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# Nannostructure and significance of fish coprolites in phosphorites

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#### Abstract

Phosphatic coprolites (0.5-2 cm long) occurring in Cretaceous-Eocene phosphorites of Tunisia, Morocco, Mauritania, Senegal, and Egypt were investigated for their petrology by using SEM. They exhibit a homogeneous porous apatite structure with a few fish remains. The nannostructures of the coprolites consist of inframicron-sized and botryoid-type apatite microparticles. Spherical cavities and cavity-infilled inframicron-sized apatite globules are typical in the coprolites and are apparently formed by extracellular precipitation of phosphate around microbial organisms. We presume that the coprolites studied here may belong to fishes, whose excrements contain abundant organic matter and phosphate. Phosphatization of excrements appears to be a microbial process controlled by the microenvironment.

The nannostructures observed in the coprolites investigated here differ significantly from the nannostructures of other studied phosphatic grains (pellets and coated grains) in the same samples which are mostly heterogeneous and consist of ovoid-type apatite particles. In the case they are not reworked, the centimetre to decimetre thick coprolite beds in phosphorites indicate a lack of detrital input and strong bottom water currents at the sediment–water interface during deposition and subsequent phosphatization of the excrements.

## 1. Introduction

Most granular or nodular phosphorites are constituted by a variety of phosphatic particles which include phosphatized skeletal or test fragments, internal moulds, nodules, coprolites, ovoids, peloids, coated grains, aggregates, and lithoclasts (Slansky, 1980, 1986; Giot, 1988). The size and origin of these particles differ. Therefore, the recognition of various types of particles and their abundance within the phosphorites/phosphate horizons may provide information about apatite formation and paleoenvironmental conditions.

Several workers have reported on the nannostructures of phosphorite particles and phosphorites in general by using scanning electron

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microscope (SEM) together with microanalysis (Riggs, 1979; Bignot, 1980; O'Brien et al., 1981; Lamboy, 1986, 1987a,b, 1990a,b, 1993; Soudry, 1987; Soudry and Lewy, 1988, 1989; Purnachandra Rao and Nair, 1988; Bréhéret, 1991). Recently, the micro- and nannostructures of various types of phosphate particles including coprolites were reported from phosphorites from the Peru margin recovered during ODP Leg 112 (Garrison and Karstner, 1990; Lamboy, 1994) and from the Oman margin during ODP Leg 117 (Purnachandra Rao and Lamboy, in prep.). The purpose of this paper is to report on the nannostructures of morphologically well defined, small and lithified coprolites (phosphatized fish excrements) which showed typical and identical nannostructures in various phosphorites. We have also compared these nannostructures with those reported in lentiform beds of primary phosphate from the Negev (Soudry, 1992).

# 2. Previous recognition criteria and meaning of coprolites

The term coprolite has been defined as fossilized animal excrement independent of its size, composition and the type of animal that produced. This term includes faecal pellets (Amstutz, 1958; Brönnimann and Masse, 1968; Cuvillier et al., 1969; Brönnimann et al., 1972; Häntzschel, 1975; Lehmann, 1975) whose origin can easily be recognized. Since many coprolites are phosphatic (Thulborn, 1991), several workers (Boujo, 1976; Slansky, 1980, 1986; Jarvis, 1980; Giot, 1988; Prévôt, 1990) have used the term coprolite in the sense of Cayeux (1939, 1941, 1950) who described coprolites in detail from phosphorites. According to Cayeux, coprolites are particles with a broad cyclindrical shape, approximately 2 cm long and 1 cm in diameter; their transversal sections are circular but rarely ellipsoidal; their ends are narrow and rounded, sometimes showing subbased cones with rounded top. All of them have a smooth surface but some show transversal constrictions. Cayeux attributed these excrements to marine vertebrates, especially to fishes and reptiles. Since reptile excrements are usually longer than 2 cm (Amstutz, 1958; Thulborn, 1991), one may consider the smaller ones (up to 2 cm) to be from fishes. Although there is a lack of good recognition criteria, we believe that the structure and constituents of coprolites are reflective of the original excrements and the animals that produced may be recognized. Therefore this kind of particles should not be clubbed with smaller faecal particles (pellets, coated grains), especially those produced by invertebrates.

Cayeux (1950) and Giot (1988) have suggested that the fish excrements are originally fragile and occur as irregular coprolite fragments in many heterogeneous sandy phosphates. The homogeneous internal aspect is characteristic of these coprolites. Thin sections show that the coprolites comprise of none or a few particulate inclusions and sometimes the fragments of bones. Apatite is isotropic or slightly anisotropic and quite limpid in these thin sections.

