations of Dong and Peacor (1996) were primarily for shales. Reynolds's observation of lack of coherency between illite packets may therefore be both correct and not in any way inconsistent with observations by TEM on shales, mudstones and the like. For such data to be definitive, the XRD observations must be coupled with TEM observations of both original rocks and separates. Otherwise, the comparisons between different rock types may be more confusing than clarifying. Reynolds's XRD data for the Zempleni sample appear to be incompatible with TEM data, however, as Ahn & Buseck (1990) showed at least partial coherency across smectite interlayers for Zempleni material, although not for the very same sample. The apparent contradiction can only be absolutely determined where the same sample is studied by all methods.

Sucha *et al.* (1996) provided a second critical set of observations relating layer sequences in separates and in original rocks, including those used for XRD studies. Using I/S of hydrothermal origin, they showed through TEM observations that layer sequences and thicknesses of crystallites in separates and in original rocks are approximately equivalent. Although they did not show single-crystal SAED patterns of separates, the euhedral shapes of separated grains implied that they consisted of translationally periodic sequences. Lattice-fringe images showed packets with exceptionally well-defined boundaries along which separation would naturally occur. Although we also find that such sharp crystal boundaries are typical of hydrothermal clays, they are in sharp contrast with images of packets in bentonites and most shales, where packet boundaries are defined only with difficulty. Nevertheless, those results are strikingly in support of the idea that thicknesses of crystals in separates and packets of unseparated samples are equivalent. They would appear to be in sharp disagreement with the fp concept, however, as the data imply that the separated particles must contain smectite interlayers. In other words, the separates did not consist only of illite interlayers, with separation having occurred along all smectite interlayers.

Separation of layers in stacks during sample preparation

Another key question must be resolved: what are the specific kinds of interlayers across which separation occurs in samples treated by mechanical or chemical means during preparation for XRD? That relation is at the heart of speculation about the relation between separates and original rocks, XRD data and TEM data for ion-milled samples, and fp *versus* "separates". Many observations imply that samples behave differently depending on the proportion of smectite interlayers. It is important to emphasize that relation here, as the author believes that at least some of the failure to resolve differences is caused by seeming inconsistencies between data for two very different rock types: (1) sedimentary diagenetic sequences in which smectite interlayers play a major role, with formation of authigenic clays in a passive burial-metamorphic environment characterized by load stress. Increasing grade is commonly measured by the proportion of smectite layers relative to illite layers; (2) low-grade metamorphic sequences, which generally range from the upper diagenetic zone, through anchizone to epizone, with smectite interlayers occurring only in the lowest grades and in minor proportions. Stress is primarily tectonically induced. Illite crystallinity is the parameter most commonly used to characterize grade in rocks which, with increase in grade, become dominated by cleavage-parallel mica.

Most would agree that smectite interlayers are sites of "separation", the term cleavage being restricted to separation across coherent interlayers if used in its usual mineralogical context, and as shown for rectorite by Ahn & Peacor (1986). Such separation is basic to the notion of fp, Środoń *et al.* (1992) noting that "...there are no smectite interlayers in a fundamental particle, but that its planar surfaces are smectitic...". Preferential separation across smectite interlayers was critical to the observations of Nadeau and coworkers, who demonstrated separation of smectite into 10-Å "elementary smectite particles", and of R1 I/S into 20-Å "elementary illite particles", the high-charge, K-rich interlayer being retained without separation. Thicker particles with numbers of layers as multiples of 10 Å, and giving rise to hexanets in SAED patterns, were interpreted as having only high-charge interfaces, and it was natural to assume that they were coherent illite units that separated *via* the smectite-like bounding interfaces of original stacks. Assuming that smectite always occurs as turbostratically related layer sequences, these underpinnings of the fp concept were internally consistent and represented a major advance.

TEM observation of dioctahedral clays and micas of relatively high-grade rocks involves layer sequences that contain no smectite interlayers, however. Where grade is sufficiently high, single crystals may be separated and studied by single-crystal XRD methods including crystal-structure analysis, and such observations invariably show that the large crystals form coherent three-dimensionally periodic arrays characterized by various kinds of defects that cause crystals to diffract as ideally imperfect arrays. It is well known that grinding such micas to produce samples for powder XRD results in separation into units whose thickness is a multiple of 10 Å, such that severe grinding results in line broadening and eventually to a loss of all XRD peaks. TEM observations verify that metamorphic clays and micas are relatively defect-free, with proportions of defects generally decreasing with grade (*e.g.*, Peacor 1992). That is, if separation is to occur, it must occur across coherent, high-charge interfaces, albeit there is some evidence that at least some cleavage must be across interlayers that contain local defects (Jiang *et al.* 1997, Li *et al.* 1998, Warr & Nieto 1998). The separate, cleaved units may therefore pass the fp test in that they give single-crystal SAED patterns, but they are not "fundamental" and not separated by smectite-like interlayers in original rocks.

