Microfacies and diagenetic evolution of the limestones of the upper part of the Crato Formation, Araripe Basin, northeastern Brazil

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Abstract
This paper presents the results of a petrographic and diagenetic study of the laminated limestones of the upper part of the Aptian to Albian Crato Formation, northeast of Brazil. The applied techniques were optical microscopy, cathodoluminescence and scanning electron microscopy (SEM) coupled to a wavelength-dispersive spectrometer (WDS). Petrographic analysis has revealed that most of the laminated limestones are calcilutites with a dominance of a micritic matrix, indicating a low-energy depositional environment. Microstructures such as microfaults, microfractures, microslumps, and loop bedding were observed. Based on textural, structural and paleontological features, seven microfacies were recognized: massive limestone, limestone with parallel laminations, limestone with undulated laminations, limestone with slumps, limestone with loop bedding, limestone with ostracods and limestone with peloids. In addition, the processes of cementation, dissolution, replacement, recrystallization and compaction, which are related to different diagenetic stages, were also recognized. The diagenetic constituents found in the sections include calcite, pyrite, silica and sulfates. We can conclude that a large part of the microstructures (microfaults, microfractures, microslumps and loop bedding) can be related to local seismicity, probably due to the reactivation of the Patos Shear Zone. The diagenetic constituents indicate an early to late diagenesis (eogenetic, mesogenetic and telogenetic stage).

KEYWORDS: Crato Formation; laminated limestones; diagenesis; Araripe Basin.

INTRODUCTION
Since the discovery of hydrocarbons in carbonate rocks of the so-called pre-salt layer in the marginal basins of Brazil, the interest increased in the search for rocky exposures as possible analogues of these reservoirs. The upper part of the Aptian to Albian Crato Formation, a well-exposed and preserved carbonate succession in the Araripe Basin (NE Brazil), is probably a good analogue. These carbonate rocks present features interpreted as algalic growth (Neumann 1999, Catto et al. 2016), similar to those occurring in the reservoir rocks of the pre-salt layer, as well as structures that can be interpreted only as event products of diagenesis (Neumann 1999). In this sense, based on petrographic, microfacies and microstructure analysis, an attempt is made, in this paper, to understand some of the issues related to the origin and diagenesis of these carbonate rocks.

The Araripe Basin is located in the interior of northeastern Brazil, occupying part of the states of Pernambuco, Ceará and Piauí, and covers an area of approximately 9,000 km², consisting of one of the most extensive interior basins in northeastern Brazil. It is located in the Piancó-Alto Brígida Terrain (Santos et al. 2004), in the western portion of the Transversal Subprovince of the Borborema Province. Its origin and evolution are related to the tectonic events of the Late Cretaceous, leading to the rupture of the Supercontinent Gondwana and the subsequent formation of the South Atlantic Ocean.

The structural configuration of this basin includes the tabular strata that formed the Araripe Plateau. This plateau is composed of the Barbalha, Crato, Ipubi and Romualdo formations and represents the post-rift sequences of the Araripe Basin (Assine 2007).

The Crato Formation represents the second lacustrine phase of the post-rift supersequence of the basin, which consists of six limestone units (informally called C1 to C6), according to Neumann (1999), deposited in the central and marginal zones of the basin forming carbonate bodies with different thicknesses and varied lateral extension. These six limestone units, 20 to 70 m in thickness, are in the lower part of the Santana Group and comprise two lithofacies, the rhythmite clay/limestone and the laminated limestone (Neumann 1999). These rocks are micritic and were deposited in internal lacustrine calm environments during the Aptian-Albian time-interval (Lima 1978, 1980, Hashimoto et al. 1987, Pons et al. 1990, Berthou et al. 1994, Coimbra et al. 2002, Batten 2007).
The carbonate section of the Crato Formation in the Araripe Basin has invariably been considered to be lacustrine (Neumann 1999, Neumann & Cabrera 1999), except for a single reference that reports the existence of marine forms of foraminifera (Arai 2012). Therefore, it is interpreted that, due to its geographic position in the continent, the marine entry in the Araripe Basin occurred with relative delay in relation to the marginal basins.

