Abstract: Following a brief summary of some fundamentals in carbonate sedimentology (sedimentary petrography) that highlights the significance of organic matter, some examples of biocrystals in carbonate grains/particles, such as bioclasts or ooids, are provided as an introductory chapter to a discussion on gravitational cements versus endostromatolites. The gravitational cements, either marine (fibrous) or continental (dog-tooth), are made of hyaline (i.e., translucent) sparitic crystals whereas endostromatolites are made of colored sparitic crystals and/or micrite. Gravitational cements forms in the vadose zone whereas endostromatolites grow in small rock cavities in the marine phreatic zone. As such the latter can grow centripetally in all directions (not only downward).

Key-words:
- asymmetrical cement;
- geopetal infills;
- endostromatolites;
- microbes;
- vadose zone;
- phreatic zone

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Résumé : La biosignature de la sparite permet de distinguer un ciment gravitationnel des endostromatolithes.- Après à un bref rappel de quelques principes fondamentaux de la sédimentologie (pétrographie sédimentaire) des carbonates qui met en évidence l’importance de la matière organique, quelques exemples de biocristes dans les grains calcaires, tels que les bioclastes ou les ooids, sont présentés dans un chapitre d’introduction à une discussion portant sur la distinction entre ciments gravitationnels et endostromatolithes. Les ciments gravitationnels, qu’ils soient marins (fibreux) ou continentaux (en “dents de chien”), sont constitués de cristaux sparitiques hyalins (c’est-à-dire translucides) tandis que les endostromatolithes sont constitués de cristaux sparitiques colorés et / ou de micrite. Les ciments gravitationnels se forment dans la zone vadose alors que les endostromatolithes poussent probablement dans de petites cavités de la roche dans la zone phréatique marine. En tant que tels, ces derniers peuvent pousser de manière centripète dans toutes les directions (et pas seulement vers le bas).

Mots-clefs :
- conservation exceptionnelle (silicification) ;
- bivalves rudistes ;
- Laluzia ;
- Caribbea ;
- Parastroma ;
- Hippuritidae ;
- Porto Rico ;
- Crétacé

1 Dépt. STU, Fac. Sci. Tech., UBO, 6 avenue Le Gorgeu, CS 93837, F-29238 Brest (France)
bgranier@univ-brest.fr

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1. Introduction

In his PhD thesis, the present author (Granier, 1987: p. 96-97, Pl. 14, fig. a; Pl. 15, fig. d; Pl. 16, figs. c-d; Pl. 17, figs. a-b; Pl. 51, figs. h-l) described from uppermost Jurassic strata in “Sierra Helada” (Serra Gelada, Alicante, Spain) "des ciments [sic] asymétriques" [asymmetric cements] made of layers of radiating fibrous sparitic crystals with an amber yellow color and micritic laminae or bulges. Granier (1987) pointed out that these structures present in former rock cavities grew in various directions, i.e., they were not necessarily formed under gravitational dynamics, and that, accordingly, they should not be called "microstalactites". He interpreted them to be of microbial origin and more specifically "endostromatolites" sensu Monty (1980, 1982) because these structures have grown up in cavities. It appears that, in the scientific literature, such features are commonly erroneously interpreted as pendant or microstalactitic cement (e.g., "gravitational cement" in Fügels, 2004: Pl. 34, fig. 6), and even as microstalacmitic (!) features (e.g., Arniaud-Vanneau, 1980; Arniaud, 1981: vol. 3, Figs. IV, VII, Pls. 33-34). Recent publications document limestones exhibiting similar features from Italy (Amadio et al., 2018) with "dendritic shrub-like fabric" and Algeria (Belkhedj et al., 2019) with "asymmetric fibrous cements (AFC)". In the present author's view, all these structures, being partly of a microbial nature, should not be considered as "cements" (i.e., a word the use of which should be restricted to crystals resulting of the purely chemical precipitation from a brine in voids) but as microbial crusts growing in cavities, i.e., "endostromatolites", as shown through the following simple and naturalistic approach.

2. Material and method

The studied material is from the present author's collection (currently stored at University of Western Brittany in Brest). It comprises a set of thin sections from various origins (France, Iran, Spain, and Ukraine), which are used either as references (e.g., bioclasts and ooids) for comparison or as case specimens illustrating either endostromatolites (Spain) or gravitational cements (France), or both of them (Ukraine).