Some very recent research has concerned measurements of crystal and grain thicknesses by TEM of both the separates used for XRD and the original rocks, both as represented by ion-milled samples. Jiang *et al.* (1997) studied shale and slate samples from a range of grade from upper diagenesis through epizone, and concluded that decrease in the size of coherent layer-sequences could be extreme, with diminution of thickness by factors as large as six for epizonal samples. As crystallite thickness decreases with decrease in grade to that typical of diagenesis, the mean thickness of separates and of original layer-sequences tends to approach equality, however. This finding implies that in clays of the upper diagenetic zone, separation occurs largely on crystal interfaces, and not within coherent sequences, but TEM observations of rocks of higher grade clearly display cleavage within coherent sequences. This result is sensible also from the point of view of rock textures, in that as grade increases to that of the epizone, textures change from those of loose assemblages of separate grains to those with interlocking textures typical of metamorphic rocks. Other studies comparing crystal thicknesses in separates and equivalent original rocks, over a range of grade from diagenesis through the epizone, which have shown similar relations of separate size *versus* grade, include those of: (1) Warr & Nieto (1998), who have correlated size data for four samples that were distributed internationally as XRD standards for measurements of illite crystallinity, (2) Li *et al.* (1998), who studied a sequence from the South Island of New Zealand ranging in grade up to biotite schists, and (3) Wang (pers. commun.), who obtained data for a number of samples from Hungary, the Franciscan Formation of California and the Welsh sedimentary basin, which were originally studied by TEM in ion-milled samples by Merriman and Roberts (pers. commun.). In some cases, XRD peak analysis has been used to calculate mean thicknesses for samples for which thicknesses in original rocks were observed by TEM (*e.g.*, Nieto & Sánchez-Navas 1994, Árkai *et al.* 1996, Jiang *et al.* 1997). In the latter two cases, significant decreases in thickness on grinding were documented.

Most of the studies described above involved samples that did not have significant proportions of smectite interlayers. At least those studied in our laboratory were prepared by grinding and ultrasonic treatment simply because they formed at grades at least as severe as that typical of late diagenesis, and were indurated. No chemical treatments were used. Although those studies do demonstrate cleavage across coherent illite interlayers, the gentler treatments possible in achieving separation in lower-grade samples may be much less severe. Indeed, the data of Li *et al.* (1998) and Jiang *et al.* (1997) imply near-equality for separate thicknesses in XRD samples and original rocks. The new method of PVP-intercalation described by Eberl *et al.* (1998) seems to have the potential of minimizing cleavage within illite interlayers in I/S. The possibility of cleavage across coherent illite interlayers in smectite-rich materials has not been thoroughly and directly tested, however.

The data from a number of studies thus show that separation occurs in very different ways for diagenetic samples and those of higher grades, as clearly demonstrated primarily by Nadeau and coworkers for the former and TEM data for original rocks in the latter case. The key element in the case of low-grade samples, however, is that it has been *assumed* that: (1) separation occurs across incoherent interfaces, and (2) smectite interfaces are turbostratically related, *i.e.*, incoherently related. Despite the growing number of studies of higher-grade samples, there has been no study by TEM of the thickness of coherent layer-sequences in original rocks as correlated with such measurements for samples separated for XRD for smectite-rich samples, with the limited but significant exception of Šucha *et al.* (1996), so the critical assumption remains effectively untested.

On the other hand, there is a growing body of evidence, much of which was reviewed above, which demonstrates that basic assumptions of the fp hypothesis are flawed with respect to the relation between separated particles and those in original rocks. The two most significant are: (1) evidence described above, especially for R1 I/S, but also even for smectite, shows that significant coherency exists in most, if not virtually all, natural samples (as does turbostratic stacking!). The evidence from TEM structure images, cross fringes, euhedral crystals, and SAED patterns has become so voluminous as to now be overwhelming. (2) The evidence demonstrating cleavage across coherent units in higher-grade crystals implies that cleavage may indeed occur in samples from all grades. That must depend critically on the severity of treatment and the degree to which it is both chemical and physical, however. The effect of treatments is extremely important, and it has yet to be studied using the array of methods now available.