#### 3. Materials and methods

The samples studied here are from Cretaceous to Eocene phosphorites of Tunisia, Gafsa and Sra el Ouartene Basins (Sassi, 1974; Chaabani, 1978; Ben-Amar, 1985; Zaier, 1984), Morocco (Boujo, 1976; Prévôt, 1990; Azzouzi, in prep.) and Mauritania (Lo, 1987), Eocene phosphorites of Senegal (Flicoteaux, 1982), and upper Cretaceous phosphorites of Egypt (Ahmed, 1988, 1992). All samples are from phosphorite beds consisting of several phosphatic coprolites and other phosphatic particles embedded in a fine-grained phosphatic matrix. Polished and thin sections were prepared. Freshly broken surfaces of coprolites and other phosphate grains of different origin (approximately 5-10 particles per sample and more than 100 in total) were examined under SEM (Jeol T-20).

#### 4. Results

## 4.1. Nannostructures of coprolites

The colour of coprolites varies from white to brown. Polished (Fig. 1A, B and D) and thin sections (Fig. 1E) of coprolites show that they are predominantly homogeneous. The peripheral area of many coprolites appears as a darker or more light coloured part, or a light coloured area underlined by an internal darker rim (Fig. 2B and D). The general fabric varies from one coprolite to the other. SEM studies at low magnification (Fig. 1C) also attest to the homogeneous structure and show a few inclusions like fish remains. Apatite nannostructures are typical and composed of inframicron globules and/or botryoids.

The fabric of inframicron globule-based nannostructure can be divided in two categories. (a) The fabric showing globules more or less agglomerated without any specific disposition (Fig. 2A). This is not frequent and can be compared with



Fig. 1. Macro- and microscopic aspects of coprolites. (A) Coprolites from Taiba (Senegal). Some of them have their ends truncated. One is broken (black arrow) and shows an homogeneous phosphate matter. (B) Polished section of a small phosphatic bed from Sra Ouertane (Tunisia) containing numerous coprolites, of about 0.2–0.5 cm in section, with various aspects. (C) SEM photograph of a coprolite from Metlaoui (Tunisia) showing 3 characteristic transversal constrictions; one small area is broken and show the homogeneous, porous, internal structure (black arrow). (D) Thin section of the sample of Fig. 1B observed in plane polarized light (PPL) showing different aspects of coprolites; the black arrow indicates the localization of Fig. 3E. (E) Detail of (D), (PPL); the dash-line corresponds to the periphery of the coprolite.

those reported in soft copronodules recovered from the Peru and Oman margins (Lamboy, 1994; Purnachandra Rao and Lamboy, in prep.). They sometimes appear compact (Fig. 2B and C). (b) The porous fabric consisting of spherical cavities of variable size  $(1-2 \ \mu m$  diameter) surrounded by inframicron-sized apatite globules (Fig. 2D and E) is more common and shows slight variation from one coprolite to the other. Around cavities, apatite globules form egg shell-like structures, sometimes with two concentric walls. Sometimes the apatite globules are randomly distributed (Fig. 2F), while in others they are regular between the cavities (Fig. 2G). Within one coprolite, the cavities are



more in one area (Fig. 2I) and cavity filled apatite globules in the adjoining area; all transitional stages exist between porous and compact structures (Fig. 2H). Similar fabrics have been observed in some phosphate particles (Bignot, 1980; Panczer, 1990; Moutaouakil and Giresse, 1993), in primary bedded (autochtonous) phosphorites (Soudry, 1992) and in gravels and coprolites from the Peru and Oman margins (Lamboy, 1994; Purnachandra Rao and Lamboy, in prep.).

The fabric showing botryoid-based microstructure is more or less compact and exhibits a radialfibrous structure (Fig. 3B). Botryoids are sometimes arranged in larger globular masses. The most typical aspect is the presence of cavities between botryoids, where one can observe the mammillated external surfaces (Fig. 3C). The botryoid-based structures and capsule-like structures coexist in some coprolites. The more compact structures show the presence of ovoidal cavities of about 1  $\mu$ m long (Fig. 3C and D) in a sporadic manner.

#### 4.2. Nannostructures of elongated faecal pellets

The phosphatic sands from Mauritania and Senegal occur as cylindrical and/or elongated pellets (Fig. 4A). They resemble faecal pellets produced by marine bivalves (Moore, 1939; Häntzschel et al., 1968; Arakawa, 1970, 1972) or echinoderms (Arakawa, 1971). Some cylindrical pellets have the same fabric as coprolites with spherical cavities and capsule-like inframicronsized apatite globules. Many other pellets contain numerous small siliciclastic and biogenous fragments (Fig. 4B and C). Although the structure is more compact, the apatite particles appear as ovoids showing a crystalline aspect, most probably corresponding to overgrowth (Fig. 4D).