The significance of organic matter in carbonate sedimentology is still noticeably undervalued, which brings us to summarize some essentials on the bioclast”. The color and the pseudopelieochroism are directly related to the organic matter.
**Figure 2: A-B)** Rudist shell with a colored primary calcitic part to the left and a secondary calcitic part to the right. The latter consists of a mosaic of calcite crystals that replaced the original aragonitic part. Ghosts of the primary structure, i.e., many linings, are locally preserved. Thin section HL 42 bis (A) 45x60, Serra Gelada, L’Alfàs del Pi, Alicante (Spain), “Calcaires à Rudistes et à Huitres”, lower Albian. A) transmitted light; B) cross polars. C-D) Radial fibrous sparitic marne ooids. Thin section BR 2530, Golan, Shirwan, North Khorasan (Iran), Tirgan Formation, upper Barremian. Transmitted light. E-F) Rudist shell (Eoradiolites) with a secondary calcitic part to the left and a primary calcitic cellular part to the right. The cellular part (right) consists of walls made of yellowish calcite and vugs cemented by a transluscent drusy sparitic cement. The secondary calcitic part (left) consists of a mosaic of calcite crystals that replaced the original aragonitic part. Ghosts of the primary structure are locally preserved, such as the irregular curved lining in the middle of both pictures. Thin section HL 74 (A) 45x60, Serra Gelada, L’Alfàs del Pi, Alicante (Spain), “Calcaires à Rudistes et à Huitres”, lower Albian: E) transmitted light; F) cross polars. All photos same scale bar = 250 µm.

Contents of these calcite crystals. In contrast, hyaline sparite is a translucent cement resulting from the direct precipitation of calcite from a brine in vugs to form palisadic or drusy cements, or it could be the mosaic replacement of a primary aragonitic or high-Mg calcite cement. Consequently, most hyaline calcite crystals that result from chemical precipitation should be treated as cement *sensu stricto*, whereas most colored calcite crystals are either primary biocrystals or re-crystallized biocrystals and accordingly they should NOT be treated as cement.

A short list of seminal books helping on identification of bioclasts in sedimentary petrography includes Böggild (1930), Mišik (1966), Majewske (1969), Horowitz and Potter (1971), Samuel et al. (1972), Fügler (1982, 2004, 2010), or the more recent Dias-Brito et al. (2017), amongst many others. They provide illustrations of 1) primarily calcitic bioclasts (e.g., echinoids, Fig. 1.A-B herein), 2) primarily aragonitic bioclasts (e.g., gastropods), eventually secondarily replaced (e.g., bivalves, Fig. 4.E herein) or dissolved and later secondarily cemented (e.g., gastropods, Fig. 4.B herein), and 3) dual calcitic-aragonitic bioclasts (e.g., serpulids or rudists, Figs. 1.C-D, 2.A-B, E-F herein). Primary biocrystals and replaced biocrystals are amber yellow colored or "cloudy" due to their residual organic matter content. A few examples are documented and illustrated herein.

### 3. Examples of primary biocrystals and replaced biocrystals

Radial ooids - samples BR2530 (Fig. 2.C-D): The material was originally collected at Golan, 10 km south of Shirwan, North Khorasan Province, Iran (GPS coordinates: 37°14′13.8″N 57°54′17.3″E). The microfacies of this rock sample from the lower part of the Tirgan Formation from the lower part of the Tirgan Formation, upper Barremian, is typically a floatstone of *Balkhiana balkhanica* Mamontova and *Pseudoactinoporella iranica* Bucur et al. with a bioclastic-oolitic grainstone matrix. Nobody ever questioned the role of organic matter in bioclasts whereas fierce debates have taken place regarding its role in ooids (e.g., Dangerd, 1936; O’Reilly et al., 2017; Diaz & Eberli, 2019, and references therein) and in microbial crusts or stromatoids *sensu* Kalkowsky (1908). Microbes and mucilage (i.e., extracellular polymeric substances or EPS) were well documented in modern and fossil stromatolites and oncoinds. On the opposite side until recently (e.g., Brehm et al., 2004; O’Reilly et al., 2017; Diaz & Eberli, 2019, and references therein), their role in the ooid factory was still questioned. Iranian ooids are calcitic with a radial fibrous fabric suggesting they were originally formed as high-magnesium calcite. Their cortices are light brown, which implies that organic matter is embedded in their crystal framework.