Agreement between XRD and TEM Observations

There has been much discussion, mostly informal, about the apparent incompatibility of XRD and TEM data. Indeed, some of the discussion above has seemingly highlighted such incompatibility. However, the author is not aware of any significant incompatibilities for samples from upper diagenetic to epizonal environments. Significant apparent inconsistencies seem to exist only for diagenetic samples with significant smectite components.

XRD and TEM data for diagenetic-grade material are in substantial agreement in one respect for samples of all grades, however: the proportions of smectite and illite in I/S. Veblen *et al.* (1990) have emphasized that their semiquantitative measures of the proportions of illite- and smectite-like layers in TEM images are in agreement with the values obtained through XRD data. With limited exceptions, all of the research of the author's group has emphasized the approximate agreement between proportions of illite- and smectite-like layers as first determined from XRD patterns. Indeed, TEM data subsequently obtained on the same samples are accepted as being representative only where such agreement is obtained. One exception relates to smectite-rich R0 I/S in Texas Gulf Coast samples, for which no illite layers were detected by TEM (Dong & Peacor 1996) in samples for which XRD data indicate the presence of *ca.* 25% illite layers. Even that apparent incompatibility has a plausible explanation in heterogeneity of interlayer charges, although it remains to be thoroughly tested. More importantly, Środoń and colleagues (*e.g.*, Środoń *et al.* 1990) have carried out comparisons of proportions of illite- and smectite-like layers as observed in TEM lattice-fringe images and found the results to be in excellent agreement. That is, there does not seem to be any significant disagreement about the relative proportions of illite- and smectite-like layers.

What, then, are the significant incompatibilities? They have only to do with the *sequences* of layers and the approach to *coherency* of crystallite interfaces, not the relative proportions of layers of different types. The specific differences are related to four factors: (1) layers may be coherently related across smectite interlayers in original rocks, (2) illite layers may be incoherently related across illite interlayers, (3) cleavage may occur across coherent illite interlayers, and (4) cleavage need not occur across smectite interlayers. Recognition that the separation process is not so simple or constrained as assumed by the fp hypothesis (*e.g.*, separation does not occur only and always across incoherent smectite interlayers) provides a means of rationalizing the apparent differences in results obtained by XRD and TEM. XRD data are obtained only on samples that have been modified during the separation process, with modification involving only the sequencing of units relative to the interlayer relations, not their proportions. Particle-size distributions in XRD samples are then clearly seen to be a function of three variables: (1) the state of crystallites in the rocks, (2) the chemical and physical process of separation, and (3) the method of aggregation of particles into a sample to be subjected to XRD. XRD data are thus interpreted to be a *function* of the original state of clay minerals in a rock, but are not a direct measure of it. Determination of the way in which separation of rocks and aggregation into XRD samples affect the state of crystallite distributions in rocks is thus seen to be critical in utilizing data on particle-size distribution to interpret crystal-growth relations in original rocks, a primary goal of clay mineralogy.

It is therefore the author's contention that TEM and XRD data are clearly in qualitative agreement, and that where that is not the case, as with smectite-rich R0 I/S in Texas Gulf Coast mudstones (Dong *et al.* 1997), the differences are not so much due to the error in one or the other, but to a lack of understanding of how both kinds of equally significant data can be rationalized with respect to new concepts.

Nomenclature of Elementary Particles

Nadeau *et al.* (1984b) discussed the innovative idea of interparticle diffraction with respect to measures of thicknesses of particles measured by TEM and relations for fp. They noted that separation of samples rich in smectite gave particles 10 Å in thickness, whereas those consisting of R1 I/S gave particles 20 Å in thickness; they referred to such particles as "elementary" smectite and illite particles, respectively. The 20-Å-thick illite particles obtained from K-saturated rectorite were inferred to be centered on a K-rich interlayer.

The author believes that the use of the term "elementary illite layer" is confusing and has led to some needless discussion in the light of more recent data. There are now abundant data that imply or directly show that the distribution of Si and Al in R1 I/S is not the same as that in clays consisting of sequences of all smectite or all illite interlayers. Dong *et al.* (1997) and Altaner & Ylagen (1997) have reviewed the evidence supporting the notion that in smectite and illite, the Al–Si distribution is symmetrical across the sheet of octahedra. This leads to approximately equal values of net negative charges (nnc) across all interlayers. By contrast, in R1 I/S (rectorite) the Al–Si distribution is almost certainly symmetrical across interlayers, leading to high- and low-nnc values across alternate interlayers. This distribution of charge provides a potential for ordering of smectite-like and illite-like interlayers.

