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EVOLUTION, CURRENT SITUATION, AND GEOLOGICAL IMPLICATIONS OF THE "FUNDAMENTAL PARTICLE" CONCEPT

FERNANDO NIETO[1]

Instituto Andaluz de Ciencias de la Tierra, Departamento de Mineralogía y Petrología,
Universidad de Granada – CSIC, Avenida Fuentenueva s/n, E-18002 Granada, Spain

JAVIER CUADROS

Departamento de Ciencias de la Tierra y Química Ambiental, Estación Experimental del Zaidín (CSIC),
Profesor Albareda 1, E-18008 Granada, Spain

ABSTRACT

The fundamental particle concept has evolved from its original formulation in order to take into account experimental evidence, in particular lattice-fringe images and the polar character of I/S. This process has brought the concept closer to the traditional mixed-layer view, the difference in interpretation lying only in the three-dimensional coherence across "fundamental particles". Recent evidence suggests that there is coherence through smectite layers. We thus consider that the fundamental particle model does not contribute to an understanding of I/S genesis.

Keywords: fundamental particle, illite, mixed-layer, smectite.

SOMMAIRE

Le concept des particules fondamentales a évolué depuis l'énoncé original, afin de rendre compte de l'évidence expérimentale, en particulier les images du réseau en haute résolution et le caractère polaire des interstratifiés I/S. Cette évolution a rapproché le concept des points de vues traditionnels des intercalations de couches mixtes, la différence en interprétation se limitant maintenant à la cohérence tri-dimensionnelle transversale à l'orientation des "particules fondamentales". D'après certaines observations récentes, il y aurait cohérence au travers des niveaux de smectite. A notre avis, le modèle de particules fondamentales ne contribue pas une meilleure compréhension de la genèse des interstratifications d'illite et de smectite.

(Traduit par la Rédaction)

Mots-clés: particules fondamentales, illite, interstratifié I/S, smectite.

INTRODUCTION

Nadeau and coworkers defined new concepts in relation to mixed-layer illite–smectite (I/S), namely illite fundamental particles (*e.g.*, Nadeau *et al.* 1984a) and interparticle diffraction (*e.g.*, Nadeau *et al.* 1984b). The original concepts implied that fundamental particles are chemically homogeneous, of illite composition, and physically independent from each other, without crystallographic continuity between them in the rock. From this point of view, I/S is simply a succession of illite fundamental particles separated by exchangeable cations. Owing to their homogeneous chemical composition, the fundamental particles are a single thermodynamic phase: illite. The smectite-to-illite transformation has been interpreted as an Ostwald ripening process in which the driving force is the minimization of the surface energy in the system (Eberl & Środoń 1988).

In this paper, we present an overview of the evolution undergone by these concepts and the difficulties encountered in their application. In our opinion, the evolution has caused a convergence in the real meaning of fundamental particles and the more traditional mixed-layer concepts.

EVIDENCE FROM LATTICE-FRINGE IMAGES

The first TEM images from I/S samples showed only layers with a thickness of 10 Å; in these images, illite and smectite layers were not distinguishable owing to

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[1] *E-mail address*: fnieto@goliat.ugr.es

the collapse of the smectite layers in the vacuum of the transmission electron microscope (TEM) or to ion milling (*e.g.*, Ahn & Peacor 1986a). Later, Guthrie & Veblen (1989) demonstrated by image simulation that smectite and illite layers can be distinguished under certain conditions of focus, which enabled them to recognize both types of layers in I/S samples (Veblen *et al.* 1990). They found that the ratios of illite and smectite layers in their micrographs are consistent with those obtained by standard XRD procedures. In addition, they found that most of the small crystals are thicker than what the fundamental particle theory predicts for their corresponding illite–smectite compositions. The same results have been reported by other authors using the focus conditions of Guthrie & Veblen (Lindgreen & Hansen 1991, Nieto *et al.* 1996) or using sample pretreatments in order to maintain smectite layers in their uncollapsed state (Ahn & Peacor 1986b, Bell 1986, Środoń *et al.* 1990,Vali *et al.* 1991).

