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Geochemical and mineralogical studies of the celestite bearing formations and associated sediments in Benghazi Formation, Ar Rajmah Group (Middle Miocene), Al Jabal Al Akhdar, NE Libya

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Highlights

- **This paper has an excellent contribution to the geology of Cyrenaica.**
- **In this paper find out geochemically and mineralogically the source of strontium element. Also the distribution of strontium in allochems, matrix and cement in the Benghazi Formation of the Ar Rajmah Group.**

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ABSTRACT

The study area is located in the northeast of Libya, in the areas of Benghazi city and Ar Rajmah village. The purpose of this study is to find out geochemically and mineralogically the source of strontium element. Also the distribution of strontium in allochems, matrix and cement in the Benghazi Formation of the Ar Rajmah Group. Celestite grade 88.3 % ($SrSO_4$) occurs only in the Benghazi Formation at Benghazi Cement Quarry as geodic nodules in the Middle Miocene rocks. Detailed mineralogical and chemical data have been collected on the celestite and associated sediments using XRF, XRD, EPMA, SEM and DSC techniques. Authigenic celestite and strontianite minerals occur both as a cement and as a replaced by other fracture filling minerals such as anhydrite. However, carbonate skeletal grains have quite high $MgCO_3$ contents, reflecting sometimes the presence of dolomite and brucite minerals. The average Sr/Ca ratio of coralline algae is 0.65. Echinoderm fragments have the highest value 1.31 of Sr/Ca and containing 0.47 to 1.8 mole percent of $MgCO_3$ in the Benghazi Formation. Echinoid spines contain higher strontium contents than other echinoderm fragments. Some bivalves are replaced totally by apatite and had a very high absolute concentration of strontium. The high strontium- low magnesium contents and vice versa are related to the diagenesis, mineralogical nature of the sediments and organic remains. Celestite occurs mainly in sedimentary beds of the Benghazi Formation, adjacent to the evaporitic environment (gypsum/anhydrite) and near sites of intense dolomitization.

Introduction

The geology of Libya is dominated by sedimentary rocks, occurring in distinct sedimentary basins. The basins of southern Libya are filled with Palaeozoic and continental Mesozoic strata. In northern Libya, the Precambrian and Palaeozoic strata are covered by marine Mesozoic and Tertiary sediments, which consist principally of carbonates and marl (Klitzch, 1968). Many general investigations have been carried out on the geology of Libya including the geology of Cyrenaica region. Most of these studies have concentrated on the surface outcrops of Al Jabal Al Akhdar, providing a general description of the lithofacies and some detailed work on the micropalaeontology. The sedimentary sequence exposed at the surface of Al Jabal Al Akhdar ranges in age from Cenomanian to Quaternary (Kleinsmiedi and Van Den Berg, 1968; Klen, 1974; Zert, 1974; Rohlich, 1974; El Hawat and Shelmani, 1993). The locations of the study area; Benghazi Cement Quarry and Ar Rajmah Quarry are illustrated in Fig. 1.

Objectives

The celestite in the study area has not yet been mentioned in the previous studies and has been considered in this work to determine geochemically the source of strontium element in Benghazi Formation. The study is also included the distribution of strontium in allochems, matrix and cement in the Benghazi Formation of the Ar Rajmah Group.

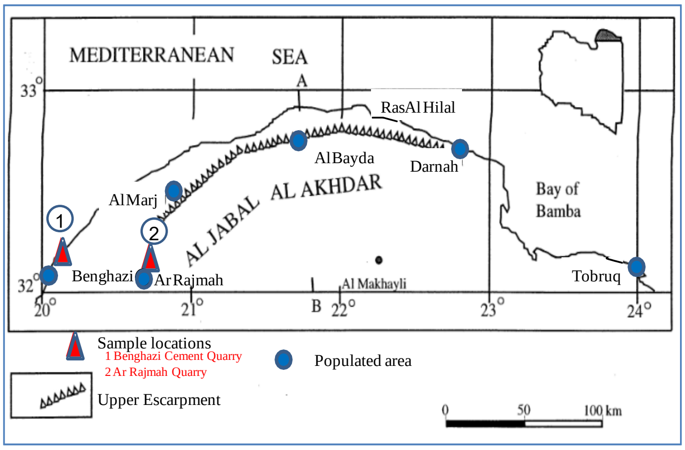

Fig. 1. Location map of the studied area, Benghazi Cement Quarry and Ar Rajmah Quarry

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Methodology

Strategic evaluations of the limestones in Benghazi Formation, the work in this paper need to involve far more than a basic geological appraisal and it was included laboratory determinations of the chemical and mineralogical properties of this rock types. A variety of techniques were used in this study included; X-Ray Fluorescence (XRF), Scanning Electron Microscopy (SEM), Electron Probe Micro-analysis (EPMA), X-Ray Diffraction (XRD) and Differential Scanning Calorimetry (DSC).

Stratigraphy of Ar Rajmah Group, Benghazi Formation (Middle Miocene)

Ar Rajmah Group is the youngest unit in Al Jabal Al Akhdar area (Fig. 2). It comprises of two formations; from the oldest to the youngest are Benghazi and Wadi Al Qattarah formations. This division is based on the differences in lithology and faunal content (Klen, 1974; Rohich, 1974).

