

## Diagenetic Fabrics of Aragonite, Calcite and Native Sulfur in the Miocene Evaporite Cycle on the Red Sea Coast, Egypt

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**ABSTRACT.** The fabrics and presence of celestite in the carbonate cements in the reefal limestone of Gebel El Rusas Formation and the sabkha sequences of the Abu Dabbab Formation (Middle Miocene) suggest deposition in a hypersaline vadose regime. The recorded carbonate cements are represented by paleoflowstone crusts; spheroidal and hemispheroidal structures and giant paleoaragonite ray - crystals deposited in fenestrae and as crusts within the Miocene sediments.

The carbonate cements occur in the form of polycrystalline calcite pseudospar and fibrous calcite resulting formed as paramorphic replacements of aragonite in a Mg poor fresh water vadose regime. Traces of native sulfur occur together with secondary gypsum and calcite in the surficial layer of the Abu Dabbab evaporites. The sulfur is formed in a localised microenvironment in the fresh water vadose regime and is a response to bacterial activity on sulfate deposits.

This paper describes the petrology, fabrics and the paragenetic relationships between the carbonate cements as well as the native sulfur, gypsum and carbonates in the Miocene evaporite cycle of the Red Sea margin between Quseir and Mersa Alam (Fig. 1). The carbonate cements occur within the fenestrae and as crusts. They are represented by polycrystalline calcite pseudospar (a term used by Mazzullo 1980, to refer to those replacement crystals which are of paramorphic rather than pore filling origin), fibrous calcite, and microcrystalline calcite forming paleoflowstone (a term used by Assereto and Folk 1980, to refer to festooned cellular crusts which lines fissures of tepee structures), spheroidal and hemispheroidal structures. El Aref *et al.* (1985) considered these structures as caliche deposits. Giant paleoaragonite ray - crystals composed of polycrystalline calcite pseudospar are reported in some fenestrae and as crusts within the sabkha sequence of Abu Dabbab Formation. Traces of native sulfur associated with

secondary gypsum and calcite are also recorded in the surficial layer of the Abu Dabbab evaporites.

### Methods of Study

The structural and textural characteristics of the carbonates were studied in the field and petrographically examined. The mineralogical composition was identified using X-ray diffractometer with Cu K alpha radiation (45 kV, 60 mA). The mineralogical analysis was carried out at the Institute of Geology, University of Bergen.