The laminated limestones are intercalated with a series of clays, siltstones and sandstones. According to Martill & Heimhofer (2007), the Crato Formation can be divided into four different members, including from the base to the top the Nova Olinda, Caldas, Jamacaru and Casa de Pedra members, but this subdivision was not adopted in this work.

The objective of this paper is to characterize the laminated limestones in the upper part (unit C6) of the Crato Formation in sedimentological, petrographic, microfaciologic, microstructural and diagenetic terms.

GEOLOGICAL SETTING

The Araripe Basin is part of the Transversal Subprovince of the Borborema Province, located between the Pernambuco and Patos Shear Zones.

The structural configuration of this basin covers the tabular strata that formed the Araripe Plateau. This E-W-elongated plateau is an outstanding geomorphic feature of northeast Brazil. It consists of sedimentary units of the post-rift sequence of the Araripe Basin, unconformably overlying older units or directly in angular unconformity upon the crystalline basement, a common configuration in the western portion of the basin, as shown in Figure 1.

According to Ponte and Appi (1990), Assine (1990, 1992) and Ponte and Ponte Filho (1996), the Araripe Basin can be subdivided into sequences linked by regional unconformities, reflecting distinct tectonic phases in the basin. Assine (2007) integrated these different proposals, identifying four large units bounded by unconformities:

• a Paleozoic sequence represented by the alluvial deposits of the Cariri Formation and characterized by medium to coarse grained sandstones of Silurian-Devonian age (Beurlen 1962, Assine 1992, 2007, Arai 2006), which is interpreted as the residual deposits of a large intracratonic basin;
• the Upper Jurassic pre-rift supersequence that corresponds to the Brejo Santo (predominantly pelites, red beds) and Missão Velha (coarse- to fine-grained sandstones and conglomeratic sandstones) formations;
• the rift supersequence represented by the Lower Cretaceous Abaia Formation formed by a succession of laterally discontinuous sandstones intercalated with calciferous shales of variegated coloration (Fambrini et al. 2012);
• the post-rift supersequence, subdivided into two sequences, the post-rift I sequence of Aptian-Albian age, corresponding to the Barbalha Formation and the Santana Group (Crato, Ipubi and Romualdo formations) according to Assine et al. (2014), and the post-rift II sequence of Albian-Cenomanian age, which is characterized by alluvial sediments of the Araripina and Exu formations, indicative of tectonic reactivation in this time interval.

RESULTS

Macroscopic analysis

The studied outcrops are located in the northern part of the Araripe basin, in the upper part of the Crato Formation. Macroscopically, these limestones are fine-grained, forming horizontal tabular layers that are locally intercalated with terrigenous rocks, such as shale and claystone (Figs. 2A and 2B). Limestones are mainly laminated with alternating light and dark laminae, of beige and brown color, respectively, sometimes being bluish. However, they have also been observed in massive and stratified forms (Fig. 2C).

Regarding the structural aspects, it has been observed that these rocks are intensely fractured and cemented by calcite and silica in some outcrops, but some unfilled fractures also occur. In addition to several shear fractures, they displace the laminations in the laminated limestones. Microfaults have also been observed, and they are mostly of the normal type.

MATERIALS AND METHODS

The classification of the limestone and the interpretation of the microfacies, the diagenetic evolution and the depositional environments are based on field observations (eight outcrops), samples and thin sections. Lithologic, sedimentological and structural descriptions were made. Samples were collected to prepare thin sections for subsequent petrographic study under optical microscope, cathodoluminescence and scanning electron microscopy (SEM) coupled to a wavelength-dispersive spectrometer (WDS).

The laboratory analysis is comprised of petrographic descriptions of 44 thin sections using a model BX-41 Olympus petrographic microscope at the Department of Geology, at Universidade Federal de Pernambuco (UFPE). At this stage, it was possible to describe the textural and structural features, as well as to identify the mineralogical (cement, grains and matrix) and paleontological compositions. Some types of porosity and diagenetic features were also identified.