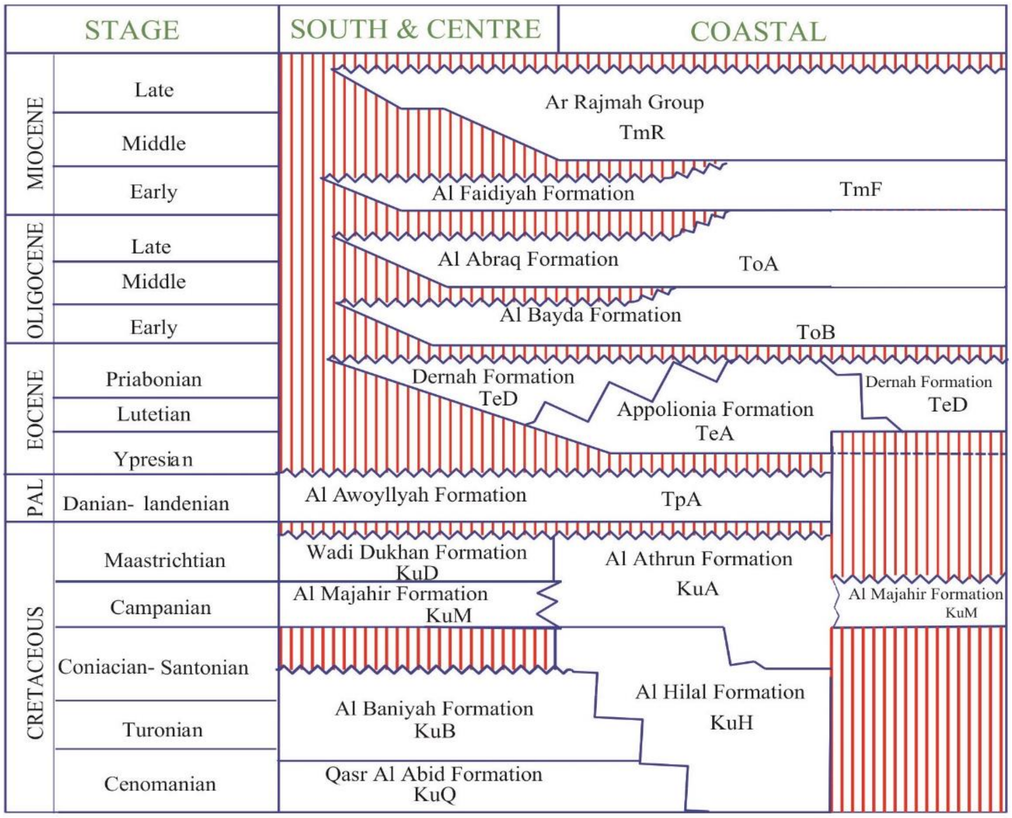

Fig. 2. Surface and Stratigraphic chart of Al Jabal Al Akhdar, NE Libya (modified after Muftah *et al.*, In press)

Benghazi Formation

The formation consists chiefly of fossiliferous limestone, white to yellowish, thick-bedded to massive (< 10 % dolomite), hard to medium hard (Fig. 3). With an abundance of fossils including large echinoids, sometimes bored (Fig. 4), bivalves e.g. Pectinids and gastropods. There is an abundance of coralline algae and one species of coral *Tarbellastraea* sp. in the lower part of the formation, was noticed in Benghazi Cement Quarry. It also contains brachiopods and echinoid spines, bryozoans and foraminifers. Celestite is also observed in the Benghazi Formation of the Ar Rajmah Group. Celestite of brownish colour and it is formed of friable prismatic crystals aggregates infilling vuggy pores (Figs. 5 and 6). XRD of a powdered specimen showed the presence of impurities such as relics of anhydrite, gypsum, calcite and dolomite. The presence of anhydrite within the celestite crystals indicates that the origin might be anhydrite nodules later replaced to celestite, as described by Wood and Shaw (1976). Authigenic celestite and strontianite minerals occur both as a cement and as a replaced by other fracture filling minerals such as anhydrite (Fig. 7).

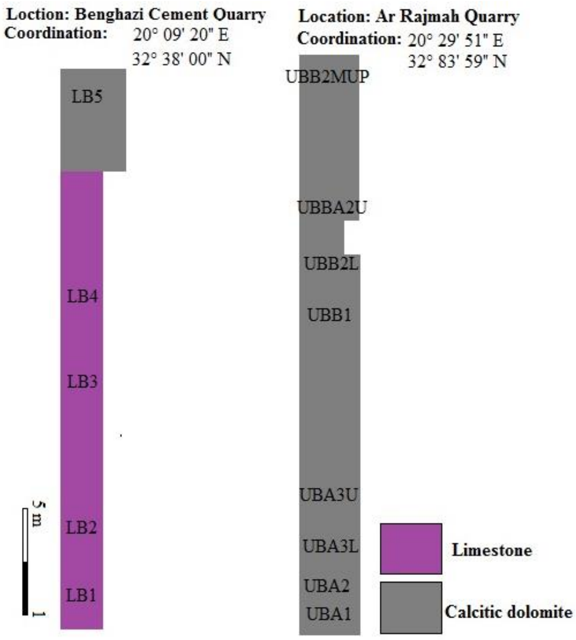

Fig. 3. Two logs of the Benghazi Cement Quarry (Lower Part), and Ar Rajmah Quarry (Upper part) of the Benghazi Formation (Middle Miocene).

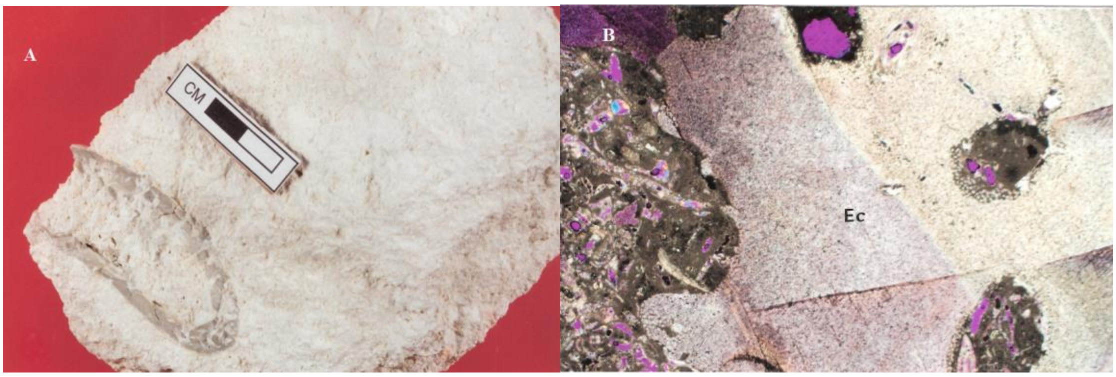

Fig. 4. A) Echinoid; B) Large echinoderm fragment (Ec), with borings filled with carbonate and bioclasts, Benghazi Cement Quarry. Field of view= 6 mm, (XPL, with gypsum accessory plate).