## 4.3. Other types of grains

Other phosphatic grains found in these phosphorites are pellets and coated grains. The study of these grains from Tunisia (Lamboy, 1987a, 1990b; Ben-Amar, 1985) and from Morocco phosphorites (Azzouzi, 1989, in prep.) shows that the microstructure of the cortex portion of coated grains and pellets is similar and exhibits heterogeneous nannostructures based on ovoid particles and/or small botryoids. Ahmed (1992) examined coated grains from the Red Sea phosphorites and found that the nannostructures are more varied than in the pelletal ones.

### 5. Discussion

The coprolites described here exhibit characteristic homogeneous and porous nannostructures, constituted by inframicron-sized globules and botryoidal apatite particles. On the other hand, the pellets and coated grains are largely heterogeneous and the nannostructures are constituted by ovoids and partly by botryoids. Detailed studies on the nannostructures of coprolites and other phosphatic particles recovered from phosphatic horizons of ODP Legs of the Peru and Oman margins (Lamboy, 1994; Purnachandra Rao and Lamboy, in prep.) are in agreement with the above observations. The size and morphology of the initial excrements were preserved. It is therefore suggested that the homogeneous nannostructure is typical for coprolites and distinctly differs from other particles. As many coprolites contain fish remains as inclusions, we believe that the initial excrements may belong to carnivorous fishes.

The nannostructures consisting of spherical cavi-

Fig. 2. Nannostructures based on inframicron-sized apatite particles in coprolites. (A) Characteristic porous aspect of more or less agglomerated apatite globules (Bofal, Mauritania). (B) and (C; detail of B) Typical aspect of a more compact nannostructure composed of inframicron-sized apatite globules (Taiba, Senegal). (D) and (E; detail of D) The capsule-like or egg shell-like structure composed of inframicron-sized apatite globules organized around micron-sized spherical cavities (Oued Tselja, Tunisie). Some numerous cavities are filled with overgrowths of apatite globules. This aspect is the most typical in coprolites; it is very similar to the one observed in primary bedded phosphatic microbial mats (compare figs. 3E, 4B and 4E of Soudry, 1992). (F) A more porous nannostructure with less cavities (Tselja). (G) Another porous aspect with numerous cavities (Metlaoui, Tunisie). (H) Contact between a porous area and a compact structure. (I) Another aspect showing small areas with numerous continguous cavities scattered within compact structures.



Fig. 3. Nannostructures based on botryoids in coprolites. (A) and (B; detail of A) characteristic porous structure composed of agglomerated botryoids with a typical botryoidal surface. Note also the characteristic internal pores between botryoids (Ouled Abdoun, Morocco). (C) Porous and compact nannostructures based on botryoids. Note the presence of some small hexagonal crystals in an internal pore. White arrows point to small ovoidal cavities (Safaga, Egypt). (D) At high magnification, the aspect of the small ovoidal cavities similar to those of Fig. 3C in a coprolite from Bofal (Mauritania).

ties and cavity filled inframicron-sized globules (Fig. 2) are the most predominant in coprolites. Similar structures were also reported in manganese mineralized stromatolitic microlayers from deep sea polymetallic nodules and crusts (Janin, 1987) and in laminated phosphorites from Israel (Soudry, 1992). These workers interpreted the nannostructures as extracellular remnants of mat-forming microorganisms. The samples studied here are mainly granular phosphorites which show neither microstromatolites nor laminated structures. It implies that the mineralization processes are

similar in both microstromatolites/laminated structures and in well defined coprolite bodies. Phosphatization apparently took place first on external surfaces of the bacteria but before the complete decay of organic matter associated with the microbial structures. The final product seemed to be a egg shell-like mould with void inside. We therefore suggest that the above structures most probably correspond to the extracellular precipitation around globular bacterial bodies which subsequently disappeared. The botryoidal-based nannostructures (Fig. 3A–C) resemble those



Fig. 4. Structures of elongated faecal pellets. (A) Macroscopical aspect of pellets; all of them are not complete and show different sections and more or less numerous longitudinal depressions (Bofal, Mauritania). (B) Characteristic broken surface of a pellet showing heterogeneous structure with numerous siliciclastic and biogenic fragments (Bofal). (C) Characteristic structure of an elongated pellet from Taiba (Senegal) showing several biogenic fragments. (D) Detail of (C), porous and compact structure, the right part showing remains of ovoids transformed into elongated bad-formed crystals by apatite overgrowth.

