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Diagenetic history of the lower oligocene algal limestone member of the Al Bayda Formation, at Al Wattayatt area, Al Jabal Al Akhdar, NE Libya.

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Highlights

- **Timing of paragenetic sequence approved to be good tool to understand diagenetic history.**
- **Diagenetic history is very important to construct the burial history of sediments.**
- **Algal Limestone Member of Al Bayda Formation has suffered extensive diagenetic processes..**

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ABSTRACT

The Algal Limestone Member of the Al Bayda Formation has been studied along Deryanah-Al Abyar road cut at Al Wattayatt area, Al Jabal Al Akhdar. The member was subdivided into five lithofacies based on petrographic examination; from bottom to top are; wackestone lithofacies, boundstone lithofacies, grainstone lithofacies, mudstone-wackestone lithofacies, and packstone lithofacies. Limestone is the dominant lithology with coralline red calcareous algal, nummulites, bryozoans, echinoids, and few corals are also present. Lithological description and paleontologic content indicate that this member was likely deposited within a shallow marine or near-surface environment that was dominated during the Lower Oligocene. However, diagenetic analysis reveals two stages of paragenetic processes, which had a great effect on the development of the depositional environment and the diagenetic history of this member. The early-stage includes grain dissolution, brittle fracture, pyrite formation, and early cement. The late-stage includes secondary cement, neomorphism (recrystallization), and dolomitization.

1. Introduction

Several previous geologic studies and researches have been conducted in the Al Jabal Al Akhdar were mainly focused on the general stratigraphy, geomorphology, structure geology, and general sedimentology. As part of the geological map of Libya, Klen (1974) mapped the area around Benghazi city and produced Benghazi Sheet (NI 34-14). Röhlich (1974) mapped the area around Al Bayda city and produced Al Bayda Sheet (NI 34-15). Zert (1974) mapped the area around Darnah city and produced Darnah Sheet (NI 34-16). Each sheet is supplemented with a 1:250,000 geologic map and explanatory booklet. Important work was done by Muftah and Erhoma (2002); Hassan and Muftah (2008); El Hawat and Abdulsamad (2004); Abdulsamad and Tmalla (2009) in which detailed identification of the algal content, age dating, and depositional environment has been studied. However, detailed diagenetic studies on the exposed rock units in the Al Jabal Al Akhdar, however, are rare to none. A paragenetic study and lithofacies examination carried out on the Algal Limestone Member of the Al Bayda Formation have been achieved by using field observations and petrographic microscope. The study includes staining the samples with Alizarin Red-S (ARS). The aims of this paper are: 1) to address and analyze the main diagenetic processes that affecting the Algal Limestone Member of the Al Bayda Formation through time; 2) to determine the paleo-depositional and diagenetic history of this member. The study area is located approximately 40 km east of Benghazi city (Fig. 1). It is bounded by longitudes 20° 27' 25" to 20° 32' 36" E and latitudes 32° 19' 30" to 32° 20' 36" N. The measured section can be accessible from the highway due to the asphaltic road that passing through the Tansalukh-Al Wattayatt area and the road cut from Al Abyar town.

2. Geological Settings

Al Jabal Al Akhdar was developed as anticlinorium and has an exceptional position within the Cyrenaica region in the northeastern part of Libya. Cyrenaica is located at the southern margin of the Mediterranean Sea and formed as the attenuated continental crust of the northern passive margin of the Afro-Arabian shield during Upper Cretaceous to Tertiary (Röhlich 1980; Anketell 1996; El Hawat and Abdulsamad 2004). Cyrenaica consists of two distinct tectonic provinces that are separated by hinge-like known as Cyrenaica Fault System (El Hawat and Shelmani 2003; El Hawat and Abdulsamad 2004). The two tectonic provinces are the mobile Cyrenaica inverted basin, which is referred as Al Jabal Al Akhdar to the north, and the more stable Cyrenaica Platform to the south. The northern mobile belt area has been influenced by the Tethys tectonic activities and events since its opening during the Jurassic. It was subsequently shaped by the reversal of the Eurasian and African plate movement that led to subduction of the latter beneath the former from the Upper Cretaceous to the present. The tectonic events assisted the sedimentation cycles that were recorded in the following strata. The exposed strata at the Al Jabal Al Akhdar are mainly marine carbonate that ranging in age from Late Cretaceous to Late Miocene. In the study area of Wadi Al Wattayatt and the surrounding areas, the exposed rock units are from bottom to top as; Darnah Formation (Eocene); Al Bayda Formation (Early Oligocene) with its two members as Shahhat Marl Member at the bottom and

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Algal Limestone Member at the top; Al Abraq Formation (Late Oligocene); Al Faidiyah Formation (Early Miocene), Ar Rajmah Group (Miocene) with its two formations as Benghazi Formation (Middle Miocene) at the bottom and Wadi Al Qattarah Formation (Late Miocene) at the bottom. These formations are separated from each other by regional unconformities. unconformities.

3. Methods

Light microscope petrography is an extremely valuable tool for the identification of minerals and their textural relationships. It is best used, in many cases, in conjunction with other techniques, such as staining, x-ray analysis, or microprobe examinations (Scholle and Ulmer-Scholle, 2003). Staining techniques are among the fastest, simplest, and cheapest methods for getting reliable mineralogical and some qualitative elemental data on carbonate phases. Alizarin Red S (ARS) and Potassium Ferricyanide staining, in particular, are normally done together (Scholle and Ulmer-Scholle, 2003). In this study, five-rock samples collected from the Algal Limestone Member of Al Bayda Formation at Wadi Al Wattayatt have been thin-sectioned. In the laboratory, Alizarin Red-S (ARS) solution was prepared for staining the thin sections. A list of carbonate minerals and their diagnostic stains can be found in Friedman (1959), Dickson (1965, 1966), Milliman (1974), Friedman and Johnson (1982).