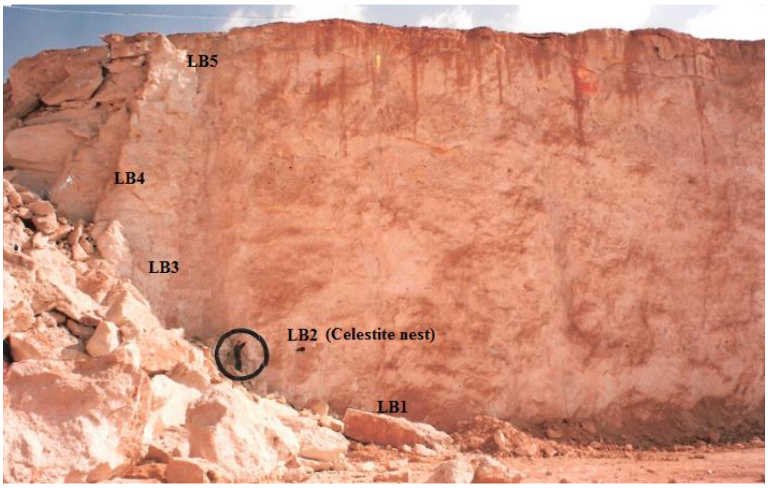

Fig. 5. A massive limestone bed of Benghazi Formation, Benghazi Cement Quarry. Displays; LB1, LB2, LB3, LB4 and LB5.

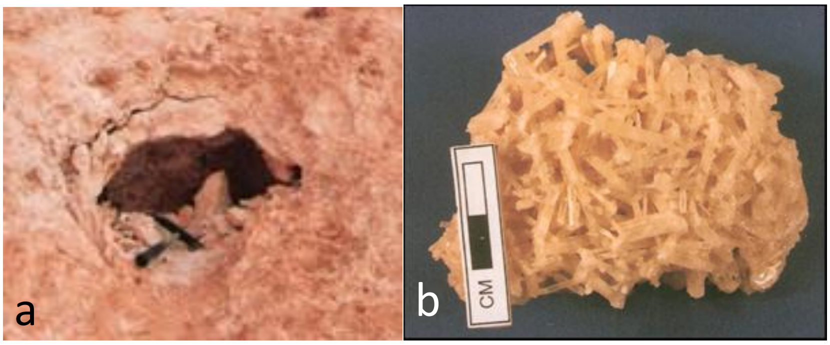

Fig. 6. a) Celestite nest at height of 4.5 m; and b) Celestite mineral sample was taken from the nest (4.5 m) and as geodic nodules, Benghazi Formation, Ar Rajmah Group, in Benghazi Cement Quarry.

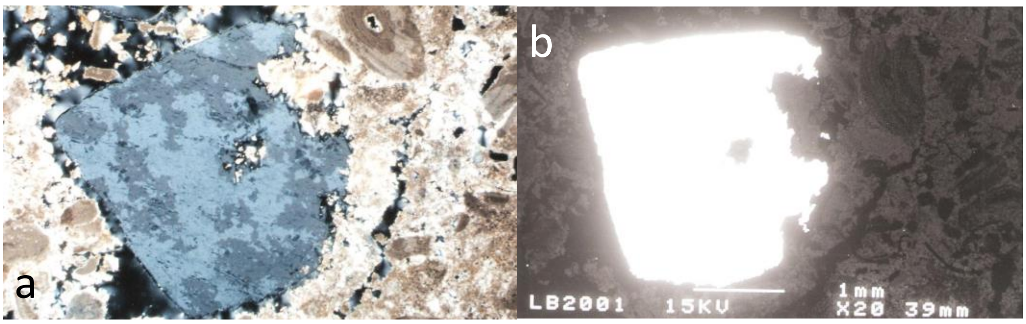

Fig. 7. a) A photomicrograph of a Strontianite mineral is etched by dolomite crystals, Field of view = 6 mm; and b) SEM (BSE) of the strontianite mineral, the brightness is due to the highest in atomic number, Benghazi Formation.

Dolomite in Benghazi Formation

Dolomite mineral is common in the Benghazi Formation in varying amounts. The lower part of the formation contains about 2 - 8 % dolomite whereas in the upper part (Ar Rajmah Quarry), consists of 10 - 80 dolomite (Table 1). Generally, dolomite crystals were present as limpid euhedral crystals with a planar (idiotopic), inequicrystalline (micron to decimicron in size) fabric (Fig. 8). It occurs both as fabric selective replacement of the carbonate mud matrix and as a cement. Grains of primary high-Mg calcite (HMC), such as red algae (Fig. 9), foraminifers, and echinoderm fragments are often more susceptible to dolomitization than low-Mg calcite (LMC); (Buchbinder, 1979; Armstrong *et al*., 1980; Sibley, 1980; Tucker, 1991). Some dolomite crystals have a hollow centre and that may result of the complete removal of metastable dolomite and/or soluble calcite zones (Sibley, 1982; Ward and Halley, 1985; Theriault and Hutcheon, 1987). Overall, the Benghazi Formation is quite porous and point-counter estimates of porosity range from 4 to 38 %.

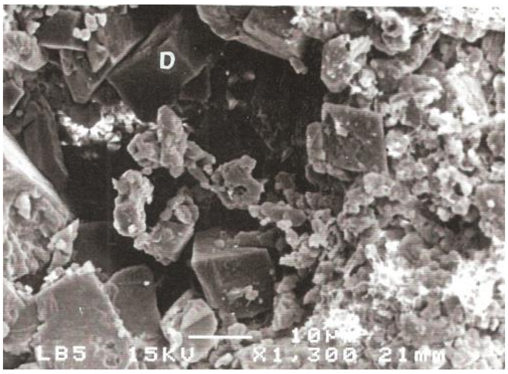

Fig. 8. SEM of the void-filling dolomite (D), Benghazi Formation (LB5), Benghazi Cement Quarry.