shown by Lamboy and Monty (1987) and Lamboy (1993, 1994) who suggested that they represent phosphatised remnants of coalesced globular bacteria. Purnachandra Rao and Lamboy (in prep.) showed nannostructures composed of botryoids in coprolites of the Oman margin; here the botryoids agglomerate into larger spherulites which showed radial fibrous structures in cross section. Laboratory studies also showed similar botryoids corresponding both intra- and extracellular microbial apatite precipitation in the form of radial fibrous structures (Prévôt et al., 1989; Hirschler, 1990; Hirschler et al., 1990; Lucas and Prévôt, 1991). It therefore appears that the bacteria that colonized the initial faeces transformed them into hard coprolites by microbial processes. The existence and number of microbes in coprolites are not surprising keeping in mind the dense populations of bacteria inhabiting the intestines of living vertebrates. Human faeces, for example, may comprise 25–50% bacteria in weight (Asimov, 1959, in Thulborn, 1991). Carnivore fish faeces usually tend to be phosphate-rich due to undigested fish remains. The initial morphology of the faeces was preserved during phosphatisation. These imply that the microbial organisms utilized phosphorus located within and phosphatization was probably aided by the microenvironment within the faeces. The uniform nannostructure in several coprolites may indicate low microbe diversity and/or reflect the severity of environmental conditions in the digestive tract of marine fishes which limit the flora to only a few microbial groups (Sera and Kimata, 1972).

There exists a slight variation in the nannostructure of coprolites from one deposit to the other and also between coprolites. Similarly, this nannostructure distinctly differs from the faecal pellets and coated grains. In fact, all these grains were formed by phosphatization of initial supports. The initial composition and microenvironmental conditions within the granular supports apparently controlled the type of colonizing microorganisms and the resultant nannostructures formed during phosphatisation.

The nannostructures of some angular grains are similar to those in coprolites; since the initial fish excrements are fragile, one may consider that these angular grains may be the broken fragments of larger excrements of fish which subsequently phosphatized. However, some elongated pellets also show similar structures as in coprolites whose origin is not clear. They may correspond to elongated excrements of small-sized fishes. It is also possible that they were produced by coprophagic invertebrates, the excrements of which may contain remains of phosphatized excrements of fishes.

The peripheral area of many coprolites appears as a darker or more light-coloured part, or a lightcoloured area underlined by an internal darker rim (Fig. 2B and D). These differentiations are due to local porosity, content of organic matter, pyrite and/or another pigments, the compactness of the apatite structure and, for thin sections, to the viscosity of the mounting product (Fikri et al., 1989). These differentiations, as well as the general colour of coprolites, can be due to syngenetic and/or diagenetic processes and sometimes are underlined by consequent weathering processes.

The significance of coprolites in the geological record is important for several reasons. (1) Our studies reported here and elsewhere indicate that the coprolites can be distinguished by their nannostructures. In contrast, phosphorite grains derived from the microbial mats also showed similar nannostructures (Soudry, 1992). It implies that the process of phosphatization is similar in both the cases. On the other hand, Lamboy (1994) has shown that the early diagenetic phosphatization of excrements is a slow process and a copronodule can be compressed into a lenticular lense in argillaceous sediments, when sedimentation rate is high. Thus, the irregular lense-shaped phosphate bodies interlayered with clays (both authigenic and detrital) in Negev phosphorites (Soudry, 1992) may actually represent compressed copronodules. We are of the opinion that in order to confirm nannostructural similarities, the primary laminated beds of unequivocal origin such as those in Cambrian age may have to be investigated. (2) The density of fish excrement at the beginning of phosphatization is very low. So the coprolite levels may represent the absence of strong currents and bioturbation. Alternatively, coprolites were sometimes reexposed at the sea floor after phosphatization and consequently concentrated through winnowing processes. (3) Frequently, in sandy phosphatic deposits, coprolites are accumulated without reworking in a two centimetre to decimetre thick level with few smaller others grains (Fig. 1B). In this case, the detritic input was nil ("the silence of the earth", Busson and Cornée, 1986) and the anoxic conditions existed at the sediment-water interface did not permit burrowing and production of invertebrate faecal pellets. Therefore, only the fish excrements were deposited and subsequently phosphatized. If one considers that the nannostructures of the well defined grains correspond to reworked fragments of primary bedded microbial mats (Soudry, 1992), the remnants of significant primary beds comprising phosphatized mats should necessarily exist in the neighbouring beds.

# 6. Conclusion

The coprolites reported here predominantly show homogeneous and porous microstructure sometimes with a few fish remains. The nannostructure is composed of inframicron-sized and botryoid-type apatite particles, the former frequently organized in spherical cavities and capsule like-structures and are very similar to those described in primary bedded phosphorites (Soudry, 1992) corresponding to phosphatized microbial mats.

The nannostructural signature reinforces the hypothesis of microbial mediation in apatitogenesis. This study also shows that all phosphate particles are the result of phosphatization of primary particles (initial granular supports) of same size and shape. The presence of abundant coprolites in a phosphate level may represent either lack of significant detrital deposition and bioturbation or a reworking process after phosphatization of the excrements. As the nannostructures of coprolites and phosphatized fragments of microbial mats are similar, we should not confuse these two types of grains. The criteria for identification of these two types of grains is difficult and may be possible only if one take into account the gross textures and associated components of the phosphorite beds.

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