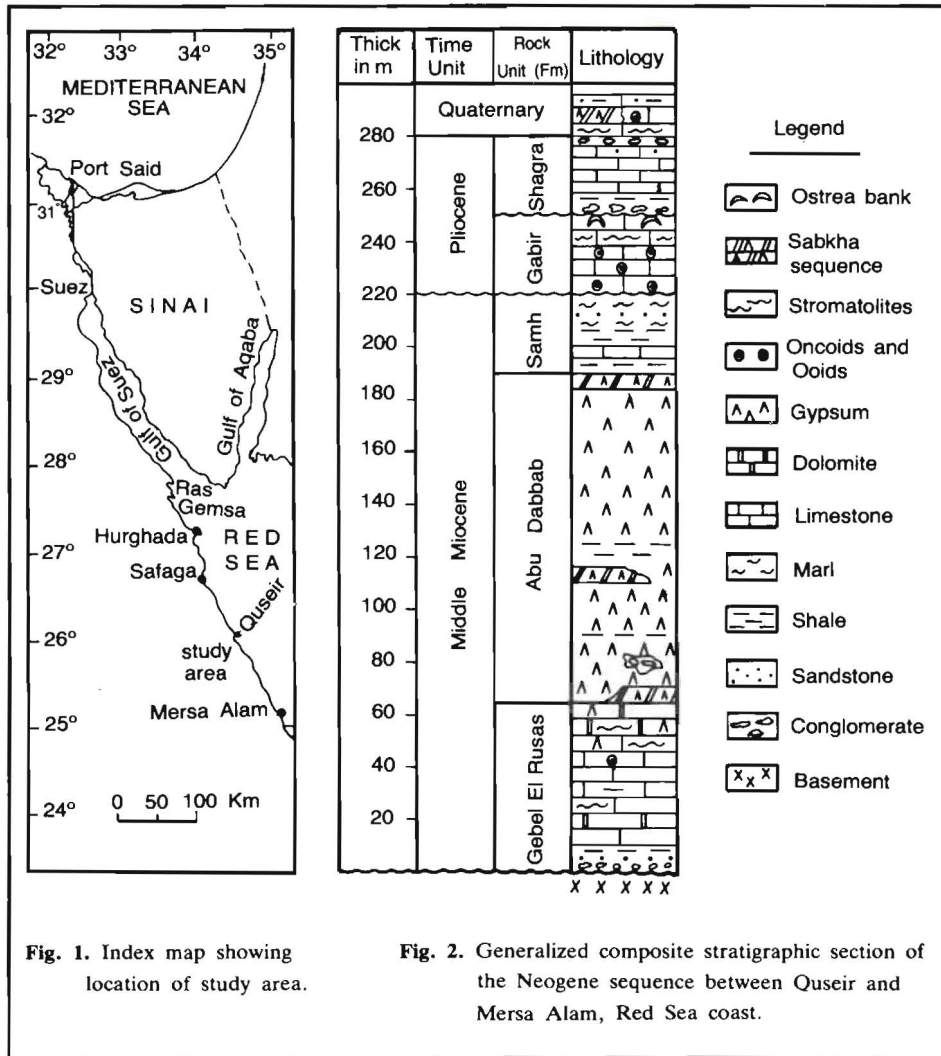
### Geologic Setting

In the study area, the Miocene sequence consists of an evaporite cycle which begins with the Gebel El Rusas Formation followed stratigraphically upward by the Abu Dabbab Formation and then by the Samh Formation (Fig. 2). These three formations are of Middle Miocene age (Said 1962, El Akkad and Dardir 1966 and others). The unconformably overlie the basement rocks and underlie the oolitic, oncolitic and stromatolitic carbonates of the Gabir Formation (Pliocene age).

The Gebel El Rusas Formation (about 50 m thick) includes from bottom to top; 1) alluvial fanconglomerate, which laterally changes into shallow intertidal cross bedded sandstone, at the base, 2) back reef facies composed of gypsiferous green shale and argillaceous biomicrite rich in miliolids and molluscs with occasional thin beds of microcrystalline dolomite, and 3) reefal facies composed of coral - algal biolithite with abundant oncolites, molluscs, echinoderms, and sometimes evaporite minerals (Youssef and Abou Khadrah 1984).

The Abu Dabbab Formation consists of about 120 m of sabkha and subaqueous evaporite sequences intercalated with shale, marl and dolomite layers. The sabkha sequence is represented by small mounds which stratigraphically overlies the Gebel El Rusas Formation as well as by thin layers overlying the subaqueous evaporite sequence (Youssef 1986). They are composed of 2-5 m of fenestral stromatolitic and nodular dolomite and sometimes nodular anhydrite. However, the subaqueous evaporite sequence is composed of about 100 m of nodular mosaic and massive crystalline gypsum covered by a surface layer of nodular white, dense powdery anhydrite.

The Samh Formation consists of about 60 m of gypsiferous olive green calcareous shale with microcrystalline limestone intercalations at the base, overlain



by red and green marls with feldspathic sandstone at the top. The facies of this unit suggest deposition in a tidal flat setting supplied with fine clastic sediments from drainage channels cross up the western basement rocks (Youssef 1986).