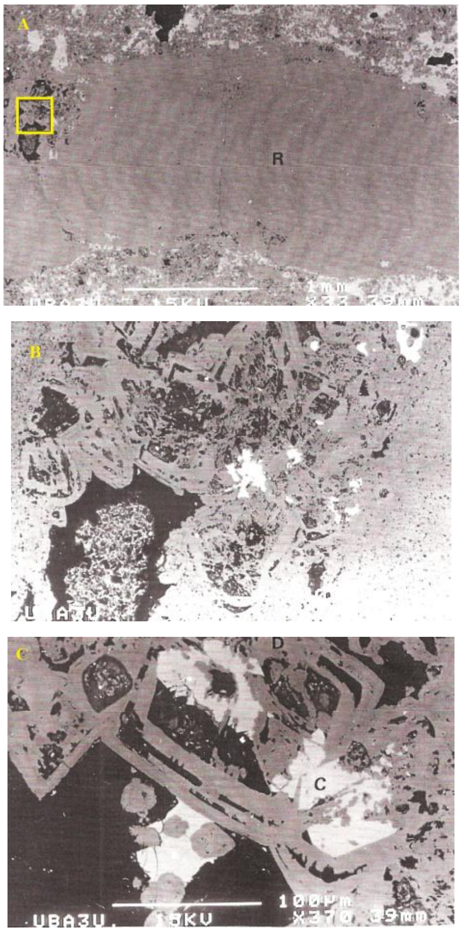

Fig. 9. SEM-BSE; A) Red algae (R); B & C) Close up view showing some euhedral dolomite (D) crystals cores filled with calcite (C).

Table 1

Chemical analysis for major and trace elements of the lower part of the Benghazi Formation (M. Miocene), Benghazi Cement Quarry.

Sample no.	LB1	LB2	LB3	LB4	LB5	UBA1	UBA2	UBA3L	UBA3U	UBB1	UBB2L	UBB2U	UBB2MUP
Oxides (wt. %)	Benghazi Cement Quarry (Lower part of the Benghazi Formation)					Ar Rajmah Quarry (Upper part of the Benghazi Formation)							
SiO_2	0.87	0.00	0.11	0.00	0.67	0.07	0.57	0.63	0.00	0.00	0.00	0.00	0.01
Al_2O_3	0.25	0.00	0.01	0.00	0.24	0.03	0.24	0.33	0.01	0.00	0.00	0.00	0.05
Fe_2O_3	0.09	0.00	0.00	0.00	0.06	0.03	0.03	0.05	0.00	0.00	0.00	0.00	0.04
$MgCO_3$	5.97	2.61	3.52	2.44	6.30	30.38	25.21	22.88	28.85	24.81	28.01	19.89	29.70
$CaCO_3$	92.78	97.7	96.65	98.11	92.66	67.21	72.03	74.25	68.95	73.81	70.22	78.98	67.78
S	0.00	0.02	0.00	0.00	0.00	0.03	0.03	0.02	0.04	0.02	0.04	0.02	0.07
K_2O	0.04	0.00	0.00	0.00	0.04	0.00	0.01	0.01	0.00	0.00	0.00	0.00	0.00
TiO_2	0.01	0.00	0.00	0.00	0.01	0.00	0.01	0.01	0.00	0.00	0.00	0.00	0.00
MnO	0.00	0.09	0.02	0.01	0.03	0.00	0.00	0.00	0.00	0.00	0.00	0.03	0.00
P_2O_5	0.03	0.00	0.03	0.01	0.03	0.12	0.18	0.30	0.20	0.24	0.17	0.17	0.31
Total	100.04	100.42	100.34	100.57	100.04	97.87	98.31	98.48	98.05	98.88	98.44	99.09	97.96
CaO	51.90	54.70	54.10	54.90	51.89	37.60	40.34	41.60	38.61	41.34	39.32	44.23	37.96
MgO	3.52	1.54	2.10	1.44	3.72	17.92	14.87	13.50	17.02	14.64	16.53	11.74	17.52
Limestone Quality	***LP**	***HP**	***MP**	***HP**	***LP**	**Impure**							
Trace elements (ppm)													
Nb	0	0	0	0	0	0	3	0	2	0	3	0	0
Zr	127	719	111	98	666	27	34	34	30	21	29	28	39
Y	40	34	41	42	31	42	43	46	43	44	42	40	45
Sr	1306	8042	1226	1082	7761	279	274	261	310	226	291	265	373
Rb	44	41	42	47	38	44	45	43	43	41	45	45	45
Zn	0	0	0	2	0	0	6	0	0	0	0	0	0
Cu	12	6	16	11	2	12	19	11	9	18	29	31	12

Table 2, continued

Ni	0	0	0	0	0	0	0	0	0	0	0	0	0
Cr	18	0	0	11	1	17	17	14	24	21	13	18	32
Ce	0	26	0	26	14	0	14	30	36	1	0	18	0
Nd	0	0	0	0	0	0	0	18	8	0	0	7	0
V	18	14	11	11	9	16	18	16	7	11	1	18	25
La	12	4	0	18	8	21	11	0	13	0	9	0	0
Ti	321	188	204	250	186	255	322	316	226	169	188	173	223
Ba	1	0	0	0	25	0	0	0	0	8	0	0	0
Sc	87	79	77	100	87	46	62	48	67	67	70	71	45

* LP = Low Purity, MP = Medium Purity and HP = High Purity; Based on Harris, 1979 Classification

Wadi Al Qattarah Formation

This formation occurs close to the main road to Deryanah Al Abyar. It is composed of hard, white, porous, oolitic limestone, with massive gypsum in the upper part. Thin section showed grainstone texture with ooid and superficial ooids, Isopachous cement is also observed (Fig. 10a). This circumgranular calcite crust cement is characteristic of phreatic precipitation. Gypsum occurs in the upper part of the formation (Fig. 10b) together with quartz grains and minor amounts of scattered micron-sized dolomite crystals filling fractures.

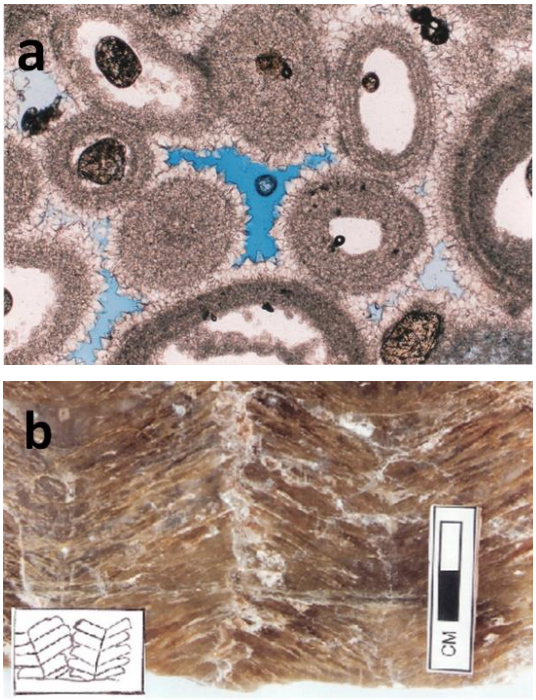

Fig. 10. a) Ooid grainstone with isopachous cement (vacuum impregnated with blue-dye resin), Field of view = 1.3 mm, (PPL); b) Coarsely crystalline swallow-tailed gypsum from a quarry NE Ar Rajmah village, Wadi Qattarah Formation.