4. Results

The Algal Limestone Member of Al Bayda Formation represents a separate cycle of carbonate sedimentation in the Al Jabal Al Akhdar during the Lower Oligocene (Rupelian age) (Röhlich, 1974; El-Hawat and Abdulsamad, 2004). At the type locality, close to Al Hamamah village at the central Al Jabal Al Akhdar, this member is composed of ~ 40m thick sequence of a wide range of lithologies from mudstone to boundstone with dominant coralline red algae, an assemblage of bryozoa, echinoids, pelecypods, and foraminifera. The Algal Limestone Member of Al Bayda Formation at the present location has been divided into five major lithofacies (Fig. 2) based on lithologic changes, from bottom to top are:

1. ***Wackestone lithofacies***: it is a 9 m thick unit composed of white, fine-grained wackestone, moderately hard, with a concentration of algae balls and some echinoid fragments.;

2. ***Boundstone lithofacies***: it is a 3 m thick unit composed of cream-pale yellow, course-grained boundstone, hard, with high concentrations of algae, some echinoid and bryozoa, and *Nummulites* sp.;

3. ***Grainstone lithofacies***: it is a 6 m thick unit composed of white, grainstone, hard, with rich algae that increase upward in the unit, rich in foraminifera, echinoids, bryozoa.;

4. ***Mudstone-Wackestone lithofacies***: it is a 4 m thick unit composed of creamy to yellowish, mudstone-wackestone, hard, few algae and *Nummulites* sp.;

5. ***Packstone lithofacies***: it is an 11 m thick unit composed of white, packstone, hard, increase in *Nummulites* sp., and algal content upward, few echinoids, and bryozoa.

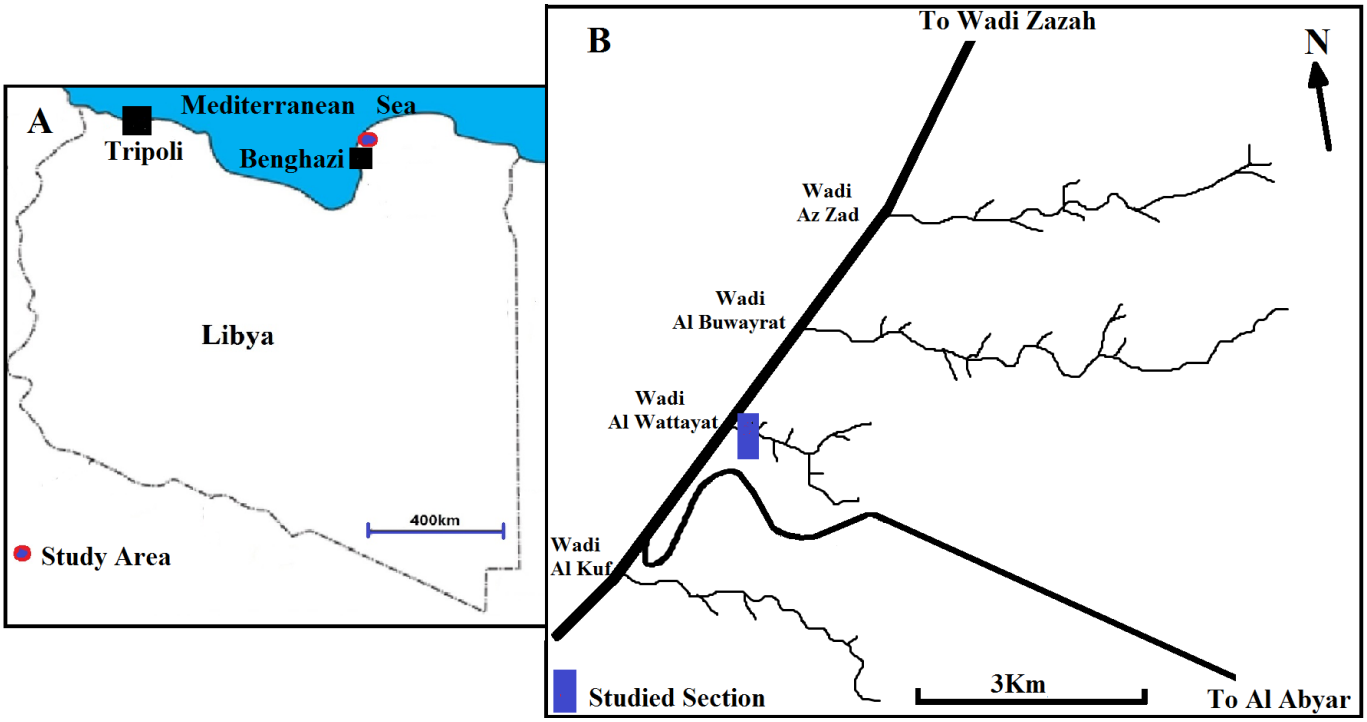


Fig. 1. (A) Location map of the study area (B) Location of the studied section along the left side of Wadi Al Wattayatt.

5. Diagenetic Processes

Sediments before lithified to a rock, pass through a sequence of diagenetic stages that take place in near-surface marine and meteoric environments, down into the deep-burial environments (Tucker, 2001). The processes that act on the sediments could be depositional or post-depositional in origin. Therefore, the texture of the sediment accompanied by the early processes of deposition is considered as depositional texture, while during the late processes that acted after deposition of the sediments, the texture will be of diagenetic origin (i.e. post-depositional). The depositional texture/fabric includes the grains and their features, while others include the diagenetic features of compaction, cementation, replacement, and dissolution of pre-existing minerals. Larson and Chilingar (1985) stated that sediments when deposited in a particular basin with the original texture as a product during deposition are the fundamental component and usually accompanied by many genetic processes. In general, the sequence of diagenetic processes that take place is in the following order: (1) biological and biochemical; (2) physical-chemical; and (3) physical (Termier and Termier, 1963). In the studied samples of the Algal Limestone Member of Al Bayda Formation, the observed diagenetic processes are (1) grain dissolution; (2) pyritization; (3) early cementation; (4) brittle fracturing; (5) secondary cementation; (6) neomorphism; (7) dolomitization.