Rudist shells - samples HL (Figs. 1.C-D, 2.A-B, .E-F): The material was originally collected at Morro de Sant Jordi, Parc Natural de la Serra Gelada, L’Alfàs del Pi, Province of Alicante, Spain (GPS coordinates: 38°34′03.0″N 0°03′21.8″W). Per definition rudist floatstones with wackestone to bacinella bindstone (Granier, 2012, and in prep.) matrices from the "Calcaires à Rudistes et à Huitres" (Granier, 1987), lower Albian, yield numerous bivalve shells. In both samples (HL42bis: Requieniaeidae, and HL74: Requieniaeidae and *Eoradiolites gr. davidsoni* Hill), the originally aragonitic inner layer of the rudist shells is replaced by a mosaic of large calcite crystals that retain few organic ghosts of the primary structure (Figs. 1.C-D, 2.A-B, .E-F).

Bivalve shell - sample 65 (Fig. 4.E): The material was originally collected on the side of the parking area near the Fierney cable-car station of Crozet, Ain Department, France (GPS coordinates: 46°17′03.3″N 6°00′22.0″E). The bioclastic grainstone facies from the uppermost part of the Chambotte Formation (Böker, 1994; Granier, 2019a), lower Valanginian, yield numerous large benthic foraminifers, including *Pfenderina neocontroversia* (Pfender) and *Pseudocyclammina lituus* (Yokoyma) (Fig. 4.F). Surprisingly all primary aragonitic bioclasts were not dissolved to form micritic envelopes (e.g., Fig. 4.B); some were replaced by a mosaic of calcite crystals that retains some organic ghosts of the primary structure (Fig. 4.E). It is suggested here that the original amount of organic matter in the original biocrystals, as well as its later degree of oxidation and the mesh density of the original aragonite needles, could be clues to this structural difference.
4. Gravitational cement from the lowermost Cretaceous (lower Valanginian) of E France

Gravitational cement - sample 65 (Figs. 3.C, 4): Same as above for the "Bivalve shell - sample 65", the material was originally collected on the side of the parking area near the Fierney cable-car station of Crozet (GPS coordinates: 46°17'03.3"N 6°00'22.0"E), from the lower Valanginian part of the Chambotte Formation.

The fabric is dominantly grain-supported with well-sorted and commonly coarse grains (Fig. 3.C). It is a beach-rock facies as evidenced by the fibrous (hence marine) gravitational cement that developed in intergranular cavities below the largest grains (Fig. 4.B, blue arrowheads). This gravitational cement documents a marine vadose setting where beach-rocks form. Some micrite is also partly infilling cavities, including some micritic envelopes following the leaching of the primary aragonitic bioclasts (Fig. 4.B). A thin isopachous palisadic cement is locally observed (Fig. 4.B), which predates the leaching of aragonite. It suggests an initial marine phreatic setting for diageneric (subaqueous) hardground formation preceding emersion and (subaerial) beach-rock formation. Not all the aragonitic bioclasts were leached. As documented above, a few were replaced by calcite (Fig. 4.E). The micrite, which later percolated through the porous network (intergranular, intragranular and moldic porosity), commonly displays a geopetal layout (Fig. 4.B, E, G-H, J-L, orange arrowheads). It postdates the leaching that in turn postdates the gravitational cement. Finally, the remaining porosity was cemented by a meteoric phreatic cement and then a deep-burial drusy sparitic cement.