These studies have shown that in the original rock, there is a real succession of two different kind of layers, recognized either by their different thickness or their different composition in terms of tetrahedrally coordinated ions (Al/Si ratio). These layers constitute packets thicker than fundamental particles. From the point of view of the fundamental particle theory, these packets can be interpreted as being stacks of fundamental particles with a high degree of orientation. According to this interpretation, the smectite layers are only the boundary between illite fundamental particles. Wilson (1990) indicated that the verification could be made by testing the packets for electron diffraction. The presence of a single-crystal pattern would favor the existence of real mixed-layer in the samples, whereas the lack of it would support the fundamental particle interpretation.

EVIDENCE FROM CHEMICAL AND SPECTROSCOPIC DATA

Chemical analysis of I/S samples covering the whole range from pure smectite to pure illite shows that there is a continuous transition in composition, with increasing Al and K, and decreasing Si (*e.g.*, Eslinger *et al.* 1979, Środoń *et al.* 1986, Lanson & Champion 1991, Cuadros & Altaner 1998). This is in contradiction with the original fundamental particle theory, which states that I/S with 50% illite and above consists only of illite particles (Nadeau *et al.* 1984). Therefore no chemical evolution would be expected from that point on. Although most of the discussion about the fundamental particle theory has focused on I/S, the original formulation of the theory also included trioctahedral mixed-layer clays. Hence, in view of this theory, corrensite would also consist of chlorite fundamental particles. Nevertheless, results of electron-microprobe analyses (EMPA) and analytical electron microscopy (AEM) have demonstrated that corrensite has a well-defined chemical composition different from chlorite and saponite (Shau *et al.* 1990).

In addition, different studies have presented evidence for the existence of two chemically different sheets of tetrahedra in I/S (different proportions of Al and Si), asymmetrically distributed in the *TOT* layer so that it acquires a polar character. This evidence was obtained by means of alkylammonium ion exchange (Lagaly 1979), nuclear magnetic resonance (NMR) (Altaner *et al.* 1988, Jakobsen *et al.* 1995), and combined X-ray diffraction (XRD) and thermal analysis (Cuadros & Linares 1995). These results are also in contradiction with the original fundamental particle concept, in which smectite was postulated to consist of particles one layer thick, and illite, to be particles two or more layers thick. These particles were assumed to have symmetrical *TOT* layers, with no polar character.

In order to reconcile the original theory with this chemical and spectroscopic evidence, the fundamental particle concept evolved so that the illite particles were assumed to have sheets of tetrahedra typical of a smectite composition at their terminations. Thus, the fundamental particles would have a different chemical composition at the external surfaces with respect to the core. These external surfaces would adsorb exchangeable cations in the same fashion as smectite interlayers (Środoń *et al.* 1992).

CURRENT SITUATION

At this point, the difference in interpretation between the fundamental particle theory and the mixed-layer concept lies only in the way illite layers are integrated in the undisturbed crystallites. The former theory views the fundamental particles as turbostratically stacked, even though they can be perfectly parallel. Their smectite-like terminations act as smectite interlayers (therefore particle terminations have a polar character). The fundamental particles grow and are physically independent of each other. The "mixed-layer" point of view considers the crystallites as actually composed of smectite and illite layers (also with polar character). The dispersion processes for XRD and TEM analyses would easily disarticulate these crystals into "fundamental particles" with no geological meaning with respect to the original sample. The physical difference between these two models is the presence or lack of tridimensional coherence within the crystallites.

Evidence about the coherence of crystallites across smectite layers is still scarce owing to the technical difficulty of obtaining bidimensional TEM images of materials that are easily damaged by the electron beam. Peacor (1998) focuses his review of the fundamental particle question on this kind of evidence. In his opinion, it suggests that there is coherence across a number of layers within the homogeneous packets that can be imaged, even where there is a significant proportion of smectite layers. The number of layers where coherence can be observed is always a minimum owing to the difficulty posed by the wavy morphology of smectite. On the other hand, Reynolds (1992) showed by means of XRD patterns of rock fragments and unoriented separates that turbostratic displacements occur at the smectite interlayers. This is in agreement with electron diffraction patterns of I/S, which show turbostratic disorder even in cases where tridimensional coherence was observed in local areas by means of high-resolution transmission electron microscopy (HRTEM). The apparent inconsistency between these two approaches seems due to the fact that the number of layers necessary to identify coherency in each method is different. With X-ray and electron diffraction, one needs a large number of coherent layers to see them as such. HRTEM is able to recognize coherence even in a series of three layers. Therefore, diffraction techniques miss short-range coherence. As I/S becomes richer in the smectite component, the number of incoherent interlayers increases, but *not all the smectite interlayers* are incoherently related, as the fundamental particle theory assumes. Very recently, Guthrie & Reynolds (1998) presented a slightly different interpretation. They indicated that the degree of rotational disorder in smectite interlayers is limited to less than 15°. These small angles would produce the loss of coherent X-ray diffraction, but local coherency could be detected by HRTEM. They refer to this angular relationship as being semicoherent.