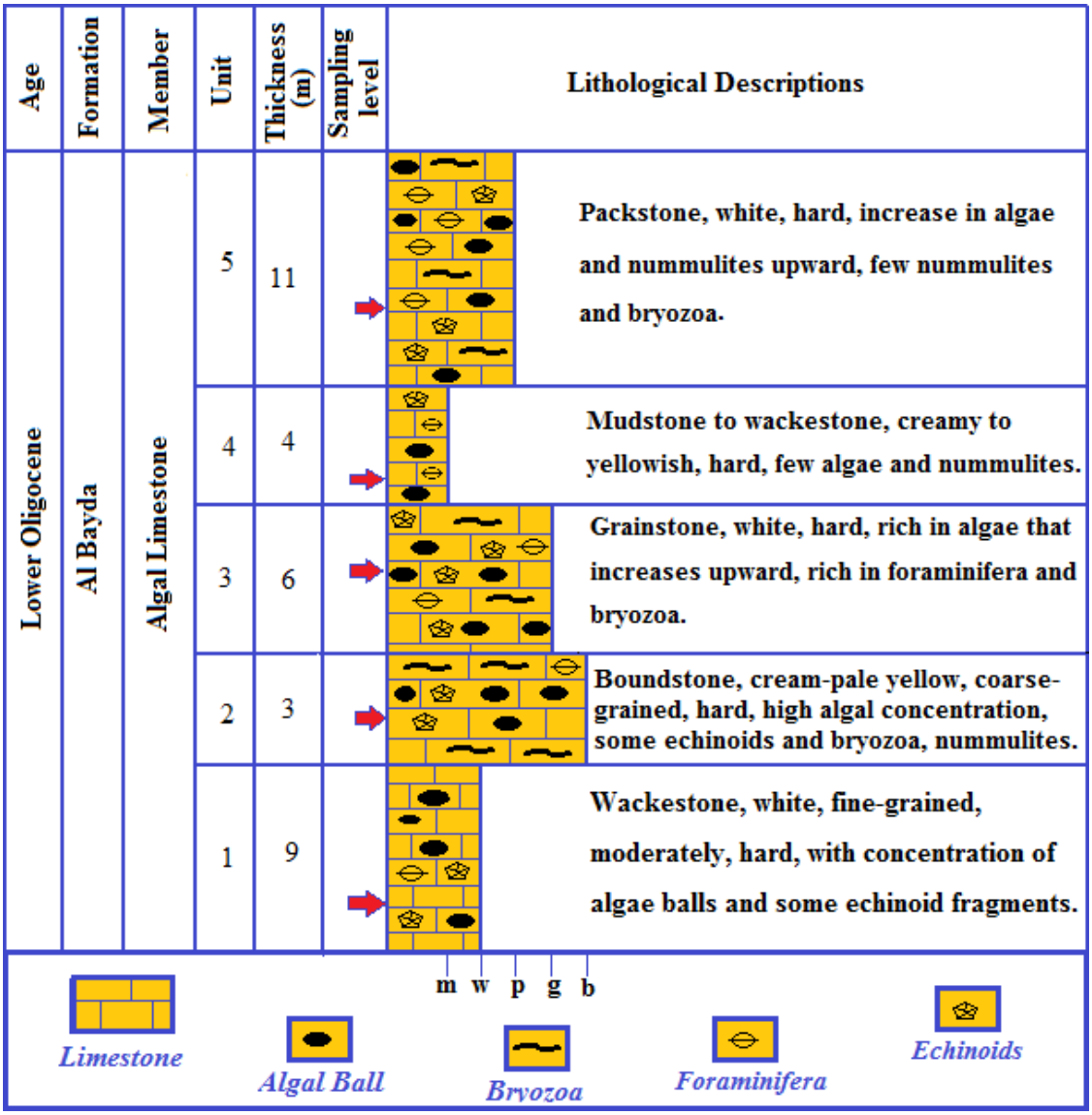


Fig. 2. Lithostratigraphic column of the Algal Limestone Member of Al Bayda Formation at Wadi Al Wattayatt section.

5.1 *Grain dissolution and pyrite formation*

Both are usually acting contemporaneously during the paragenetic sequence, particularly in carbonate rock. The dissolution of carbonates grains always commencing by corrosion along the cleavage planes of the minerals followed by continuous dissolution this will collapses the fragment of the grain, and in time, may remove the entire grain (Tucker, 2001). The resulted pores would be exposed to minerals-loaded solutions that usually precipitates pyrite mineral. Commonly, pyrite forms as 1) sooty (smoky) fine film on faunal skeletons; 2) minute pyrite sphere occurring in aggregates; 3) cubic forms. Pyrite forms in reduction organic matter regime under negative Eh (Krumbein and Sloss, 1963) and due to sulfide activity with the bacterial reduction to the sulfate in pore fluid (Tucker, 2001). Sometimes precipitate before calcium carbonate cement but may be replaced by both the textural grains and calcite cement. Pyrite can be formed in deep marine and euxinic basins (Larson and Chilingar, 1985). The minerals are distinguished from other opaque minerals by it is yellowish color in reflected light (Tucker, 2001). This feature is mostly observed in samples from the wackestone lithofacies.

5.2 *Early and secondary cements*

They are chemically precipitated minerals into voids and in-situ onto the surface of the host framework. The calcareous cements may be of different crystal sizes as micrite, microspar, and sparite; these are used to describe the cement size with the increase in grain size, respectively. The early cement occurs by crystal coat to adhered with grains or sediment, whereas the sparite or secondary cements look larger and fill the remained voids of the dissolved grains. These features are mostly observed in all samples that contain fossil grains, such as echinoid and foraminifera.

5.3 *Brittle fracturing*

It takes place with the continuous increase of the overburden pressure (Tucker, 2001). This may open empty space, which would be filled by newly precipitated materials. This feature is mostly observed within the samples contain algal grains

5.4 *Neomorphism*

It is applied when the processes made of the older crystals to be gradually consumed and their place simultaneously occupied by new crystals of the same mineral or could be of polymorph (Folk, 1965). This feature is observed mainly in the mudstone lithofacies.

5.5 *Dolomitization*

It is a major alteration process in carbonate diagenesis formed as a precipitate in near-surface and burial environments. This feature is mainly observed in the mudstone lithofacies.

The history and timing of these diagenetic features depend on the observed features, which are linked to their occurrence and development and the disturbance of the former feature by the younger. The timing of the above processes that acted on the Algal Limestone Member of Al Bayda Formation is shown in Fig. 3. Dissolution of fossils grains appears to form in the early stage of the diagenetic process and predates all other processes. This is evidenced by the presence of a remarkable amount of dissolved fossils (voids) fragments (Fig. 4). These dissolved fossil grains were originally composed of unstable aragonitic and/or high magnesium calcitic shells. This is followed by fracturing that mostly affecting the algal grains (Fig. 5). Few fractures and most voids are filled later with pyrite minerals in the forms of spherical aggregates known as framboids (Fig. 6). Early cement of syntaxial overgrowth type surrounding partially dissolved echinoid grain and equant cement type at the wall of some voids indicate that cement postdates the grain dissolution (Fig. 7). Secondary cement, on the other hand, is present as sparite filling the space between the grains and voids (Fig. 8). This indicates that secondary cement postdates the earlier diagenetic features. Neomorphism seems to affect the muddy matrix in a later stage. This appears as the micritic fine materials were recrystallized by coarser pseudospar-microspar in a process known as aggrading neomorphism (Fig. 9). Dolomite rhombic crystals formed as sucrosic texture have been observed at some places, indicating a later stage (Fig. 10).