Cathodoluminescence analysis was used in the study of diagenetic processes, to highlight the presence of fractures and the evolution of porosity in rocks and to recognize the different generations of cement. The study was conducted using a Cambridge Image Technology Ltd. (CITL) CL8200 model coupled to an optical microscope.

A JEOL KAL 6460 SEM with a wavelength-dispersive spectrometer from the Nanostructure Laboratory (LDN) was also used to aid in the identification and semiqualitative analysis of chemical elements.

The integrated analysis allowed the classification of the limestones in the upper part of the Crato Formation using the classification of Grabau (1904) and Dunham (1962). In addition, it was possible to characterize the seven microfacies of these laminated limestones based on textural and microstructural features and fossil contents, and to define the diagenetic evolution of these rocks.
Locally, calcite veins with cone-in-cone structures (Fig. 2D) have been observed as evidence of the recrystallization process. Salt pseudomorphs, such as hopper-faced halite, have also been seen (Fig. 3A).

It was possible to verify the presence of micropores caused by dissolution. Some of these pores are cemented by recrystallized calcite or quartz, and geode structures formed by calcite may also occur.

In some outcrops, where the surface of the upper part of the C6 level can be observed, it was possible to note that the limestone laminae showed undulations (Fig. 3B) and a botryoidal surface characterized by an irregular and wavy feature which is associated with pedogenetic or diagenetic processes. In addition, a silicification process was perceived in these portions (Fig. 3C) in the macroscopic analysis and confirmed by microscopy.

Locally, a collapse-breccia with various fragments of laminated limestone cemented by carbonatic and siliceous matrix was observed (Fig. 3D).

Microfacies of level C6 of the Crato Formation

Only the lithofacies of the laminated limestone was observed, since it refers to the upper part of the Crato Formation and, in this case, the C6 level proposed by Neumann (1999). This is the main and most representative carbonate lithofacies occurring in the Crato Formation. It is represented in this work by beige to brown and sometimes gray to blue laminated limestone, with parallel laminae, locally undulated and with the presence of sliding structures (microslumps), microfaults, microfractures, loop bedding, ostracods and peloids.

The microscopic characteristics allowed us to characterize seven microfacies (m1 to m7) according to texture, structures and bioclastic content: massive limestone, limestone with parallel laminations, limestone with undulated laminations, limestone with slumps, limestone with loop bedding, limestone with ostracods and limestone with peloids (Tab. 1).

**Massive limestone microfacies (m1)**

The massive limestone microfacies (m1) consists of a carbonate mud composed of micritic calcite with colors ranging from beige to light brown, usually without visible lamination under the microscope, and contains, locally, frambooidal pyrite and iron oxide stains (Fig. 4). Tectonic features can also be observed, such as microfractures filled/cemented by calcite and/or silica. Primary or secondary porosities are typically absent in this microfacies.

**Limestone with parallel laminations microfacies (m2)**

This is the microfacies with the largest occurrence in these limestones. It is represented by limestone with millimeter-thick parallel laminae, organic matter, opaque minerals (frambooidal pyrite), and iron oxide resulting from the alteration of these pyrites. In addition, several filled or unfilled microfractures were observed. The beige laminae are composed of micritic calcite, whereas the dark one is more often defined by the presence of opaque minerals, iron oxide and organic matter (Fig. 5). Organic matter, silica and calcite were also observed filling the microfractures.
Limestone with undulated laminations microfacies (m3)

This microfacies was found in only two outcrops located at the contact of the Crato and Ipubi formations. It is characterized by crenulated organic lamellae, which in most cases are wavy and deformed (Fig. 6). Framboidal pyrite, iron oxide and various fractures filled with fibrous calcite were also observed. In addition, silica (cryptocrystalline quartz, megaquartz and chalcedony) and sulfates (gypsum, anhydrite and barite) were found to occur as substitutions.