The 20-Å particles obtained by cleavage along low-charge interlayers of rectorite are therefore not "illite" particles. They are illite-like insofar as the K-rich interlayer is concerned. They are very different from illite in Al–Si distribution, however. They also must differ in the inevitable adjustments of structure topology that result from variations in cation-ordering patterns. Such adjustments have been amply demonstrated through structure refinements of compositional variants of a given silicate structure-type, such as that of micas. The use of the unmodified term *illite* for such particles is therefore confusing and serves to mask their unique nature. It incorrectly implies, for example, that R1 I/S can be formed simply by stacking up illite and smectite layers, and obscures the uniqueness of rectorite insofar as it consists of three-dimensionally coherent layers "like" those comprising illite and smectite. Although the term "rectorite-like" would be most apt for 20-Å particles obtained by separation of R1 I/S layer sequences, the term "illite-like" would at least be a distinct improvement over "illite".

Recommendations

As emphasized also by Li *et al.* (1998), separation and reconstitution in XRD preparations or other kinds of samples may not only alter the layer interfaces and mixed-layer sequences, but also diminish the effective sizes of crystallites. With respect to crystal size, Li *et al.* recommend that in order to define the relations between samples prepared for XRD and those of original rocks, it is necessary to characterize crystal size in the original rocks by TEM and, for the same samples, in the separates as reconstituted for XRD analysis. Because the separation effects are dependent on the severity of the process, standardization of the process with respect to both physical and chemical treatment is essential. But the process of disarticulation is also dependent on texture, which varies both according to rock type (*e.g.*, shales *versus* bentonites), and to grade, diagenetic or metamorphic. It is essential that such relations be defined; otherwise conclusions from one rock type may appear to be at variance with those of others, leading to apparent disagreement. That is especially apparent for coarse-grained clastic rocks and hydrothermally affected rocks, for which TEM generally shows discrete euhedral crystals, whereas in bentonites, shales and mudstones, packet boundaries may be vague.

Those same kinds of recommendations are equally valid for a determination of layer sequences and interlayer coherency. Furthermore, they would provide a rigorous evaluation of the possibility that differences between XRD and TEM data regarding coherency are based on differences in the sensitivities (*e.g.*, Veblen *et al.* 1990) or averaging capacities (Reynolds 1992) of the two methods. Careful and complete characterization by TEM of both the separates and original rocks studied by Reynolds using XRD would definitively speak to that question, for example.

Such comprehensive studies must be based on the methods involving laborious measurements over large numbers of individual crystallites and layer interfaces. It is a daunting task. But once determined, the effects of sample disarticulation would be known, and measurements could subsequently be made by the infinitely more efficient method of XRD, with reasonable confidence of the relation between separates and those of undisarticulated rocks. As emphasized above, where separated clay grains have commonly been called fp, no such test as recommended by Nadeau and coworkers has generally been used. The relation to layer sequences in original rocks therefore requires a leap of faith. It may well turn out that the kinds of tests described above actually will demonstrate that separates now described as fp are exactly that, replete with implications for layer sequences in original rocks. Especially with respect to the evidence reviewed here that is to the contrary, the author pleads only that where separates have not been adequately tested to be fp, and where separates are claimed to be representative of relations in the original rock, that they be described simply as "separates". Labeling them as fp without adequate tests, and with significant implications for geological relations, is inappropriate. Suggestions that the definition of fp be changed so that separates are generally accepted as fp, without tests and clear implications for the states of clay minerals in original rocks, are also inappropriate. A changing definition would lead to even more confusion regarding the meaning and significance of the term fp. Indeed, one of the purposes of this paper was to provide a review of the definition of the term fp, so that discussion could proceed *via* common understanding.