Results and Discussions

DSC curve of celestite:

In air, the DSC curve shows two steps resulting from weight losses. The first weight loss of 3.7 % is associated with peaks 785 °C and 801 °C. The second peak showed a 0.37 % loss at about 1060 °C. The last endothermic peak is very sharp at about 1155 °C (Fig. 11). The first and second peaks represent traces of dolomite and that's confirmed by XRD analysis. The small peak at 1060 °C and very sharp peak at 1155 °C are probably due to the presence of some foreign material such as impure strontianite. Celestite was heated at a range of temperatures (800°C, 1000°C, 1150°C, and 1500 °C; Fig. 12). The only effects to be detected using XRD (approx. 0.5 mg) include a change in the sequence of peak intensities (Table 2). There is no change in mineralogical composition, but the degree of order changes, reducing at a higher temperature.

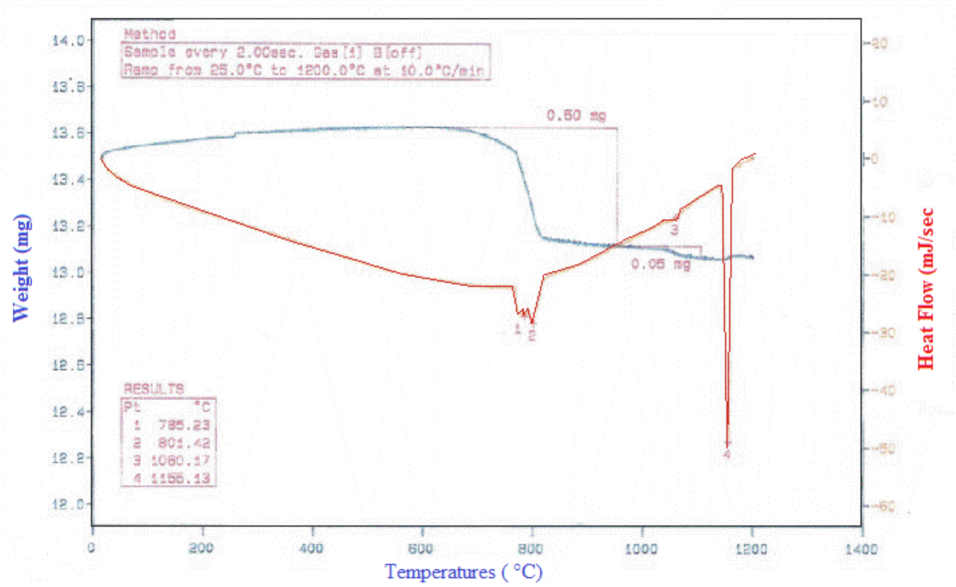

Fig. 11. TG-DSC curves of celestite of the Benghazi Formation, in the air (sample size = 13.5 mg) and CO_2 (sample size = 18.82 mg) at Benghazi Cement Quarry.

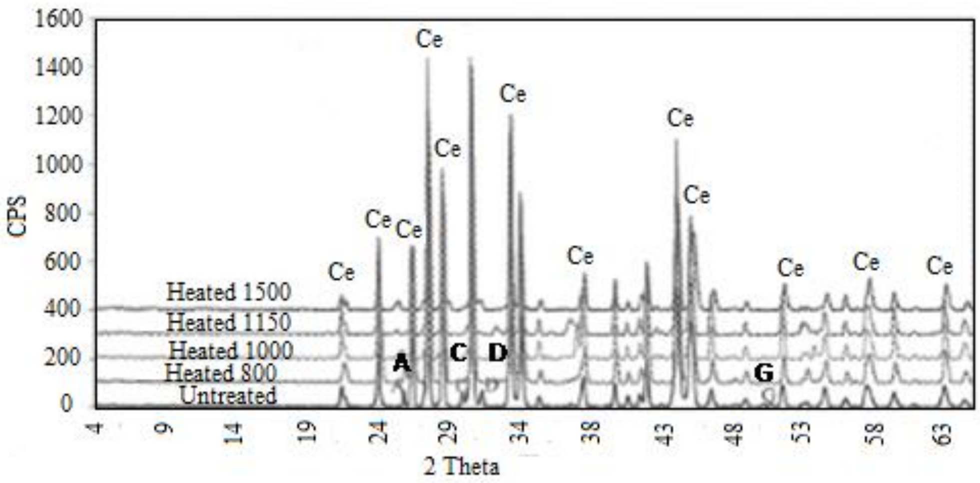

Fig. 12. XRD pattern of celestite mineral of the Benghazi Formation showing; (Ce) Celestite; (A) Anhydrite; (G) Gypsum; (D) Dolomite; (C) Calcite. Untreated (Air treated) and treated (heated to 800°C, 1000°C, 1150°C, 1500°C.

Table 2

Dominant X-ray diffraction peaks of celestite, in the Benghazi Formation

Age	Formation	Temperature (°C)	2 Theta	d-spacing d (Å)	Int. (%)	hkl
Middle Miocene	Benghazi	untreated	30.095	2.967	100	211
			27.077	3.291	98.80	210
			28.093	3.174	77.27	102
			32.793	2.729	67.02	112
			44.321	2.042	62.38	113
		800	27.210	3.275	100	210
			30.217	2.995	93.08	211
			44.431	2.037	68.49	113
			32.908	2.720	65.56	112
			28.216	3.160	59.56	102
		1000	27.104	3.287	100	210
			30.121	2.965	98.99	211
			32.805	2.728	60.89	112
			28.103	3.173	58.16	102
			44.337	2.041	52.89	113
		1150	27.094	3.289	100	210
			30.107	2.966	97.57	211
			32.807	2.728	74.07	112
			28.107	3.172	60.83	102
			44.317	2.042	54.16	113
		1500	30.178	2.959	100	211
			27.147	3.282	82.88	210
			32.837	2.725	80.79	112
			33.485	2.674	53.89	020
			28.165	3.166	50.55	102
			44.358	2.041	39.33	113

Occurrence and distribution of Sr in Benghazi Formation

Most the limestones in the study area contain skeletal materials such as shells and coral skeletons. Coralline algae contain a large amount of an $MgCO_3$, sometimes more than 35 $MgCO_3$ mole percent. The distribution of high Mg-Calcite (HMC) and low Mg-Calcite (LMC) in coralline algae and the other components were discovered and illustrated in Fig. 13.