Figure 2. (A) Outcrop (Aurélio’s Quarry) with laminated limestone intercalated with claystone; (B) laminated limestone intercalated with shale in Três Irmãos Quarry; (C) fractured stratified limestone in outcrop located in Abaiara; (D) calcite vein with a cone-in-cone structure.
**Limestone with slumps microfacies (m4)**
This microfacies presents the same characteristics of the m2 microfacies, but with smooth laminations and the presence of sliding features or microslumps in soft-sediment (Fig. 7).

**Limestone with loop bedding microfacies (m5)**
In this microfacies, laminated limestone is produced with microstructures called loop bedding, consisting of "small groups of laminae that are sharply constricted or that terminate at intervals, giving the effect of long, thin..."
loops or links of a chain” (Bates & Jackson 1980), similar to sedimentary boudinage. The loop bedding can be classified as simple or complex. However, here, we have observed only simple loop bedding, generated diagenetically by overload (Fig. 8).

**Limestone with ostracods microfacies (m6)**

This microfacies was found in both laminated and massive limestones. Bioclasts identified as ostracods have been observed, the interior of which one filled by micritic or sparry calcite or by pyrite, which mostly occurs with whole and articulated valves that suggest low energy sedimentation, but locally they have compaction features, with disarticulated and flattened valves. These ostracods are replaced by micritic and/or sparry calcite (Figs. 9A and 9B) and by pyrite (Figs. 9C and 9D).

**Limestone with peloids microfacies (m7)**

This microfacies is poorly represented in the thin sections studied. A level of millimetric peloids having circular and ellipsoidal forms was observed without internal structure and composed of micrite and distributed in a micritic matrix. The origin of these peloids may relate to the alteration of bioclasts (Fig. 10).

**Diagenetic processes**

Diagenesis typically involves a variety of physical and chemical processes. The most common and identified here are dissolution, cementation, replacement, recrystallization and compaction.

The dissolution process is represented by secondary porosities (Fig. 11A), which are represented by the vuggy and fenestral types of porosities and are classified according to Choquette and Pray (1970).

With respect to cementation (Fig. 11B), calcite cements were found to occur in sparry, prismatic and fibrous forms (Fig. 12A and 12B), in addition to silica, both of which filled fractures.

In these limestones in the upper part of the Crato Formation, several mineral phases were observed, which replaced these carbonates. These substitutions were then referred to as pyritization, silicification and sulfation.

The pyritization is characterized by the most common iron sulfide (FeS₂) found in carbonate rocks, whose presence is very often found in all the microfacies defined herein. It is represented by microcrystalline and framboidal pyrite, which usually forms anhedral replacement masses. It is produced in a fairly distributed manner in the rocks and locally replaces organic matter and some bioclasts, in this case, ostracods (Fig. 12C).

![Figure 4. Massive limestone microfacies: (A) and (B) carbonate mud with diagonal fractures filled by calcite; (C) carbonate mud with horizontal fracture; (D) carbonate mud with framboidal pyrite. Photomicrographs: parallel nicols.](image-url)
The silicification process is characterized by silica (SiO$_2$), a diagenetic mineral phase that is very common in carbonate rocks and can occur as cement or as a replacement of the original material. Although there are several diagenetic species of silica, only cryptocrystalline quartz (chert), megaquartz and chalcedony (fibrous and radial forms and spherulitic structures) were observed (Fig. 12D).

Other minerals that are present in the contact between the Crato and Ipubi formations are sulfates (Figs. 12E and 12F), thus giving origin to the sulfation process, which is represented by the occurrence of gypsite, barite (Fig. 13) and anhydrite.

The recrystallization process was observed locally in these limestones, where micritic calcite change occurs, thereby generating sparry calcite. These calcites occur as fracture fill.

Finally, compaction was observed, exhibiting only mechanical compaction (Bathurst 1986). It is defined by deformation features in the clay-organic laminations, in addition to the presence of some displaced and sometimes flattened valves of ostracods.

DISCUSSION

The macro- and microscopic analysis in this research confirmed the sedimentological and structural aspects previously described by Neumann (1999) and Silva (2003) as the dominance of micritic calcite, microfaults, microslumps and loop bedding structures.

