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Petrographic and Geochemical Evidence for Paragenetic Sequence Interpretation of the Lower Cretaceous Limestones in the Eastern Binalood Mountain Range, NE Iran

A., Mahboubi[1], R., Moussavi-Harami[1], V., Yahya-Sheibani[1] M., Najafi[1] and L., Gonzalez[2]

[1]*Department of Geology, Faculty of Science, Ferdowsi University of Mashhad, Mashhad Iran 91775-1436*

[2]*Department of Geoscience, The University of Iowa, Iowa City, IA, USA, Current address, Department of Geology, The University of Kansas, Lawrence, KS, USA 66045-7613*

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Abstract

The Lower Cretaceous sediments (mainly limestones with a few horizons of sandstones and conglomerates) are present south of Mashhad along the Binalood Mountain range. Petrographic studies reveal that the carbonate rocks were deposited on a shallow carbonate platform of ramp type including open-marine, shoal, lagoon and tidal-flat facies belts. Diagenetic processes include compaction, cementation, micritization, replacement (silicification), neomorphism and fracturing.

Oxygen and carbon isotope signatures of the limestones range from -11.2 to -2.1‰ and 0.2 to 4.3‰ PDB respectively. This variation reveals meteoric and burial diagenetic environments. Decreasing trend in the value of oxygen isotopes with increasing the amount of Fe and Mn as well as decreasing Na and Sr also indicate that these rocks may have been exposed to the meteoric diagenetic environment too. The calculated temperature for the ambient water in which calcite was deposited is about 26^{o} C.

Keywords: *Geochemistry, Lower Cretaceous, Stable isotopic, Binalood.*

Introduction

The Binalood Mountain range in NE Iran (Fig. 1) extends sinuosoidaly in an east-west direction and contains Palaeozoic, Mesozoic (Jurassic and Cretaceous) and Cenozoic successions. The strata in the Binalood region are folded and faulted as a result of collision between the Iran and Turan plates in NE Iran (Alavi, 1991 and 1992).

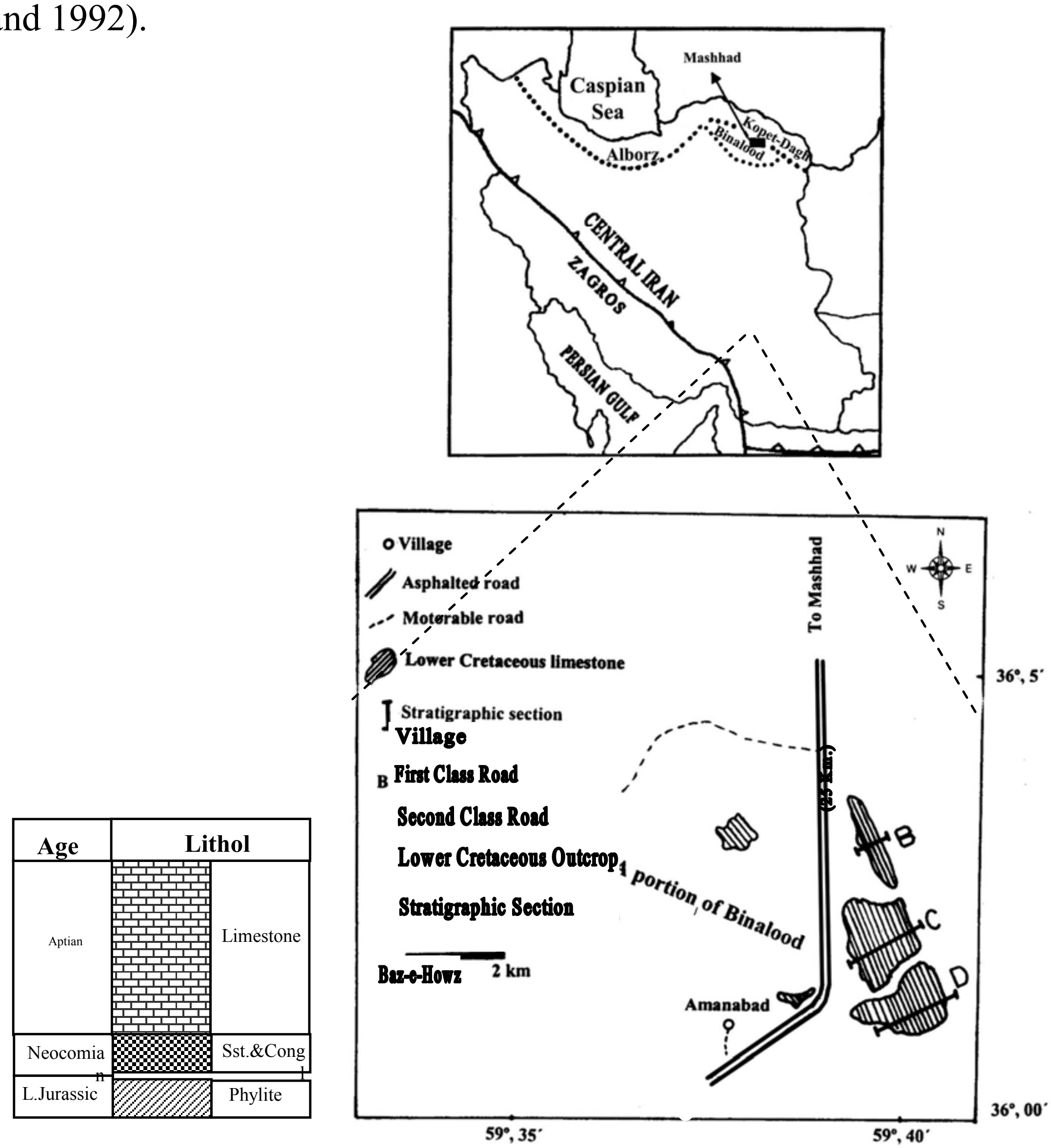

Figure 1. Location map and simplified stratigraphic section of the study area in NE Iran. Location of measured stratigraphic sections are labeled by A, B, C and D.