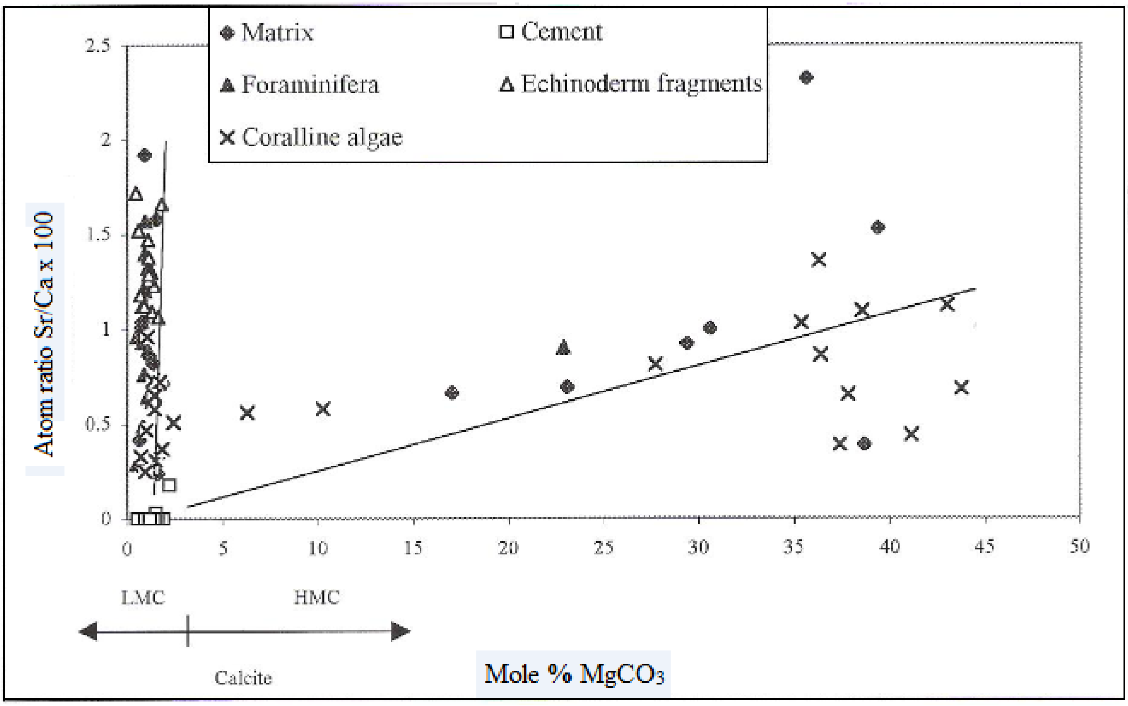

Fig. 13. Scatter diagram of Sr/Ca x 100 atom ratios vs Mole % $MgCO_3$ concentrations of the major components (coralline algae, echinoderm fragments, foraminifera, matrix and cement) in Ar Rajmah Group, Benghazi Formation.

Low magnesium calcite has less than 4 mole percent $MgCO_3$ and high magnesium calcite greater than 4 mole percent (Chave *et al.*, 1964; Blatt *et al.*, 1980; Carter, 1990; Tucker, 1991). Because of their lower stability, carbonate skeletal materials with high magnesium calcite, such as coralline algae are most likely to be replaced with metastable phases (e.g brucite). However, carbonate skeletal grains have quite high $MgCO_3$ contents, reflecting sometimes the presence of dolomite and brucite minerals. The average Sr/Ca ratio of coralline algae is 0.65. Echinoderm fragments have the highest value 1.31 of Sr/Ca and containing 0.47 to 1.8 mole percent of $MgCO_3$ in the Benghazi Formation. Echinoid spines contain higher strontium contents than other echinoderm fragments. Some bivalves are replaced totally by apatite and had a very high absolute concentration of strontium (Fig. 14; Table 3).

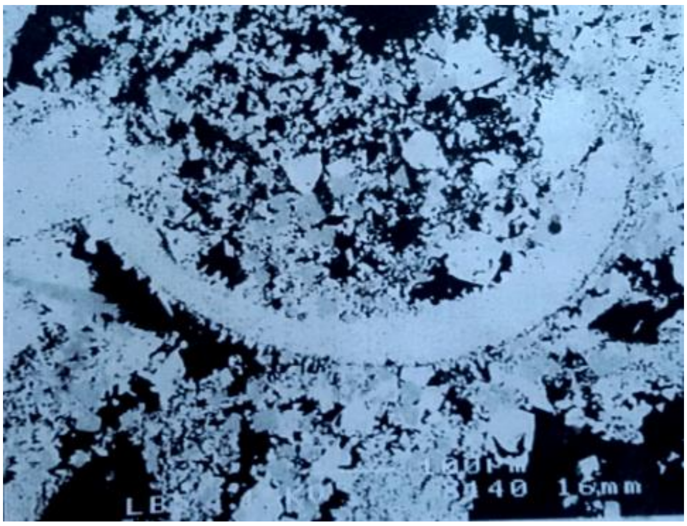

Fig. 14. BSE-SEM image of bivalve, totally replaced by apatite in Benghazi Formation

Table 3

High-resolution EPMA analyses of the bivalve shell fragment in (Sample no. LB5) Benghazi Formation

Oxides (wt. %)	Sample no. LB5 (Benghazi Cement Quarry)		
P_2O_5	36.495	39.047	36.456
SO_2	1.061	0.327	1.041
TiO_2	0.002	0.016	0.016
MgO	0.149	0.140	0.154
CaO	50.470	50.305	50.105
MnO	0.000	0.000	0.004
FeO	0.185	0.321	0.195
NiO	0.000	0.007	0.000
CuO	0.000	0.000	0.002
SrO	0.226	0.161	0.299
BaO	0.000	0.000	0.034
Na_2O	1.120	1.024	1.100
K_2O	0.021	0.017	0.015
Total	89.729	91.365	89.421

Magnesium and strontium relationship

The amount of strontium in the lower Benghazi Formation (Benghazi Cement Quarry) increases due to contamination of the limestone rocks with celestite and dolomite as well. The magnesium content decrease as strontium content increases and reaches a maximum where the strontium content reaches a minimum (Table 4). The high strontium- low magnesium contents and vice versa are related to the diagenesis, mineralogical nature of the sediments and organic remains. The loss of Sr is connected with observed high dolomitization, Sr % vs Mg % (Fig. 15) showed a decrease in Sr content with increasing dolomitization and showed also the Sr distribution and its relation to facies types.