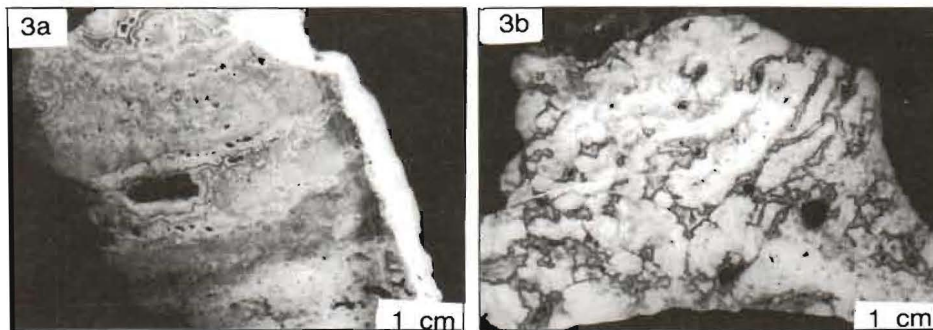
### Fabrics and Petrology

The studied sediments are characterized by fenestral cavities and solution veins. The fenestrae are either horizontally subparallel arranged voids or irregular

cavities randomly oriented and distributed throughout the rock (Fig. 3a). However, the solution veins are irregular in shape, range from 1 to 3 mm in width and filled by brownish red micrite (Fig. 3b). The fenestrae are mostly filled with festooned cellular crusts (paleoflowstone), spheroidal and hemispheroidal structures, and giant paleoaronite ray - crystals. Some fractures and cavities, produced by dissolution of former evaporite nodules, are filled with native sulfur, calcite and secondary gypsum.

### *Paleoflowstone*

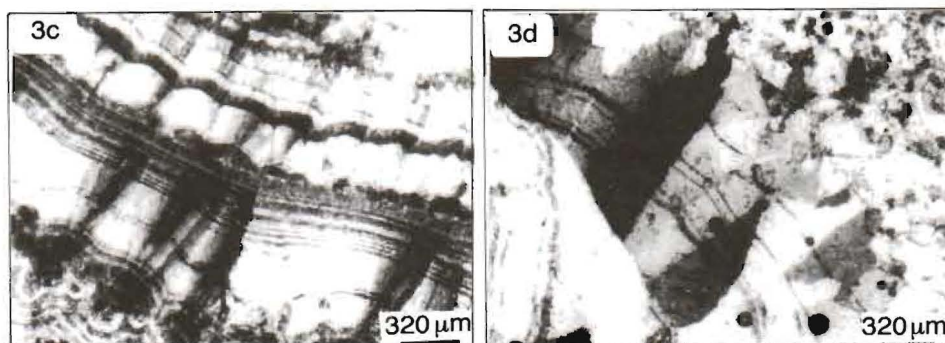
Some fenestrae in the studied sediments are filled with curved and wavy discontinuous carbonate crusts ranging in length between 1 and 6 cm and in thickness between 0.1 and 0.2 cm (Fig. 3a). They are composed of light fibrous calcite laminae which are separated by thinner dark micrite laminae (Fig. 3c). The fibrous calcite is similar to the aragonite cement seen in Recent hypersaline environment (Purser and Loreau 1973). It was previously defined as fascicular optic calcite by Kendall (1977). The fibrous calcite does not show uniform extinction, each crystal has its inherited composite extinction pattern which is different from that of the neighboring crystals. Some of these fibrous calcite crystals contain minute inclusions concentrated in a radiating form and parallel to the substrate. Sometimes, although the fibrous aragonite is replaced by sparry calcite (calcite pseudospar), the fibrous texture is observed in it (Fig. 3d). The crystallographic orientation of the fibrous calcite and calcite pseudospar is generally cut across the inclusions.



**Fig. 3a.** Fenestral stromatolitic carbonates with paleoflowstone crusts (about 2 cm thick) showing scalloped laminations. The light laminae are fibrous calcite whereas the dark laminae are micrite.