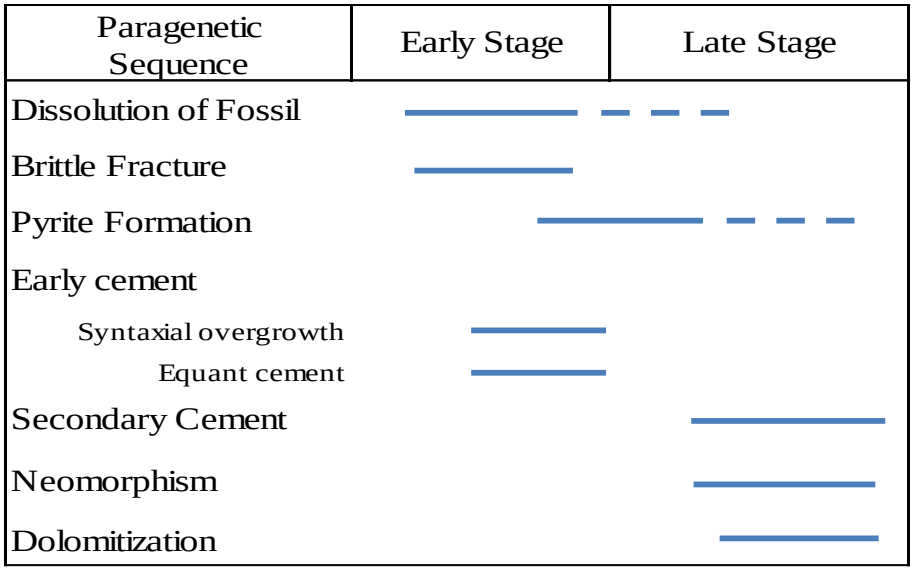


Fig. 3. Sequence of paragenetic processes observed within the major lithofacies of the Algal Limestone Member.

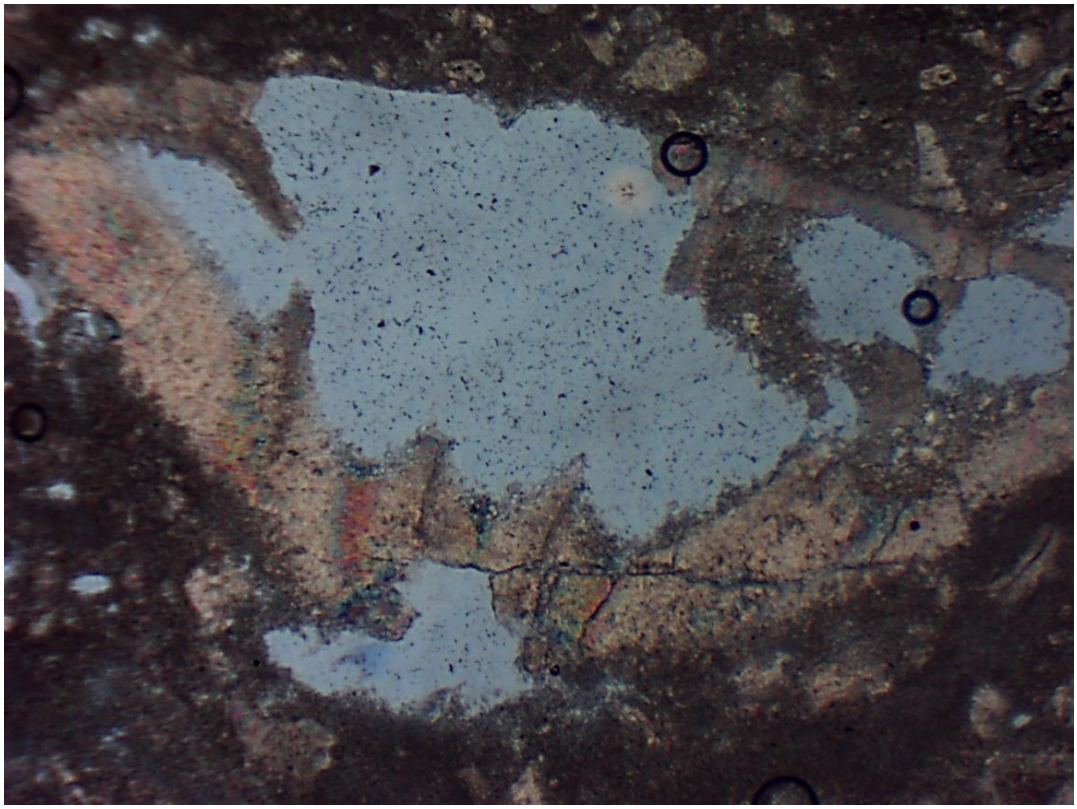


Fig. 4. Thin section photomicrograph showing partial dissolution of foraminifera fossil (10x-XPL).