We would also like to briefly comment on the use that has been done of scanning electron microscopy to support the fundamental particle theory. SEM micrographs of I/S samples that have been critical-point dried show a more delicate morphology than those that are not treated prior to SEM observation [compare Fig. 1 in Nadeau *et al.* (1985) with Figs. 4 to 22 in Keller *et al.* 1986)]. This is because the critical-point drying treatment better preserves the original morphology by preventing coalescence of particles while drying in the microscope vacuum. It has been stated that the very fine fibers observable in the critical-point micrographs argue in favor of the fundamental particle concept. A simple calculation demonstrates that these fibers are at least two orders of magnitude thicker than the assumed dimensions of fundamental particles. Hence, SEM resolution is too faint to provide information about this question.

DISCUSSION

In our view, the evolution of the fundamental particle concept in order to meet the experimental evidence has brought it very close to the traditional mixed-layer interpretation. In fact, Wilson (1990) stated "... and perhaps (the fundamental particle model) can be regarded as differing only subtly from the MacEwan crystallite model". This difference lies only in the crystallographic coherence across smectite interlayers. The accumulated evidence in this respect, though still scarce and partial, points toward the presence of crystallographic continuity, as previously stated. If this is the case, the contribution of the fundamental particle concept to the genetic interpretation of I/S is not clear.

Moreover, the fundamental particle model would seem to present a number of difficulties with regard to accepted mineralogical concepts. It requires the acceptance of a systematically different chemical composition at the borders and the core of illite particles that is not related to changes in the physical and chemical conditions during their crystallization, but only to the fact that the particle has reached its boundary. Also, if illite is accepted as the mineral phase present in fundamental particles, the exchange cations adsorbed in the external faces of the particles, necessary to balance its residual negative charge, have no clearly defined relation with the chemistry of the mineral.

The driving force for the growth of fundamental particles toward thicker crystallites has been considered to be the decrease of surface energy through a process of Ostwald ripening. In agreement with Primmer's (1994) point of view, we believe that this would only be possible in the case of a chemically homogeneous system, as Ostwald ripening is defined for homogeneous thermodynamic phases. In contrast, we observe that during the smectite-to-illite transformation, there is not only an increase of the mean thickness of particles, but also a constant chemical evolution toward a less silicic and more aluminous and potassic composition. Furthermore, the mechanism by which Ostwald ripening is supposed to operate is not clear to us, because it would imply that every time a new layer of illite is accreted on a fundamental particle, the previous sheet of tetrahedra should change composition from smectite type to illite type, which would seem to be unlikely (Primmer 1994, Altaner & Ylagan 1997).

In conclusion, we contend that the fundamental particle concept does not contribute to an understanding the genesis of I/S and illite with respect to the traditional mixed-layer concept; on the other hand, it has supplied clay mineralogy with useful concepts, as in the case of interparticle diffraction, which in our view is demonstrated in the experiments of Nadeau *et al.* (1984b). Today, the only difference between the two models is the size of the coherent crystallites and, therefore, the inclusion of smectite layers as crystallite components. Present evidence from HRTEM supports the mixed-layer model. The only important geological implication of the quandary between the two views is the thermodynamic consideration of I/S. The fundamental particle theory implies that illite is the only thermodynamic phase for I/S with 50% illite layers and above. Finally, the fundamental particle concept admits dissolution and precipitation as the only mechanism operating during smectite-to-illite transformation up to 50% illite, followed by Ostwald ripening. The "mixed-layer" view is compatible with both solid-state transformation and dissolution – precipitation mechanisms.

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