5. Endostromatolites from the uppermost Jurassic of SE Spain

Endostromatolites - samples SHJ (Figs. 3.A-B, 5): The material was originally collected at Morro de Sant Jordi, Parc Natural de la Serra Gelada, L’Alfàs del Pi, Province of Alicante (Spain) (GPS coordinates: 38°33'07.3"N 0°03'32.0"W). The stratigraphic unit was improperly named “Mud mounds à Clypéines” (GRANIÉR, 1987), Tithonian-lower Berriasian “Aloisalthella Limestones” based on the latest systematics of the algal epynym (GRANIÉR & LETHIERS, 2019). The rock sample corresponds to a microbial boundstone (Fig. 3.A), probably formed in a very shallow-water setting. Some large cavities between grains were sites for endostromatolitic growth. These commonly zoned endostromatolites comprise yellowish fibrous sparitic crusts and micritic crusts and bulges. Although they commonly look like they were formed under some gravity control, there are places where that was obviously not the case (Fig. 5.C-D). As already documented by GRANIÉR (1987), the remainder of the cavities were then filled with 1) dog-tooth sparitic cements (Fig. 5.C, E, white arrowheads), which are the mark of a continental phreatic setting, 2) red-colored marine micrites that have percolated downward through the remaining porous network and a fracture network, and finally 3) a drusy sparitic cement, which corresponds to the deep burial setting. At Serra Gelada, the Tithonian-lower Berriasian “Aloisalthella Limestones” are topped by an angular unconformity (GRANIÉR, 1987, Pl. 13, fig. d) in the form of a bored surface, a feature which suggests the occurrence of at least one episode of emersion and erosion preceding a flooding. The fracture network is post-early Valanginian in age (see GRANIÉR, 1987; GRANIÉR et al., 1995). It was polyphasic, as documented by several episodes of fracturing, phreatic cementation and red-colored micrite infill (e.g., GRANIÉR, 1987, Pl. 16, fig. a), which have occurred over part of the late Valanginian – earliest Aptian time interval.

Figure 3: A) Polished slab of an oncoid floastone to bindstone facies from Serra Gelada, L’Alfàs del Pi, Province of Alicante (Spain), “Aloisalthella Limestones”, Tithonian-lower Berriasian. White arrowheads point to stratiform binding micritic films, red arrowhead points to a patch of red-colored micrites corresponding to upper Valanginian – lower Aptian pelagic mud that has percolated downward into the porous network. Sample SH 19; B) Detail of a colored fibrous sparitic calciflower-like structure growing in a cavity of the “Aloisalthella Limestones” (GRANIÉR, 1987, Pl. 15, fig. d). Thin section SHJ 1B, Serra Gelada, L’Alfàs del Pi, Province of Alicante (Spain); C) Polished slab of the beachrock facies from Crozet, Ain Department (France), Chambotte Formation, lower Valanginian. Sample 65; D) Polished slab of a coquina facies from Bukivna/Буківна, Ivanofrankivsk Oblast (Ukraine), “Niżniower Kalkstein”, Tithonian-Berriasian. The yellow arrowhead points to a former shelter cavity below a bivalve shell, now fully cemented. Sample F3. Thick black arrowheads are upward-oriented; scale bars: red = 1 cm, white = 500 µm.
6. Endostromatolites and gravitational cement from the uppermost Jurassic of W Ukraine

Endostromatolites and gravitational cement – sample F3 (Figs. 3.D, 6): The material was originally collected near Bukivna/Буківна, Ivano-Frankivsk Oblast/Province, Ukraine (GPS coordinates: 48°59'04.5"N 24°58'07.4"E). The stratigraphic unit corresponds to the "Niźniower Kalkstein" (ALTH, 1882; GLĄZEWSKI, 1937; GRANIÈRES, 2019b, 2020), upper Tithonian-lower Berriasian, the Clypeus podolica (A. von) (1878) (see GRANIÈRES, 2019b, 2020), upper Tithonian-lower Berriasian, the stratum typicum of Actinoporella podolica (ALTH, 1878) (see GRANIÈRES, 2019b), that of Aloisaitheilla sulcata (ALTH, 1882) (see GRANIÈRES & LETHIERS, 2019), and also that of some agglutinated foraminifers described by CUSHMAN and GLĄZEWSKI (1949).