Summary

TEM and other data clearly demonstrate that structural coherency, as reflected in coherent diffraction relations, is common in dioctahedral smectite, I/S, and illite, but with samples from lowest grades having larger proportions of disordered, incoherent interfaces. That finding is consistent with the general trend observed by TEM over samples from a wide range of grades, that disorder in general, and the proportion of incoherent interfaces in particular, decrease with increasing grade of diagenesis or metamophism. Smectite-rich crystals in low-grade rocks are generally neither entirely turbostratic nor completely ordered, but have proportions of both coherent and incoherent interfaces. Likewise, packets of illite occurring at low grades and without smectite interlayers have significant proportions of incoherent interfaces. Although the degree of disorder is certainly greatest in smectite, the fundamental tenet of the fp hypothesis, that such interfaces are incoherent and the only sources of separation, seems to be in error based on currently available observations. Cleavage, preferentially across incoherently related smectite interlayers relative to illite interlayers, but also across both illite interlayers and coherent smectite interlayers, with subsequent reconstitution into samples for XRD, may result in production of both altered layer-sequences and altered proportions of coherent interfaces. Especially where the effects of separation are taken into account, the observations by TEM (primarily) on unseparated samples and those by XRD (primarily) on separated samples, give rise to conclusions that are almost entirely compatible.

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References

AHN, JUNG-HO & BUSECK, P.R. (1990): Layer-stacking sequences and structural disorder in mixed-layer illite/smectite: image simulations and HRTEM imaging. *Am. Mineral.* **75**, 267-275.

________ & PEACOR, D.R. (1986): Transmission electron microscope data for rectorite: implications for the origin and structure of "fundamental particles". *Clays Clay Minerals* **34**, 180-186.

ALTANER, S.P. & YLAGEN, R.F. (1997): Comparison of structural models of mixed-layer illite/smectite and reaction mechanisms of smectite illitization. *Clays Clay Minerals* **45**, 517-533.

ÁRKAI, P., MERRIMAN, R.J., ROBERTS, B., PEACOR, D.R. & TOTH, M.N. (1996): Crystallinity, crystallite size and lattice strain of illite-muscovite and chlorite: comparison of XRD and HRTEM data for diagenetic to epizonal pelites. *Eur. J. Mineral.* **8**, 1119-1137.

BANFIELD, J.F., JONES, B.F. & VEBLEN, D.R. (1991): An AEM–TEM study of weathering and diagenesis, Albert Lake, Oregon. II. Diagenetic modification of the sedimentary assemblage. *Geochim. Cosmochim. Acta* **55**, 2795-2810.

BRINDLEY, G.W. (1980): Order – disorder in clay mineral structures. *In* Crystal Structures of Clay Minerals and their X-Ray Identification (G.W. Brindley & G. Brown, eds.). The Mineralogical Society, London, U.K. (125-195).

DONG, HAILIANG & PEACOR, D.R. (1996): TEM observations of coherent stacking relations in smectite, I/S and illite of shales: evidence for MacEwan crystallites and dominance of $2M_1$ polytypism. *Clays Clay Minerals* **44**, 257-275.

________, ________ & FREED, R.L. (1997): Phase relations among smectite, R1 illite-smectite, and illite. *Am. Mineral.* **82**, 379-391.

EBERL, D.D., NÜESCH, R., SUCHA, V. & TSIPURSKY, S. (1998): Measurement of fundamental illite particle thicknesses by X-ray diffraction using PVP-10 intercalation. *Clays Clay Minerals* **46**, 89-97.

FREED, R.L. & PEACOR, D.R. (1992): Diagenesis and the formation of authigenic illite-rich I/S crystals in Gulf Coast shales: TEM study of clay separates. *J. Sed. Petrol.* **62**, 220-234.

GRUBB, S.M.B., PEACOR, D.R. & JIANG, WEI-TEH (1991): Transmission electron microscope observations of illite polytypism. *Clays Clay Minerals* **39**, 540-550.

GUTHRIE, G.D., JR. & VEBLEN, D.R. (1989a): High-resolution transmission electron microscopy applied to clay minerals. *In* Spectroscopic Characterization of Minerals and their Surfaces (L.M. Coyne, S.W.S. McKeever & D.F. Blake, eds.). *Am. Chem. Soc., Symp. Ser.* **415**, 75-93.

________ & ________ (1989b): High-resolution transmission electron microscopy of mixed-layer illite/smectite: computer simulations. *Clays Clay Minerals* **37**, 1-11.

________ & ________ (1990): Interpreting one-dimensional high-resolution transmission electron micrographs of sheet silicates by computer simulation. *Am. Mineral.* **75**, 276-288.

HOVER, V.C., WALTER, L.M., PEACOR, D.R. & MARTINI, A.M. (1996): Early marine authigenesis of K- and Mg-rich smectite in a hypersaline tidal flat, Salina Ometepec, Mexico. *11th Int. Clay Conf. (Ottawa), Program Abstr.*, A37.