In general, these lacustrine limestones of Aptian-Albian age were classified as calcilutites, due to granulation, according to the classification of Grabau (1904). As it was observed, these rocks are composed of a micritic matrix with less than 10% grains and can be classified as mudstones according to Dunham (1962).

High luminescence in these lacustrine limestones was observed in all thin sections examined using cathodoluminescence analysis. This high luminescence could be associated with the relatively high Mn/Fe ratio, typically obtained under reducing conditions during the early stage of burial diagenesis.

According to Heimhofer et al. (2010), the origin of the Crato Formation carbonates is traditionally attributed to the chemical precipitation associated with clastic fine-grained sediments and is unaffected by any organic organism. Catto (2015) observed the presence of the organic matrix extracellular polymeric substance (EPS) in the laminated limestones of the Crato Formation and used this substance as a diagnostic criterion for the biotic influence on the precipitation of carbonate minerals. The last author has identified calcified organisms (filamentous and coccus bacteria, and cyanobacteria) and suggested

![Figure 5. Limestone of the parallel laminations microfacies: (A) and (B) the dark bands are composed of microcrystalline calcite, organic matter and pyrite, while the light bands are constituted of microcrystalline calcite without pyrite. Evidence of recrystallization of calcite in the lower portion (lighter color) is seen in C. The dark stain in the lower part of D refers to an arboreal manganese. Photomicrographs: parallel nicols.](image-url)
that the Crato Formation carbonates come from biologically
induced precipitations.

In this study, some laminations similar to algae mats have
been observed, as shown in the m3 microfacies. Therefore,
the origin of these limestones could be bio-induced by algae
growth, as observed in Catto (2015), and related to chemical
precipitation (Heimhofer et al. 2010).

The organic laminations observed in the m3 microfacies
are compound algae mats that represent a hypersaline environ-
ment, whose observed deformation has been related to a desic-
cation and compaction process acting on the lacustrine system.

The greatest importance of these carbonate rocks is their
potential for the reservoir rocks, and here it has been observed
that the outcrops of the carbonate rocks in the upper part of the

![Figure 6. Limestone with undulated laminations microfacies: limestone with organic laminae, brownish-colored, crenulated and deformed with recrystallized calcite. Photomicrographs: (A) parallel nicols, (B) crossed nicols, (C) and (D) parallel nicols.](image1)

![Figure 7. Limestone with slumps microfacies: microslumps located in the center of the illustration are highlighted by a dashed line. Photomicrograph: parallel nicols.](image2)

![Figure 8. Limestone with loop bedding microfacies: laminated limestone with simple loop bedding highlighted by hatching contour. Photomicrograph: parallel nicols.](image3)
The Crato Formation are quite fractured. However, a large part of these fractures are sealed. Hence, in addition to the fractures, the vuggy and fenestral porosities are also mostly cemented. Therefore, in this work, with respect to the potential of the reservoir rocks, the lithotypes of the upper part of the Crato Formation were characterized by a low grade of permeability and, thus, a low reservoir potential.

According to Miranda (2015), based on the geological and perm-porosity characteristics obtained for the laminated limestones in the upper part of the Crato Formation, it has been possible to classify them as an analogue of naturally fractured type 4 (Nelson 1987, 2001), in which the matrix has a medium to high primary porosity and low permeability, and the fractures are cemented. Thus, it could be classified as an unconventional reservoir of “tight” type (when porosity is associated with the microporosity). In general, Miranda (2015) considers these limestones to have a medium primary porosity and very low permeability, as well as to contain a series of fractures cemented by calcite that form a hydraulic barrier, as observed in this study.

The structural features observed in these laminated limestones are microslumps, loop bedding, microfractures and microfaults (Neumann 1999), which may be related to local seismicity occurring in the region and probably related to the reactivation of the Patos Shear Zone, in the North of the Araripe Basin. This fact was also noted by Silva (2003), who recognized three deformational stages (D1, D2 and D3) in this unit. These microstructures were generated during the D1 event, brittle-ductile extensional, located in some thin limestone levels and are the result of small seismic pulses. Silva (2003) suggested that these structures occur in a few levels of the carbonate units, which may indicate sporadic events of seismic pulses over long time intervals, since some levels do not exhibit such characteristics.