**Fig. 3b.** Solution veins (about 3 mm wide) filled with micrite.





**Fig. 3c.** Thin section in paleoflowstone crust showing that the fibrous calcite crystals are arranged perpendicular to the micrite laminae. Crossed Nicols (C.N.).

**Fig. 3d.** Paleoflowstone crust composed of calcite pseudospar crystals cutting across the micrite laminae and inclusions. C.N.

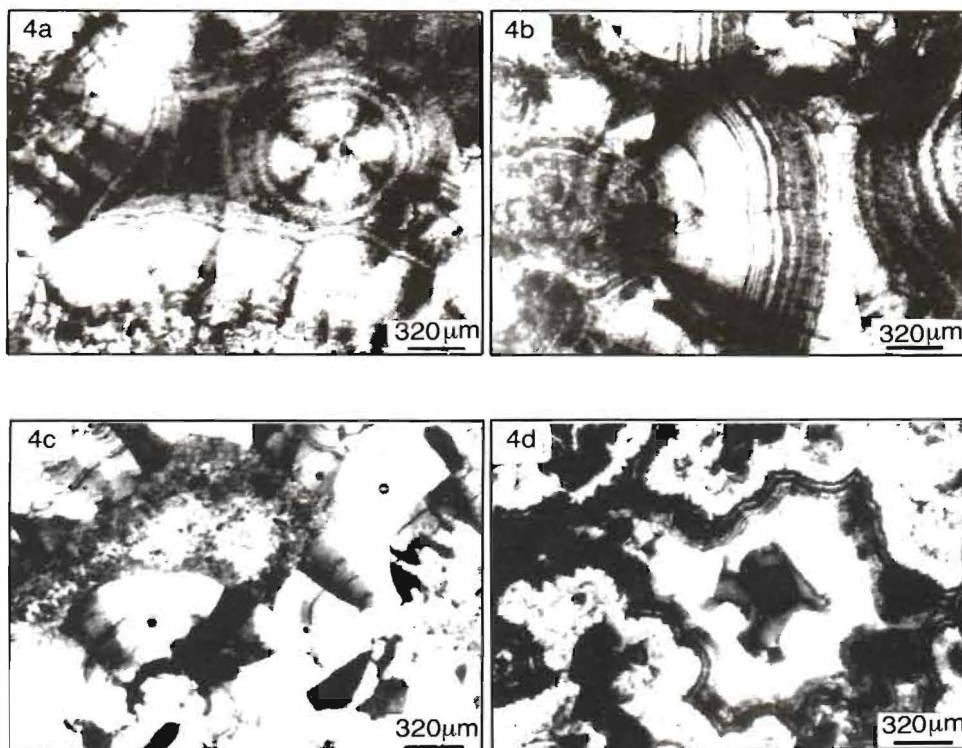
#### *Spheroidal and hemispheroidal structures*

Spheroidal and hemispheroidal structures ranging in diameter between 1 to 4 mm are generally exist in the paleoflowstone crusts (Figs. 4a and 4b). They are composed of nuclei surrounded by envelopes of thick light fibrous calcite laminae alternating with thinner dark micrite laminae. Some envelopes are composed of calcite pseudospar (Fig. 4c), in which the calcite crystals grow perpendicularly to the surface of the nucleus. The nuclei are mostly composed of fibrous calcite balls (Fig. 4a), algal nodules (Fig. 4b), or mosaic sparry calcite (Fig. 4c). Some other nuclei are composed of silica or evaporites. The hemispheroids may coalesce to form continuous crust in the paleoflowstone. The coalescing crust generally show either smooth flat or a mammilated form representing a section in a spherulitic mass (Fig. 4d).