Table 4

Strontium, magnesium and calcium analyses (XRF) of sediments of the traverse LB1 to LB5 of the Benghazi Formation (Benghazi Cement Quarry).

Location	Ca Wt. %	Mg Wt. %	Sr Wt. %	Atom ratio Sr/Ca x 1000 (Siegel, 1961)	Celestite & strontianite + Facies
Benghazi Cement Quarry					
LB1	37.09	2.12	0.13	1.61	Present + Dolomitic limestone
LB2	39.09	0.93	0.13	1.54	
LB3	38.66	1.27	0.12	1.45	
LB4	39.24	0.87	0.11	1.26	
LB5	37.09	2.24	0.11	1.39	

In other places such as Ar Rajmah the facies becomes calcitic dolomite to dolomite with absent of celestite minerals and diminishes in Sr content.

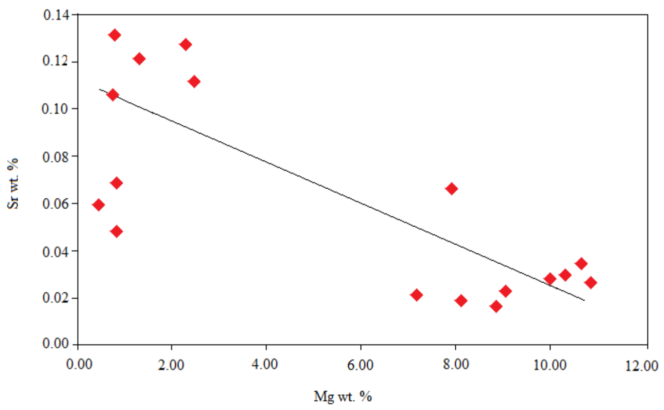

Fig. 15. Sr (wt. %) vs Mg (wt. %) of the Benghazi Formation, Ar Rajmah Group

Dolomite, anhydrite, celestite and replacement

Celestite ($SrSO_4$) is a member of the barite group and the principal source of strontium in many respects but it has a considerably lower density. Celestite occurs mainly in sedimentary rocks, by the interaction of gypsum or anhydrite with Sr-rich (Harben and Bates, 1990; Ober, 1994; Chang *et al.*, 1996). Celestite in the study area contains the strontium that was released from limestone in the formation of dolomite (such dolomites are extremely low in strontium content). Dolomite and celestite are often associated with the presence of evaporate deposits such as gypsum and/or anhydrite and the percolation within limestone by waters containing high sulphate. Generally, the composition of celestite depends on three main factors; 1) the nature and intensity of diagenesis such as the chemical composition of interstitial waters on their movement and renewal; 2) dolomitization; 3) recrystallization as described by West (1964), Lloyd and Murry (1965), Jorgensen (1994), Purser (1998) and Rosell *et al.*, (1998). The celestite in the lower part of Benghazi Formation occurs mostly as clear prismatic or tabular euhedral crystals and is coarse to extremely coarsely crystalline. Most of the replacement celestite occurs in nodular anhydrite; a complete replacement of isolated anhydrite nodules was occasionally observed. XRD analysis of the central portion of celestite nodules showed numerous relicts of anhydrite (Fig. 16).

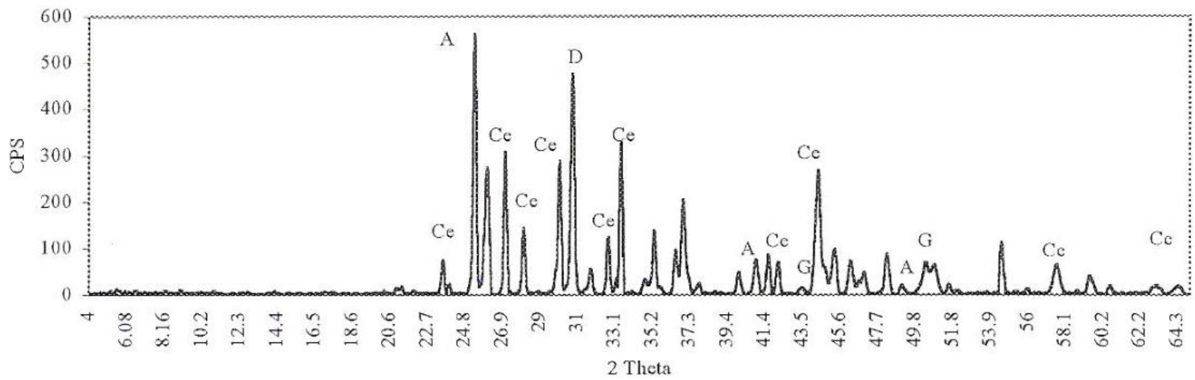

Fig. 16. XRD pattern of sediment taken from celestite nest of the Benghazi Formation (Benghazi Cement Quarry). Showing Celestite (Ce), Anhydrite (A), Dolomite (D) and Gypsum (G).

In Rhaetia limestones of southern England, gypsum veins originating from underlying Triassic strata have similarly been converted to celestite (West, 1964). Because of its low solubility in water, celestite of Ar Rajmah Group could be also precipitated previously in association with dolomite (in high porosity) and after removal took place, celestite re-deposited in the lower part of Benghazi Formation (Benghazi Cement Quarry).