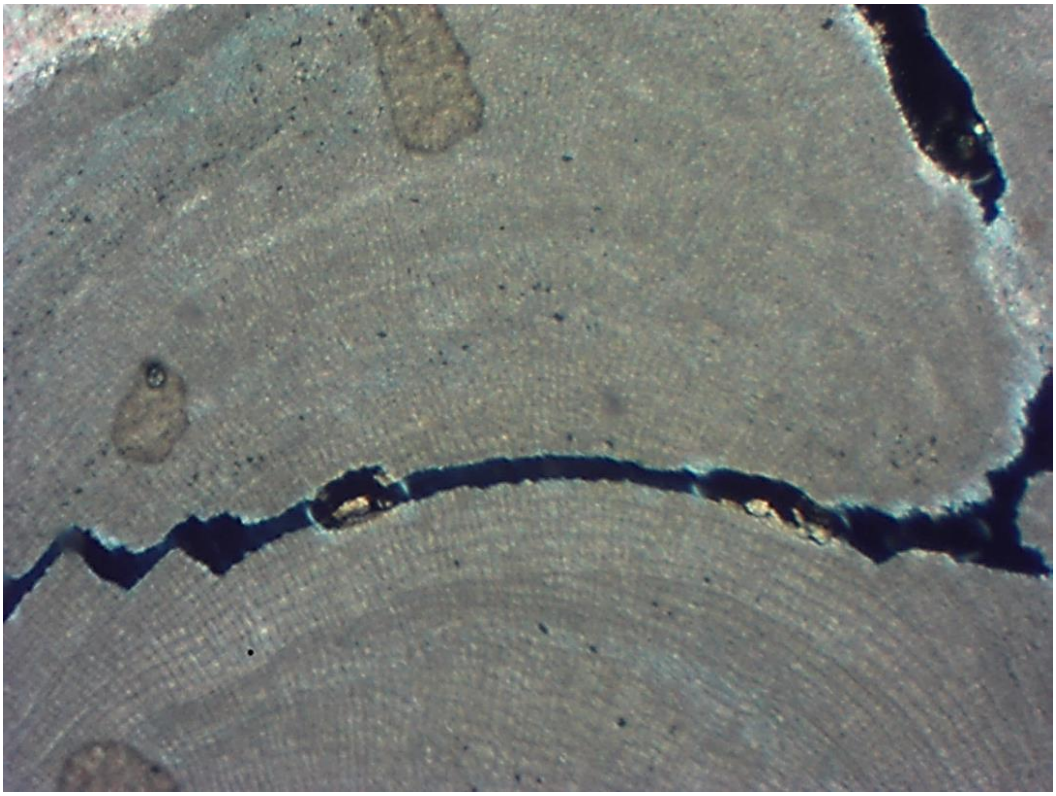


Fig. 5. Thin section photomicrograph showing a fractured algal grain (10x-XPL).

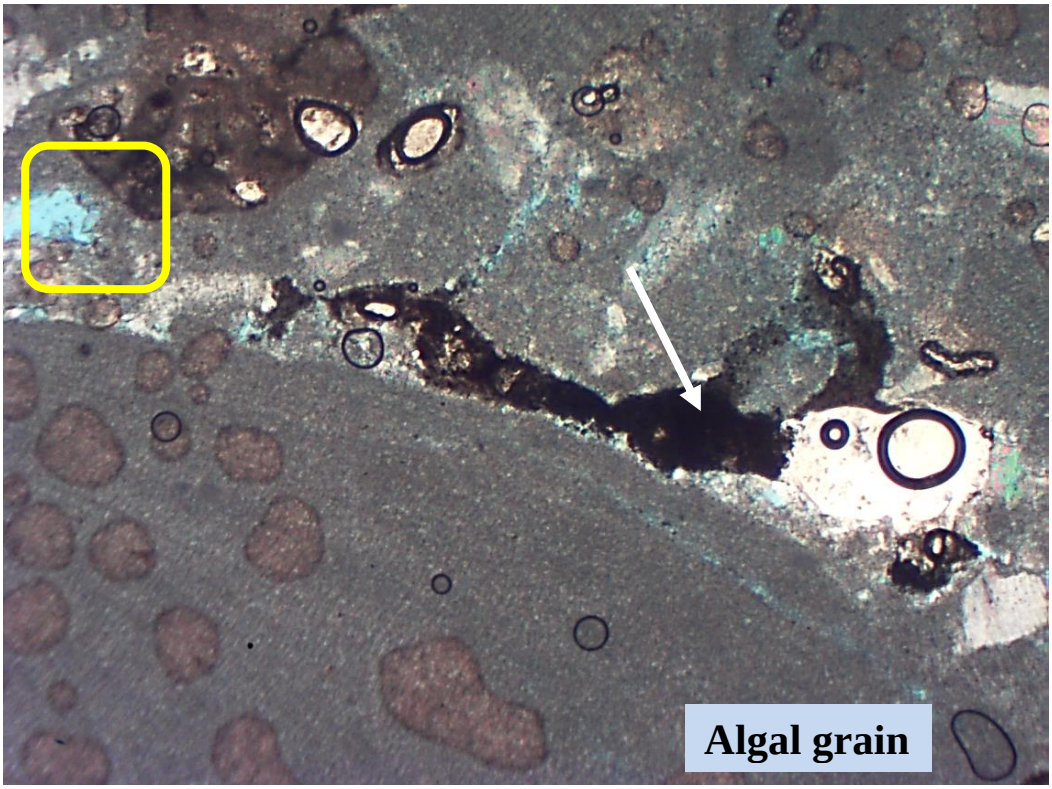
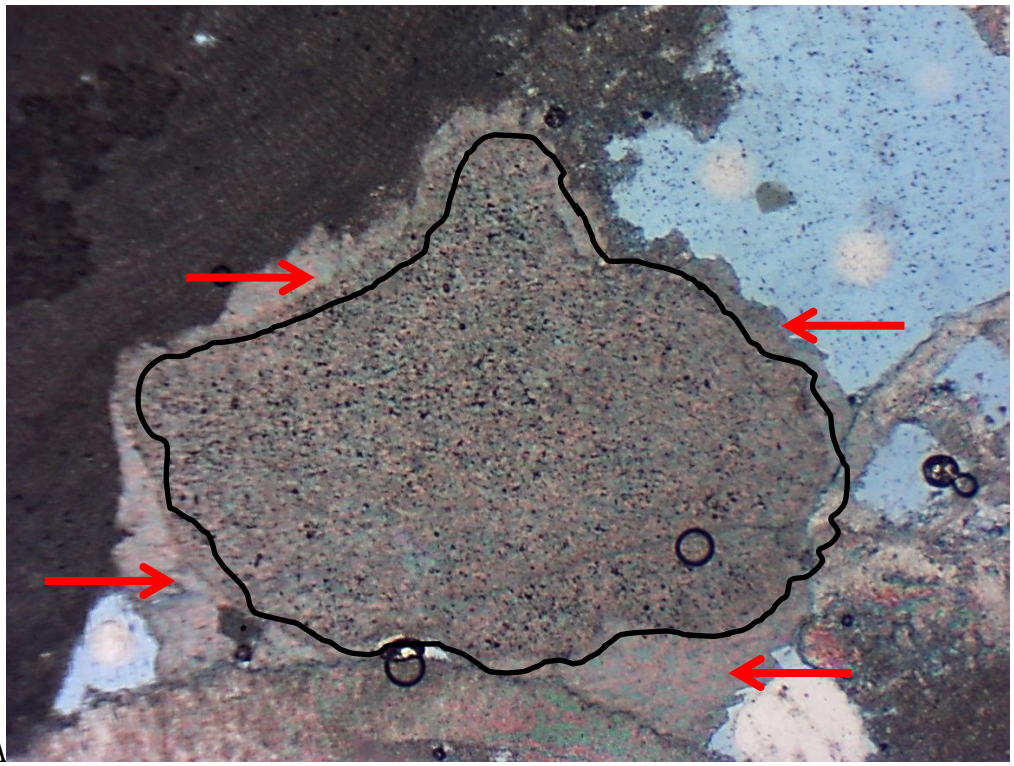
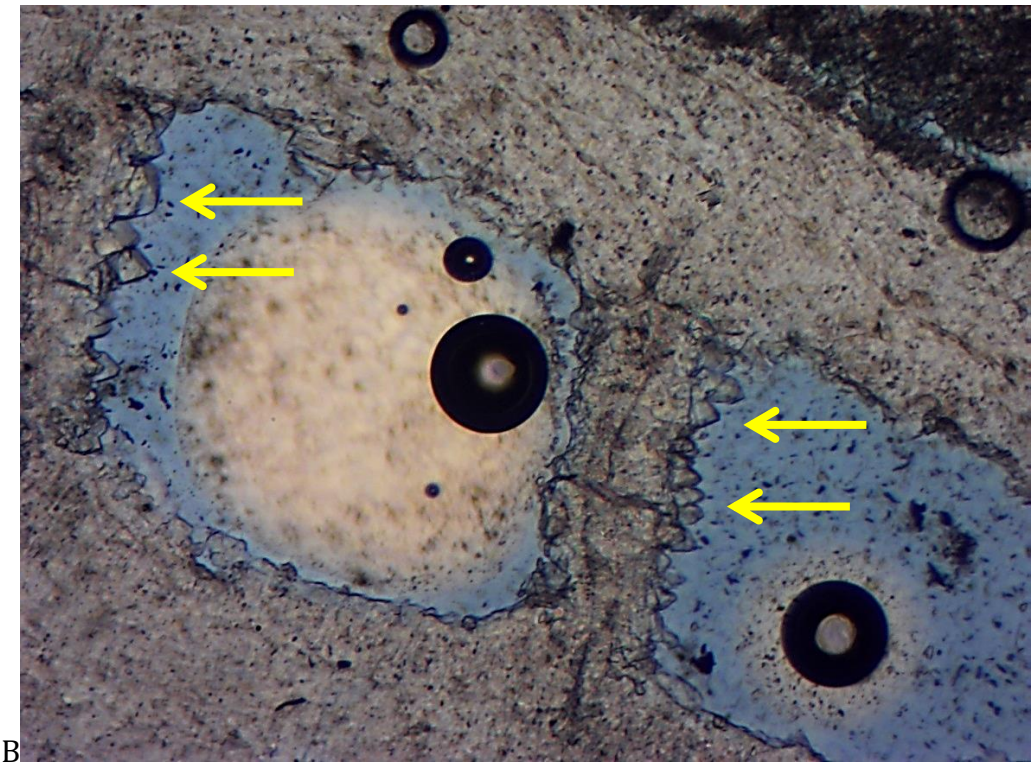