#### *Giant paleoaronite ray - crystals*

Parallel and/or radial paleoaronite ray - crystals are generally form crusts in the sabkha sequence. The elongation direction of these crystals diverges away from the substrate in a manner similar to Holocene aragonite fan druses (Fig. 5a). Other giant paleoaronite crystals either line walls of cavities in the reefal limestone or grow downward (Fig. 5b) to form gravitational cement (stalactites) as well as on the floor (stalagmites). The crystals have sharply defined straight edges and square ends. They range in length between 5 and 16 cm and width from 1 to 4 mm (Figs. 5a and 5b) Some crystals behave optically as a single crystal, although they represent a group of mutually parallel crystals. Other crystals internally consist of a mosaic of

optically unoriented calcite pseudospar crystals with irregular interlocking boundaries (Fig. 5c). In most cases the calcite pseudospar tends to show long straight boundaries parallel to the ray sides. The pseudospar crystals range in length between 0.1 and 2 mm and sometimes approach 10 mm in length. They clearly appear as if controlled by the original aragonite structure. When the ray crystals are seen in transverse section, they show rhombic form as well as quadrant shape (Fig. 5d), probably of the carbonate (aragonite).

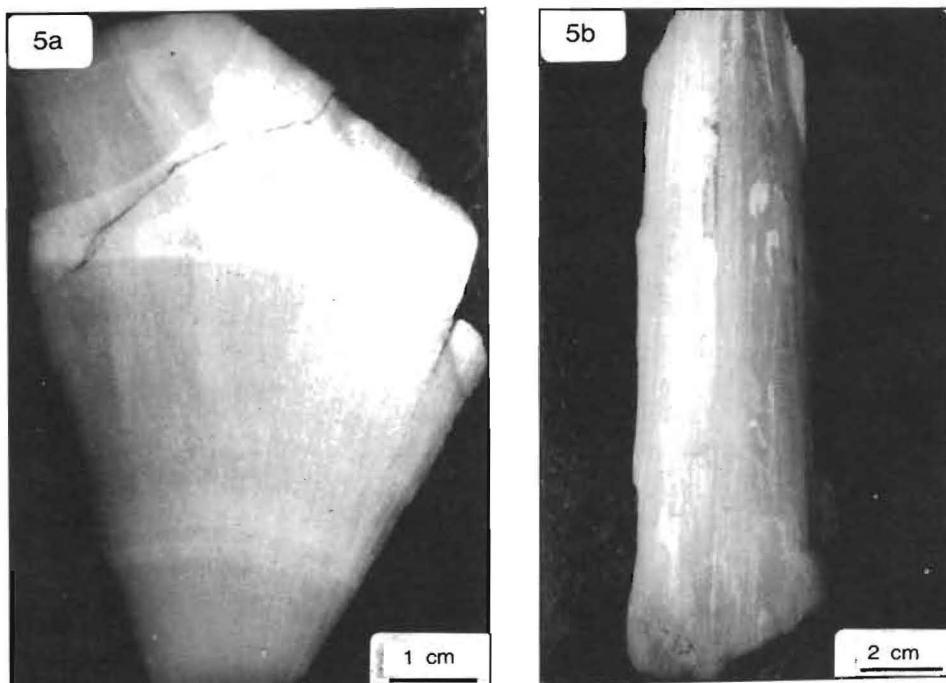


**Fig. 4a.** Spheroidal structure composed of fibrous calcite ball nucleus surrounded by fibrous calcite and micrite laminae. C.N.

**Fig. 4b.** Hemispheroidal structure composed of algal nodule nucleus surrounded by fibrous calcite and micrite laminae. C.N.