HUGGETT, J.M. (1995): Formation of authigenic illite in Palaeocene mudrocks from the central North Sea: a study by high resolution electron microscopy. *Clays Clay Minerals* **43**, 682-692.

JIANG, WEI-TEH, PEACOR, D.R., ÁRKAI, P., TÓTH, M. & KIM, JIN-WOOK (1997): TEM and XRD determination of crystallite size and lattice strain as a function of illite crystallinity in pelitic rocks. *J. Metamorph. Geol.* **15**, 267-281.

KIM, JIN-WOOK, PEACOR, D.R., TESSIER, D. & ELSASS, F.A. (1995): A technique for maintaining texture and permanent expansion of smectite interlayers for TEM observations. *Clays Clay Minerals* **43**, 51-57.

KODAMA, H. (1966): The nature of the component layers of rectorite. *Am. Mineral.* **51**, 1035-1055.

LANSON, B. & CHAMPION, D. (1991): The I/S-to-illite reaction in the late stage diagenesis. *Am. J. Sci.* **291**, 473-506.

LI, GEJING, PEACOR, D.R., BUSECK, P.R. & ÁRKAI, P. (1998): Modification of illite–muscovite crystallite-size distributions by sample preparation for powder XRD analysis. *Can. Mineral.* **36**, 1435-1451.

NADEAU, P.H. (1985): The physical dimensions of fundamental clay particles. *Clay Minerals* **20**, 499- 514.

________ (1998): The fundamental particle model: a clay mineral paradigm. Proc. 11th Int. Clay Conf., Ottawa, Canada (in press).

________ & BAIN, D.C. (1986): Composition of some smectites and diagenetic illitic clays and implications for their origin. *Clays Clay Minerals* **34**, 455-464.

________, TAIT, J.M., MCHARDY, W.J. & WILSON, M.J. (1984c): Interstratified XRD characteristics of physical mixtures of elementary clay particles. *Clay Minerals* **19**, 67-76.

________, WILSON, M.J., MCHARDY, W.J. & TAIT, J.M. (1984a): Interstratified clays as fundamental particles. *Science* **225**, 923-925.

________, ________, ________ & ________ (1984b): Interparticle diffraction: a new concept for interstratified clays. *Clay Minerals* **19**, 757-769.

________, ________, ________ & ________ (1985): The conversion of smectite to illite during diagenesis: evidence from some illitic clays from bentonites and sandstones. *Mineral. Mag.* **49**, 393-400.

NIETO, F. & SÁNCHEZ-NAVAS, A. (1994): A comparative XRD and TEM study of the physical meaning of the white mica "crystallinity" index. *Eur. J. Mineral.* **6**, 611-621.

PEACOR, D.R. (1992): Diagenesis and low-grade metamorphism of shales and slates. *In* Minerals and Reactions in Atomic Scale: Transmission Electron Microscopy (P. Buseck & P. Ribbe, eds.). *Rev. Mineral.* **27**, 335-380.

REYNOLDS, R.C., JR. (1992): X-ray diffraction studies of illite/smectite from rocks, <1 μm randomly oriented powders, and < 1 μm oriented powder aggregates: the absence of laboratory-induced artifacts. *Clays Clay Minerals* **40**, 387-396.

ŚRODOŃ, J., ANDREOLI, C., ELSASS, F. & ROBERT, M. (1990): Direct high-resolution transmission microscopic measurement of expandability of mixed-layer illite/smectite in bentonite rock. *Clays Clay Minerals* **38**, 373-379.

________, ELSASS, F.D., MCHARDY, W.J. & MORGAN, D.J. (1992): Chemistry of illite–smectite inferred from TEM measurements of fundamental particles. *Clay Minerals* **27**, 137-158.

ŠUCHA, V., ŚRODOŃ, J., ELSASS, F. & MCHARDY, W.J. (1996): Particle shape versus coherent scattering domain of illite/smectite: evidence from HRTEM of Dolna Ves Clays. *Clays Clay Minerals* **44**, 665-671.

VEBLEN, D.R., GUTHRIE, G.D., JR., LIVI, K.J.T. & REYNOLDS, R.C., JR. (1990): High-resolution transmission electron microscopy and electron diffraction of mixed-layer illite/smectite: experimental results. *Clays Clay Minerals* **38**, 1-13.

WARR, L.N. & NIETO, F. (1998): Crystallite thickness and defect density of phyllosilicates in low-temperature metamorphic pelites: a TEM and XRD study of clay-mineral crystallinity-index standards. *Can. Mineral.* **36**, 1453-1474.

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