Fig. 6. Pyrite formation (white arrow) fills pore space around algae fossil grain, other porosity is in blue epoxy (yellow square) (10x-PPL).



A



B

Fig. 7. Thin section photomicrographs showing (A) echinoid fragment (black line) surrounded by syntaxial overgrowth calcite cement (red arrows) (10x-XPL). (B) an early equant calcite cement (yellow arrows) lining the foraminiferal chambers (10x-XPL).

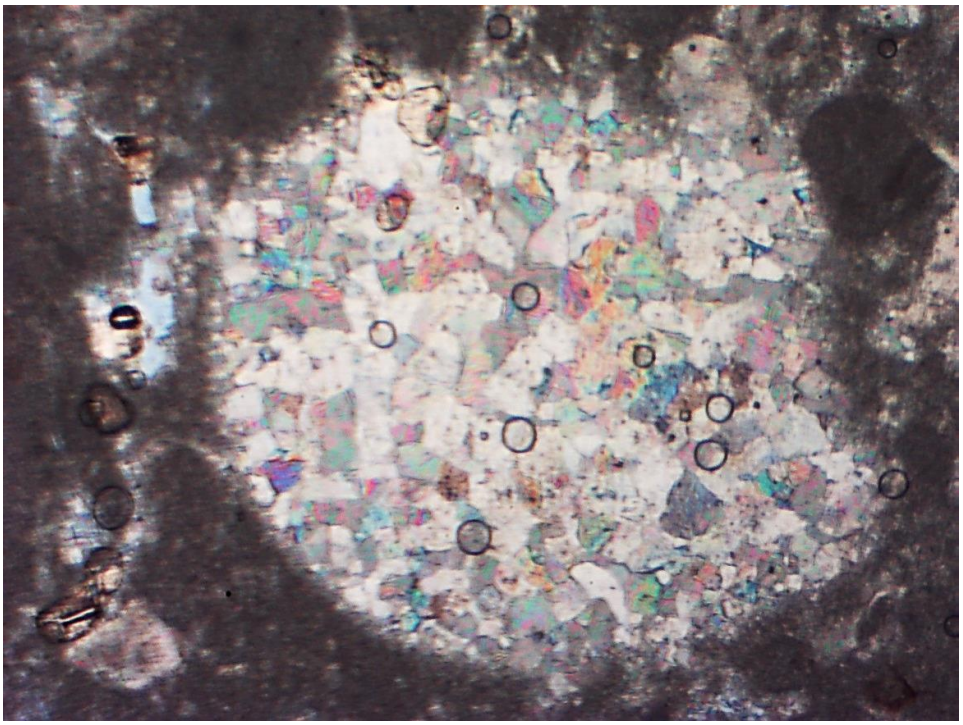


Fig. 8. Thin section photomicrograph showing secondary cement filling an opened pore space (4x PPL)