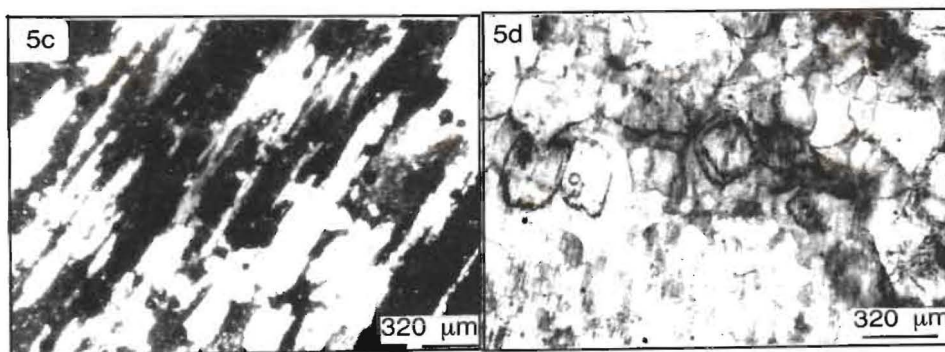
**Fig. 4c.** Spheroidal and hemispheroidal structures composed of spar-filled cavities as nuclei, surrounded by calcite pseudospar. C.N.

**Fig. 4d.** Coalesced hemispheroidal structures showing mammillated form which reflect section through spherulitic masses. C.N.



**Fig. 5a.** Radial paleoaragonite crystals arranged vertically from substrate in the sabkha sequence.

**Fig. 5b.** Paleoaragonite crystals grow downward in cavities of the reefal limestone.



**Fig. 5c.** Paleoaragonite crystals formed of mosaic of optically unoriented calcite pseudospar crystals. C.N.

**Fig. 5d.** Thin section perpendicular to the elongation of calcite pseudospar crystals showing rhombic shape of the carbonate crystals. C.N.



### *Native Sulfur*

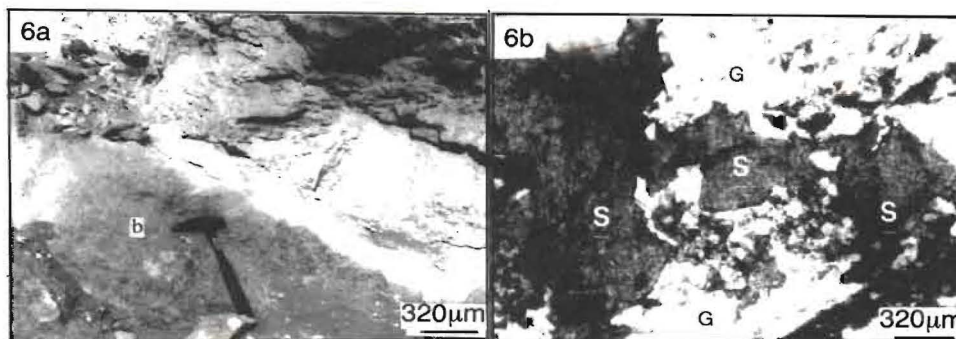
The origin of native sulfur in the Miocene evaporite sequence on the Red Sea coast, Egypt, attracted the attention of many authors. Shukri and Nakhla (1956) concluded that the sulfur ore at Ras Gemsa is of epigenetic origin. They stated that the sulfur was brought up from a lower source, most probably dissolved in hydrocarbons along fault planes. They added that the sulfur in anhydrite was formed by reduction. Roufaiel and Samuel (1975) concluded that the sulfur at Range area, Red Sea coast was formed by an epigenetic process of reduction of the enclosing gypsum - anhydrite by low temperature hydrothermal solutions containing appreciable amounts of Pb, Sr and Ba. El Aref (1984) concluded that the sulfur at Ranga area is of syndiagenetic origin, deposited with their gangue associations by generations of fractional crystallization in shallow marine environments. Youssef (1988) concluded that the sulfur ore at Ras Gemsa area was formed as the result of bacterial activity on the Miocene sulfate deposits in the presence of organic matter (hydrocarbon derived from the lower reefal limestone of the Miocene age) which acted as nutrient for the activity of the sulfate reducing bacteria.