Formation of Celestite

Celestite has been formed from pore water enriched in both SO_4 and Sr contents. A possible mechanism for concentration of strontium ions may be related to the loss of strontium by; 1) aragonite to calcite transformation (Dodd, 1967; Bathurst, 1975; Nicholas, 1978); and 2) intense dolomitization of aragonite mud. The Sr released during the replacement of aragonite (7000 - 8000 ppm Sr) by dolomite (600 - 700 Sr) is added to interstitial water and form celestite (Kinsman, 1969; Garea and Baitwaite, 1996). Nickless et al., (1976), concluded that sulphate is derived from anhydrite and gypsum, suggested that the strontium became available when aragonite converted to calcite. Celestite is much less soluble than gypsum and anhydrite and in most cases, celestite is formed by the replacement of those two minerals (West, 1973). Gypsum or anhydrite both contain small amounts of strontium, more strontium can be accommodated in solid solution in anhydrite (up to 0.74 % = 7400 ppm) than gypsum (up to 0.1 %; Dean and Tung (1974)). Therefore, in the transformation from anhydrite to gypsum, Sr can be released to form celestite (de Brodtkorb, 1989). Olaussen (1981) presented three models to explain the formation of celestite in a subtidal to supratidal facies in the Wenlock of Norway; 1) early diagenetic dolomitization of aragonite mud release Sr, which may react with the brine to form celestite; 2) Sr enrichment of the interstitial fluid produced by transformation of aragonite to calcite; and 3) late diagenetic dolomitization in which Sr released may precipitate as late-stage authigenic celestite in geodes and veins.

Conclusion

- Celestite occurs mainly in sedimentary beds of the Benghazi Formation, adjacent to the evaporitic environment (gypsum/anhydrite) and near sites of intense dolomitization.
- The high strontium content of the bulk sample (LB2) examined by XRF (see Table 1) is due to contamination by celestite relicts.
- The significant variations in their strontium contents commonly accompany the transition from one facies to another, influenced mainly by diagenetic processes such as inversion of aragonite to calcite and replacement of calcite by dolomite dolomitization as well as part of the strontium content of the originally aragonitic bioclasts have been mobilized during late diagenesis. All these factors have provided considerable amounts of strontium.
- Fossils from low permeability/porosity sediments such as shale or claystone (closed system) may better indicators of the amount of strontium present originally within the fossils than highly porous and permeable limestones (open system). The fossil echinoderm fragments reported in the literature are low in strontium content, but in fact, have commonly the highest Sr/Ca values of all allochems in the Benghazi Formation of the Ar Rajmah Group (Table 5).

Table 5

EPMA analyses (average of about 20 readings each) of strontium contents of the main components in the Benghazi Formation.

Group	Formation	Bioclasts					
			Red algae	Echinoderm fragments	Foraminifers	Matrix	Cement
Ar Rajmah	Benghazi	Mg (wt. %)	7.08	0.32	0.62	3.67	0.33
		Ca (wt. %)	28.70	39.94	38.68	35.28	40.56
		Sr (wt. %)	0.20	0.10	0.06	0.05	0.001
		$MgCO_3$ (Mol.%)	24.45	1.08	2.14	12.68	1.13
		Atom ratio Sr/Ca ×1000	0.72	0.94	0.69	0.63	0.01

In the study area, Wadi Al Qattarah Formation, the upper part of Ar Rajmah Group is consists of white, porous oolitic limestone with gypsum. Freshwater infiltrating through the gypsum of this formation and may have dissolved calcium sulphates, resulting in the groundwater being enriched with SO_4. The Benghazi Formation has high porosity (secondary porosity), and due to dolomitization and dissolving aragonite fossils, and these are giving a high Sr content in the interstitial water. However, Sr reacted with SO_4 within and formed celestite in pore structures of the lower part of the Benghazi Formation (Fig. 17).

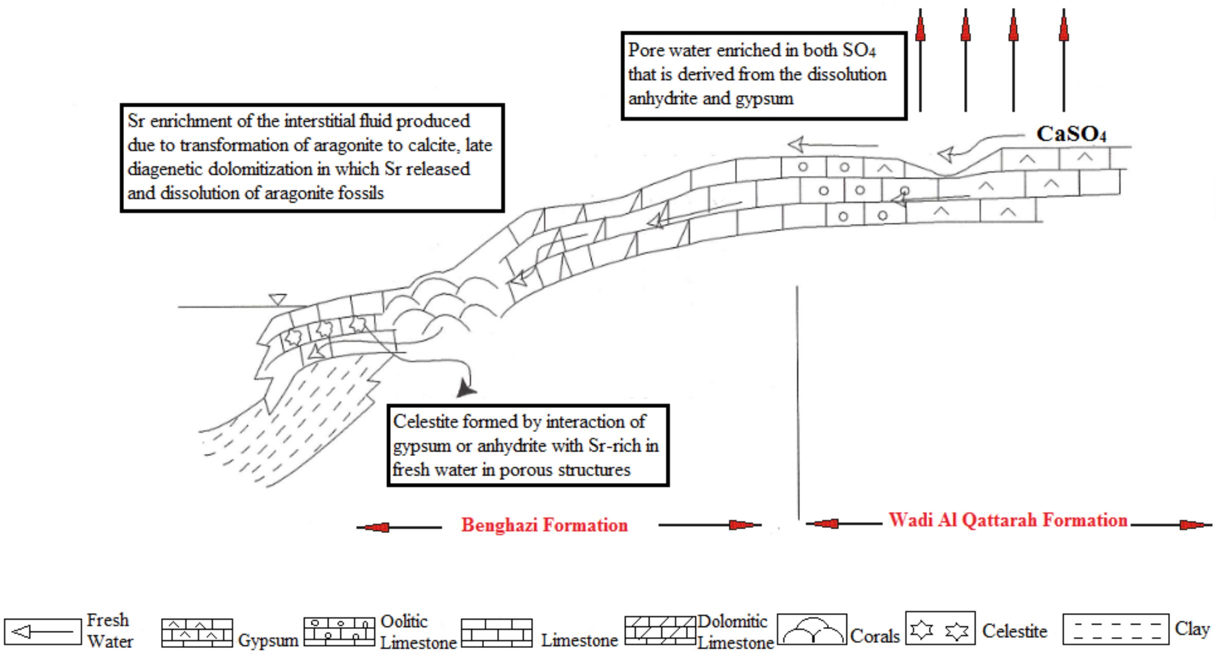

Fig. 17. A schematic model for the formation of celestite in the lower part of the Benghazi Formation of Ar Rajmah Group.

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