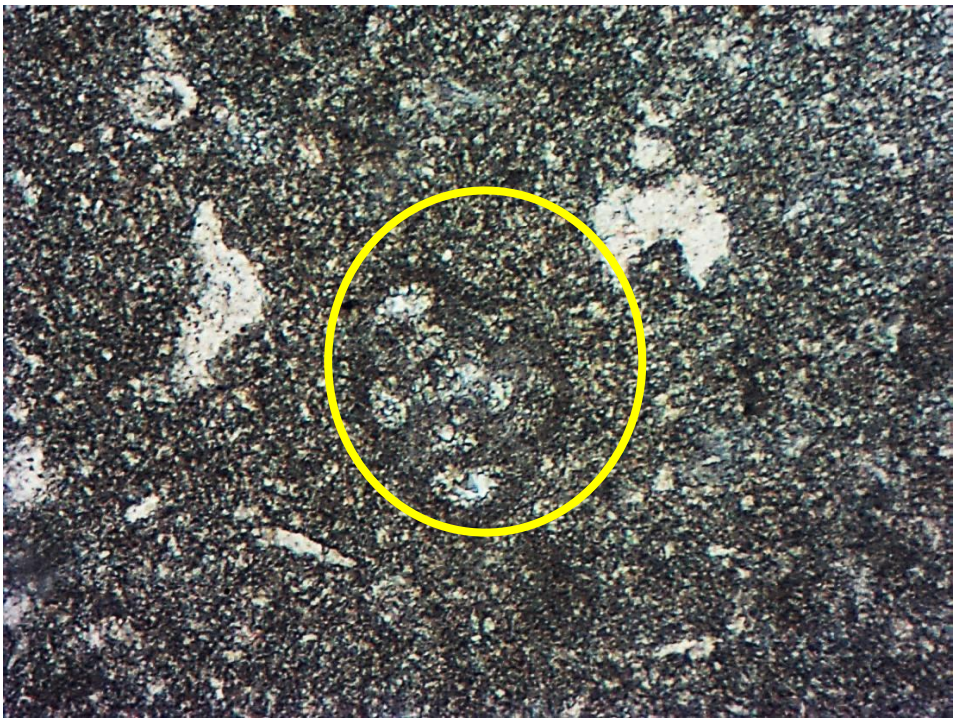


Fig. 9. Thin section photomicrograph showing microspar matrix formed due to aggrading neomorphic process of mudstone. Note: the foraminifera miliolid-test (yellow circle (10x-XPL).

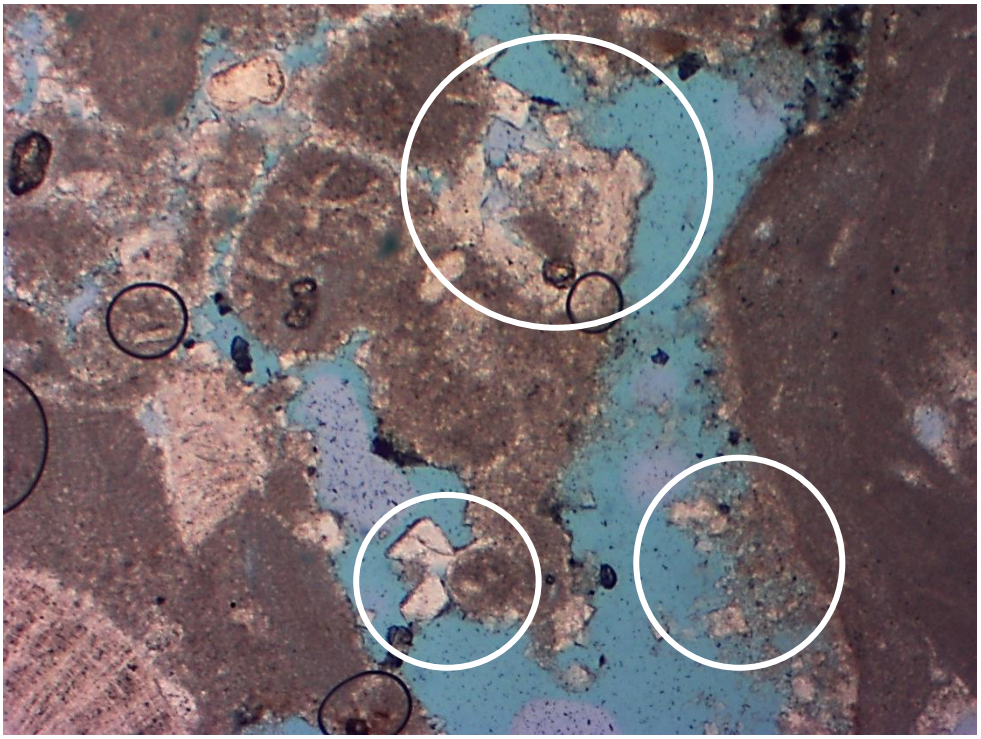


Fig. 10 Thin section photomicrograph showing dolomite crystals (white circles) formed at later stage inside a pose space(4x-PPL).

6. Discussion

6.1 Diagenetic and Environmental Interpretation

Choquette and Pray (1970) summarized the diagenetic environment of the sedimentary rocks into three regimes (phases); shallow or near-surface environment, burial environment, and during uplift or unconformity related environment. The Algal Limestone Member of the Al Bayda Formation was likely deposited within a shallow marine or near-surface environment that was dominated during Lower Oligocene (Fig. 11). Division of the diagenetic environments shows that each environment is characterized by the presence of different received diagenetic processes that can be distinguished by petrographic study. This is due to the variability of the percolatedsolution, sediments position and the accessibility of the solution to attain the sediments, hence, are shown with distinguishable features. Cement developments on the other hand depend entirely on the environment of cementation, adding to the texture of the sediment, which are playing an important role in the establishment of the diagenetic environment. Furthermore, the morphology and mineralogy of the pore-filling cement crystals in carbonate rocks may yield information concerning the environment of cementation, wherever it forms, cement can be observed and distinguished from the other fabrics. It is obvious that the Algal Limestone Member of Al Bayda Formation has suffered extensive diagenetic and chemical processes. The solution that penetrated the sediment must have a concentration of HCO_3 in charge of the occurrence of the grain dissolution of the bioclastic grains and other minerals that have an unstable component of $CaCO_3$. It seems also selectively affected the lime mud to leach out, forming pore space. The dissolution process offers the development of pyrite mineral (FeS_2) that comes from bacterial reduction of dissolved sulfate in the pore water (Tucker, 2001). More effective was the burial stage after the reducing environment and bacterial activity, which acted to exceed the compaction and fracturing of the rock constituents. These processes are considered pre-cementation, including pyritization and grain dissolution. However, early cementation of microcrystalline calcite is present in the early-stage process during the basinal environment in the phreatic zone (Tucker, 2001) whereas secondary cementation is present in the late-stage process during burial and/or uplift. The dolomite formation in the coralline algae of Algal Limestone Member of Al Bayda Formation formed possibly due to the seasonal temperature variation or the influence of the metabolism of the algae, thus the algae as it grows may contain dolomite nuclei that could be enlarged by diffusion of the originally adsorbed Mg ions in the structure and/or by addition of Mg ions from the seawater. Similar features were recognized in the Australian algal reef complex (Wolf, 1963).