In the studied sequence, the native sulfur is associated with the surficial layer at the contact between the secondary gypsum and the white dense powdery anhydrite forming a crustal layer on the evaporite sequence of Abu Dabbab Formation. It is restricted to the channels of circulating fresh water (Fig. 6a) and is associated with white dense calcite and carbonaceous material. It may also associated with laminated evaporite probably of sabkha origin. Sulfur occurs in three main forms: a) small lenses of about 10 cm in length, b) fracture fillings, and c) disseminated granules (Fig. 6b). It is associated with granotopic, alabastrine, porphyrotopic and satinspar secondary gypsum. The secondary gypsum represent the late stage of transformation of anhydrite to gypsum by circulating fresh water (Youssef 1986). The existence of sulfur in the surficial layer (fresh water vadose regime) and its association with the secondary gypsum suggest that it was formed in a localized reducing microenvironment which is different in geochemical character from the surrounding fresh water vadose oxidizing setting.

### **Diagenetic Models**

The fenestrae in the studied sediments were probably originated due to gas generation or as contraction fractures formed during compaction and drying of the sediments in the early stages of diagenesis (Shinn 1968 and 1983, and Kendall 1969). Flooding of these sediments by hypersaline marine water was resulted in the deposition of flowstone, spheroids, hemispheroids, and giant aragonite ray - crystals in the fenestrae and as crusts in the sabkha sequence.





**Fig. 6a.** Channel of ground water composed of secondary gypsum, anhydrite, calcite and traces of sulfur granules. b = bottom of channel.

**Fig. 6b.** Disseminated sulfur granules in the secondary gypsum. S = sulfur, G = gypsum. Plane Polarized Light.

Similar paleoflowstone were reported in Permian limestones of New Mexico ("stromatactis" of Otte and Parks 1963), in Triassic rocks from Austria (Zankl 1971) and from Jurassic tepee structures in Morocco (Burri *et al.* 1973) and were interpreted as organic crusts. On the contrary, Assereto and Folk (1980) interpreted the festooned cellular crusts (paleoflowstone) and the hemispheroids from the Triassic Calcare Rosso, Italy as inorganic structures formed penecontemporaneously within the fissures of the tepee in deformed supratidal sediments when flooded by marine brines. Spheroidal structures similar to the studied spheroids were also described from the hypersaline supratidal environment in the Arabian (Persian) Gulf (Purser and Loreau 1973, Evamy 1973, Shinn 1973, and Picha 1978), and from the lower Carboniferous Llanelly Formation of South Wales (Wright 1981).

#### *Aragonite - calcite relations*

The studied paleoflowstone, spheroids and hemispheroids are composed of light thick fibrous calcite laminae alternated with thin dark micrite laminae, or of calcite pseudospar, whereas, the paleoaragonite ray - crystals are composed of calcite pseudospar. Similar fibrous calcite with inherited composite extinction pattern has been frequently considered to represent replacement of acicular marine aragonite or high magnesium calcite cements without the formation of significant solution voids (Kendall and Tucker 1973, Kendall 1977, Davis 1977, Assereto and Folk 1976, Mazzullo 1980 and others). The radial and horizontal inclusions in the fibrous calcite probably reflect growth bands (Kendall and Tucker 1973, and Kendall 1977).

In agreement with Scholle and Kinsman (1974), Mazzullo (1980), and Assereto and Folk (1980), the studied fibrous calcite and calcite pseudospar of the paleoflowstone, spheroids, hemispheroids, and the giant ray-crystals were deposited as aragonite in a hypersaline vadose regime. Later flushing by Mg-poor fresh water in fresh water vadose regime was inverted the aragonite to calcite. Traces of celestite in these carbonate cements could be identified by the X-ray diffraction analysis. The author believes that it was formed as the result of diagenetic alteration of high strontium bearing aragonite into low strontium bearing calcite and the subsequent reaction between  $\text{Sr}^{2+}$  and  $\text{SO}_4^{2-}$  (originating from solution of calcium sulfate existing in the studied facies by fresh water).