Diagenesis was subdivided into three main stages: eodiagenetic, mesogenetic and telodiagenetic. Therefore, with the study of diagenetic processes, it was possible to determine the stage at which each event occurred.

Typical eodiagenetic features were observed, such as intergranular porosity in the micritic matrix and the presence of microcrystalline and frambooidal pyrite (FeS2). Additionally, at this stage, the presence of sulfate minerals that form closer contact with the Ipubi Formation was observed.

A dissolution process was noticed that gave rise to a vuggy and fenestral secondary porosities that represents the mesogenetic stage. Additionally, cementation by silica and calcite was verified and also a process of silicification, which occurs in form as cryptocrystalline quartz, megaquartz and chalcedony.
At this stage, the recrystallization process was characterized by microfractures that are often sealed by blocky or fibrous calcite cement.

The telodiagenetic stage is represented by the presence of iron oxide, which were fairly observed in these limestones and are associated with the alteration of pyrite by oxidation processes.

According to Boggs Jr. (2009), the origin of silica can be essentially a product of dissolution by meteoric waters (weathering), dissolution of the skeletons of organisms, dissolution of quartz grains, pressure solution of quartz grains and release of silica by reactions between minerals, as occurs with clay minerals. In the case of the Crato Formation limestones, the origin is mainly related to meteoric water saturated in SiO₂, which, due...
to the increase in pressure (P) and the temperature (T) of the dissolved silica, precipitates later in the pores.

According to Martill et al. (2007), the existence of halite pseudomorphs in these limestones indicates that the basin experienced aridity conditions, where hypersalinity prevailed in the deposition period of the laminated limestones. These authors have recognized five types of pseudomorph shapes of halite. Here, only the so-called type 1 is observed, which occurs as a kind of cross and, occasionally, star, in which a series of concentric circles appear in layers of laminated limestone.

Sulfate minerals, as well as evaporites, are generally formed in environments with a low supply of terrigenous material. They begin their precipitation in a dry climate with increased evaporation due to a decrease in the lake level that makes the waters more concentrated and allows the formation of brines.

Evaporites are deposits from arid environments, and, in the case of the Araripe Basin, they are representatives of the Ipubi Formation. Evaporative solutions are highly mobile due to their low density. Evaporites can migrate to adjacent or underlying strata and precipitate as diagenetic sulfates (usually as crystals

Figure 12. (A) Fibrous calcite at parallel nicols; (B) the same image as A where the fibrous calcite shows high luminescence under cathodoluminescence analysis; (C) pyrite occurring in a widespread form, replacing bioclasts (parallel nicols); (D) chalcedony spherulite replacing the micritic matrix (crossed nicols); (E) anhydrite in the upper portion of the image and barite in the lower right portion (crossed nicols); (F) gypsum with gray coloration and anhydrite colored with high birefringence (crossed nicols).
and nodules or as carbonate replacements) in units that may be unrelated to arid environments, such as events occurring in the upper part of the Crato Formation.

CONCLUSIONS

The laminated limestones in the upper part of the Crato Formation consist mainly of calcilutite with a micritic matrix, frambooidal pyrite and iron oxide. The observed paleontological content was ostracods and peloids.

These limestones have secondary porosity. However, it has been found that these porosities are locally filled due to diagenetic processes. Hence, these rocks become less porous.

The processes identified from different diagenetic stages that affect these laminated limestones are cementation, dissolution, replacement, recrystallization and compaction. Additionally, the diagenetic constituents found in the thin sections include calcite, pyrite, silica and sulfates.

From the obtained data, it can be concluded that a large part of the microstructures may be related to local seismicity, probably related to the reactivation of the Patos Shear Zone. However, they were also generated diagenetically by overload.

In general, these limestones in the upper part of the Crato Formation represent a gradual growth in an arid and salty depositional environment, where silica and sulfates were observed in the upper part in contact with the Ipubi Formation. In addition, algae mats were noted, and the presence of which is also linked to a high salinity environment.

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