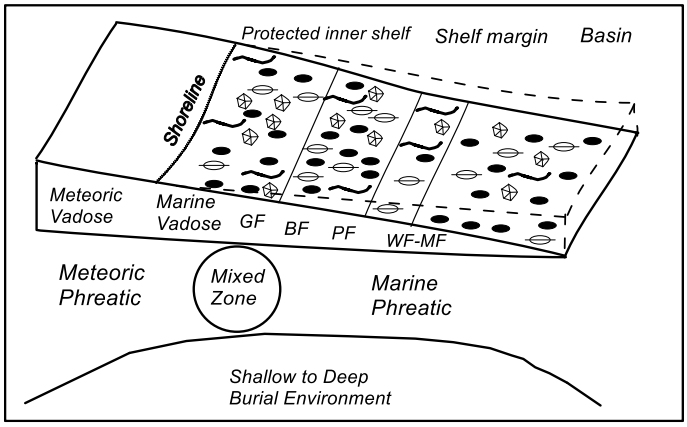


Fig. 11. Schematic diagram showing depositional environment that was dominated during the deposition of the Algal Limestone Member in the study area. GF-Grainstone Lithofacies; BF-Boundstone Lithofacies; PF-Packstone Lithofacies; WF-Wackestone Lithofacies; MF-Mudstone Lithofacies (Modified from Hassan and Muftah, 2008).

6.2 Diagenetic History

The burial history of the sequence (Fig. 12) has been reconstructed from thickness data in Klen (1974), Röhlich (1974), and Zert (1974). It is probable that these overestimate the local thickness of the overburden that was once present at the study area. However, no decompaction of units has been attempted and this may affect thicknesses and lead to an underestimate of thickness in parts of the diagram. The burial history indicates that the Algal Limestone Member of Al Bayda Formation would have undergone prolonged but persistent burial. It would have been fairly rapidly removed from the shallow diagenetic environment to deeper zones in which burial diagenesis could have taken place, and in which it

resided for a few million years. The diagenesis of this carbonate member within the studied sequence shows some broad similarities and can be clearly divided into two suites, early and later suites of processes (Fig. 3). The early suite of processes includes grain dissolution, pyrite formation, precipitation of early cement, and fracture. Slight variations in the temporal sequence of these processes suggest that they are closely related in time. Pyrite formation (Berner, 1970) clearly takes place soon after deposition or at very shallow depths. The other processes are also consistent with a shallow diagenetic environment, though grain fracturing may indicate some build-up of overburden pressures. The stable non-ferroan calcite early cements indicate a meteoric influence, could be of marine phreatic origin. Interestingly no hardgrounds or early vadose cements have been detected within the studied samples. The later diagenetic suite is dominated by the precipitation of a pore filling sparry calcite cement and by extensive neomorphism. The later processes clearly affected the micrite. These processes are interpreted as taking place in a deeper burial environment.

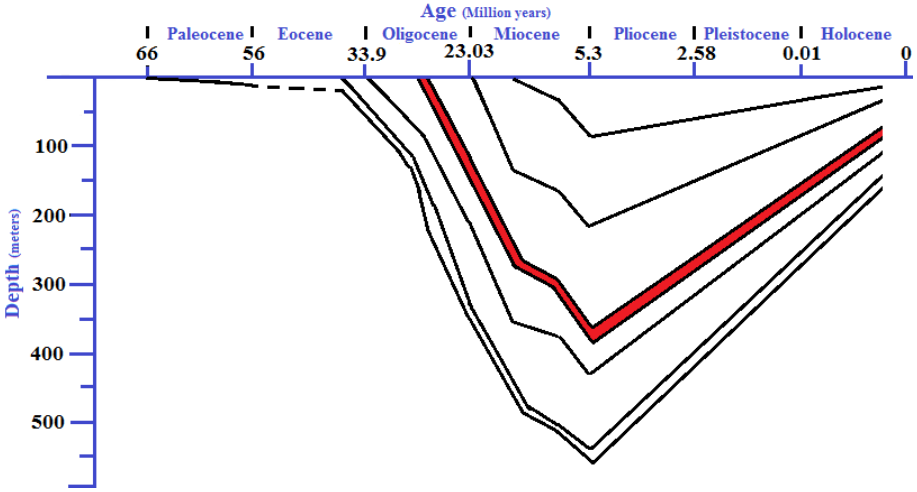


Fig. 12. Diagram illustrating the history of subsidence and uplift of the Paleocene to Miocene sediments. The red-lined period (~33 - 28 my) represents the Middle-Upper Oligocene. Time interval is based on Geological Time Scale by Cohen et al. (2013).

7. Conclusions

- The Algal Limestone Member of Al Bayda Formation at Deryanah-Al Abyar road cut, Al Wattayatt Area, Al Jabal Al Akhdar, has been divided into five major lithofacies based on the macro and microscopic study. They are wackestone; boundstone, grainstone; mudstone-wackestone, and packstone lithofacies.
- The Algal Limestone Member of Al Bayda Formation has suffered extensive diagenetic and chemical processes.
- The diagenetic analysis applied to this member reveals two stages of paragenetic processes. Early-stage includes dissolution, brittle fracture, recrystallization, pyritization, and early cement. Late-stage includes secondary cement, neomorphism, and dolomitization.
- Dissolution of fauna and other minerals that have an unstable component of $CaCO_3$ indicates penetration of solution enriched with HCO_3. The formation of pyrite mineral (FeS_2) comes from the metabolism of the anaerobic bacterial activity under the reduced condition
- Early cementation of crystalline calcite is present in early-stage process during basinal environment in phreatic zone, whereas secondary cementation is present in late-stage process during burial and/or uplift.
- Several processes could be responsible for dolomite formation. It may be formed due to; i) variation in the seasonal temperature; ii) the algal metabolism; iii) by addition of Mg-ions from the seawater.

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