Assereto and Folk (1980) believed that the fibrous calcite correspond to slow precipitation from brine films, while they related the micrite to rapid precipitation, possibly by marine water during water table fluctuation. However, the author observed that the studied micrite laminae are sometimes include algal remains represented by minute tubes similar to Recent calcified Cyanophytes. Therefore, the author agree with Wright (1980), that the micrite laminae were built at least in part by filamentous Cyanophytes.

#### *Formation of Native Sulfur*

The mechanism of formation of the native sulfur, calcite and secondary gypsum is outlined as follows:

- 1) The downward percolating ground water partially dissolved sulfate in localized microenvironments and transformed the secondary anhydrite into secondary gypsum.
- 2) Sulfate reducing bacteria acted on the sulfate in the presence of organic matter as nutrient. The mode of occurrence and the small quantity of native sulfur suggest that the organic matter necessary for sulfate reduction was either from algae growing under such conditions and/or plant matter derived from the continent by circulating water.
- 3) The reduction of sulfate resulted in the formation of hydrogen sulfide gas and secondary calcite. Some  $\text{H}_2\text{S}$  escaped while a part was oxidized to elemental sulfur by the dissolved oxygen in the percolating water.

#### **Conclusion**

The coral-algal reefal limestone of Gebel El Rusas Formation was deposited in a shallow phreatic marine environment and represents the early stage of an evaporite cycle. A fall of sea level resulted in the development of local hypersaline

ponds and supratidal environments, in which the subaqueous evaporites and sabkha sequences of the Abu Dabbab Formation were deposited. The reefal limestone and evaporite facies were occasionally flooded by storm tides which provided marine water vadose condition. This resulted in the deposition of paleoflowstone crusts, spheroidal and hemispheroidal structures and aragonite ray - crystals.

Temporary influx of fresh water on the Miocene sequence (fresh water vadose regime) resulted in:

- 1) Formation of solution veins and cavities.
- 2) Transformation of aragonite into fibrous calcite and calcite pseudospars that may retain the original aragonite form.
- 3) Transformation of secondary anhydrite into secondary gypsum and formation of native sulfur and calcite from the action of sulfate reducing bacteria on sulfate minerals in a localized reducing microenvironment. The organic matter necessary for sulfate reduction was either derived from algae growing under such condition and/or plant matter derived from the continent by circulating water.

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## أنسجة ما بعد الترسيب للأراجونيت ، الكالسيت والكبريت في متبخرات الميوسين على ساحل البحر الأحمر (مصر)

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يهدف هذا البحث إلى دراسة أنسجة ما بعد الترسيب ودلالة هذه الأنسجة في صخور الميوسين الأوسط على ساحل البحر الأحمر، مصر. بدأ ترسيب هذه الصخور بترسيب الحجر الجيري المرجاني الطحلي (مكون جبل الرصاص) في بيئة بحرية ضحلة، تبعتها ترسيب صخور المتبخرات (مكون أبو دباب) في لاجونات وسبخات شاطئيه ثم صخور فتاتية قارية مختلطة مع صخور بحرية ضحلة (مكون سمح). غمرت هذه الصخور بواسطة المد العالي الأعصاري من وقت لآخر مما نتج عنه ترسيب مواد جيرية لاحمة من معدن الأراجونيت في فجوات هذه الصخور في صورة أنسجة كروية، نصف كروية وفلوستون. بعد ذلك تأثرت صخور مكون جبل الرصاص ومكون أبو دباب وكذلك المادة اللاحمة بمياه عذبة فقيرة في عنصر المغنسيوم مما نتج عنه:

- ١ - تحول معدن الأراجونيت إلى كالسيت.
- ٢ - تكوين معدن السيلستيت.
- ٣ - تحول معدن الأنهدريت الثانوي إلى جبس ثانوي.
- ٤ - تكوين معدن الكبريت والكالسيت كنتيجة لفعل البكتريا المختزلة على معادن الكبريتات في وجود بعض المواد العضوية.