The rock sample corresponds to a coquina (Fig. 3.D), probably in a supratidal wash-over setting. Its texture is that of floatstone to rudstone because the allochons are large, including numerous gastropods, some corals, sponges, Arbitroides, and Gyroporella microstructures. The related large cavities between grains or inside them were sites for endostromatolithic growth (Fig. 6). Although the yellowish fibrous sparitic layers commonly look like they were formed under gravity control, that is not obvious in some places. These endostromatolites, commonly zoned, also comprise micritic bulges, either embedded or developing on the side of the crust. The emersion that affected the Niźniow limestone led to the dissolution of most aragonitic shells, leaving behind micritic envelopes, in a (?) continental vadose setting. The development of non-isopachous, hence partly gravitational, dog-tooth sparitic cement in all cavities, including in the micritic envelopes, means that the rock remained in a dominantly continental vadose setting (with at least one episode in a phreatic setting). The micrite, which percolated later through the porous network in a marine phreatic setting and almost completely filled in the cavities, postdates the dog-tooth cementation and is probably the mark of the Late Cretaceous marine transgression in the area.

7. Conclusions

As documented above, sparitic crystals forming gravitational cements, whether they are 1) fibrous in shape and the signature of a vadose marine setting (e.g., in the French sample, Figs. 3.C, 4) or 2) dog-tooth shaped and the signature of a vadose continental setting (e.g., in the Ukrainian sample, Figs. 3.D, 6), are hyaline/translucent. In contrast, sparitic biocrystals that form bioclasts are commonly colored by organic matter, which is also the case of sparitic crystals forming colored crusts in some rock cavities and that should be assigned a biogenic origin. Such individual colored crusts are here called endostromatoids whereas the assembly of colored sparitic and/or clotted micritic crusts in a similar setting is called endostromatolite, not cement. Besides being colored, contrary to the crystals forming gravitational cements that are always hyaline, endostromatolitic biocrystals are not necessarily growing gravitationally as documented herein (e.g., Fig. 5). Because no specific laboratory investigation of the organic contents of such endostromatolites has hitherto been attempted, it is felt that the features given above should serve as a preliminary guide to distinguish between endostromatolites (e.g., "gravitational cement" in FLÜGEL, 2004: Pl. 34, fig. 6) and genuine gravitational cement (e.g., "pennat (or gravitational) cement" in FLÜGEL, 2004: Pl. 33, fig. 4).

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**Figure 5:** Endostromatolites (red arrowheads) from Serra Gelada, L’Alfàs del Pi, Province of Alicante (Spain), “Aloisathella Limestones”, Tithonian-lower Berrissian. Thick black arrowheads in A-D are upward-oriented; E-I are oblique or horizontal sections (i.e., oblique or parallel to stratification). A) infill, cf. GRANIER, 1987, Pl. 51, fig. h. The red arrowheads point to non-strictly gravitational crusts. Thin section SHJ 1B; B) Thin section SHJ 1 b; C) The red arrowhead points to a non-gravitational crust whereas the white arrowhead points to a dog-tooth like sparitic cement. Thin section SHJ1B (covered); D) The red arrowheads point to a non-gravitational crust. Thin section SHJ 1B (uncovered); E) The white arrowheads point to the dog-tooth like sparitic cement. Thin section SHJ 1B (?); F) Thin section SHJ1B (covered); G) Thin section SHJ1B (covered); H) Thin section SHJ 1B (?); I) The yellow arrowhead points to vertical of Aloisathella sulcata (AITH), the calcification of which is yellowish-colored as the endostromatolitic crusts. Thin section SHJ 1B (?). All photos same scale bar = 500 µm.
**Figure 6:** Endostromatolites (red arrowheads) from Bukivna/Буківна, Ivano-Frankivsk Oblast (Ukraine), “Niżniower Kalkstein”, Tithonian-lower Berriasian. A) White arrowheads point to the micritic envelop of a former aragonitic gastropod shell. Thin section BR 3008. B) As in the previous picture, the micritic infill of the interparticular and moldic cavities postdates a phreatic cementation, that in turn postdates the dissolution of aragonitic shells. Thin section BR 3005. C) Detail of a colored fibrous sparitic crust growing in a cavity. Thin section BR 3008. D) Colored fibrous sparitic crusts in a cavity below a gastropod shell. Thin section BR 3010. E) Colored fibrous sparitic crusts growing in cavities. Thin section BR 3006. Red arrowheads point to endostromatolites (crusts); thick black arrowheads are upward-oriented; scale bars: white = 1 mm, black = 250 µm.


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