The Lower Cretaceous sediments in the study area with 30 to 85 meters are mainly composed of skeletal and non-skeletal grainstone to mudstone with interbedded thin shale beds. Siliciclastics (sandstones and conglomerates) are present in the lower part and skeletal and non-skeletal (oolitic) limestones in the upper part of the sequence. The sandstone and conglomerate units seem to be equivalent to the Neocomian Shurijeh Formation, whereas the limestones are likely an equivalent of the Tirgan Formation in the Kopet-Dagh basin, north of the study area.

In this paper we focus on the diagenetic history of the Lower Cretaceous limestones by using petrographic and geochemical data.

Material and Methods

We used a data-base of 160 samples collected from four measured stratigraphic sections through the study area (Fig. 1). Thin sections were stained with an Alizarin Red and Potassium Ferricyanide mixture following the procedure outlined by Dickson (1966). Limestones and siliciclastics were classified after Dunham (1962) and Folk (1980). 50 polished thin sections with thickness of 80-100 μm were examined under luminescent light (based on Miller, 1988; Marshal, 1988; Frank et al., 1995; Budd et al., 2000). Cathodoluminescence studies were conducted with a Technosynosyn Cold CL (Model 8200 MK3) at 12 KV and 160-195 μA.

21 samples were selected for carbon and oxygen isotopes analysis. Samples for isotope measurements were extracted with a dental drill and a 500 μm bit from polished slabs. We selected about 0.2 mg of each sample, including bioclasts, micrite, cement and ooids. Stable isotope analysis was performed using a Finnigan MAT 251-KIEL at the University of Iowa. $\delta^{18}O$ and $\delta^{13}C$ values are reported relative to PDB. In addition, 17 samples of micrite, ooids and bioclasts were analyzed by atomic absorption spectrophotometry to determine their Sr, Na, Mn and Fe contents, using a Shimatzu AA-670/670 G at the Ferdowsi University of Mashhad.

Description of Lithofacies and their Environmental Interpretation

Based on thin section observations, four lithofacies were identified:

Lithofacies A: Skeletal packstone/wackestone

This lithofacies is brown-grey and medium to thin bedded in the field. In thin section it shows abundant fine to medium-grained skeletal debris including bryozoans, brachiopods and echinoderms that are abraded, bored and partially micritized. The great number of debris of stenohaline organisms together with the abundant lime mud matrix suggests that lithofacies A was deposited below fair-weather wave base in open-marine environment (outer ramp).

Lithofacies B: Skeletal-ooid-intraclast grainstone

These coarse-grained grainstones are grey to light tan-grey at the surface and commonly show cross-bedding and ripple cross-lamination. In thin section they contain abundant ooids, intraclasts and skeletal debris (including red algae, orbitolinids, echinoderms and bryozoans); locally peloids are common. The size of ooids ranges from 0.15-1 mm and they have concentric and radial fabrics. The nuclei of ooids consist of bivalve and echinoderm fragments, as well as detrital quartz. Intraclasts vary in size, ranging in diameter from 0.4 to about 1.8 mm, and are subangular to subrounded. They are composed of a variety of skeletal debris and non-skeletal grains. In some cases they are completely composed of lime mud. The interstitial material is sparite with blocky, granular and in some cases acicular morphology. Cross-bedding and coarse grain-size as well as abundant ooids and intraclasts suggest that lithofacies B was deposited in a mobile sand-shoal environment in a shallow-water setting (middle ramp).

Lithofacies C: Peloidal-skeletal packstone/ wackestone/mudstone

This lithofacies consists of thin-bedded, light grey to tan limestones with silt to very fine sand-sized components and mm-scale laminae. Under the microscope peloids are common and skeletal grains include green algae (mainly dasycladae), miliolids, textularinids, orbitolinids and gastropods. Ooids, oncoids and some intraclasts (similar to lithofacies B) are also present.

The thin bedding, abundant lime mud and peloids and diverse lagoonal type biota suggest that the formation of this lithofacies took place in a relatively shallow, semi-restricted, low-energy lagoon environment.

Lithofacies D: Unfossiliferous lime mudstone

This grey to light tan-grey and thin-bedded lithofacies consists of unfossiliferous micrite. In a few samples terrigenous quartz is common. Predominant lime mud without marine biota suggests that this lithofacies was deposited in supratidal and high intertidal environments.

Summarizing the above; based on field and laboratory observations we conclude that these Lower Cretaceous limestones were deposited on a shallow-water platform of a ramp-type, in accordance with the models presented by Read (1985).

Diagenetic History

The diagenetic history of the Lower Cretaceous limestones in the eastern Binalood is inferred from the results of petrographic and geochemical investigations.

Petrographic evidence

The most important diagenetic processes which affected the limestones are compaction, cementation, micritization, neomorphism, silicification and fracturing. These processes are briefly discussed below.

Compaction: Compaction is the most common process which operated in the limestones. Gradual increase of overburden (lithostatic and hydrostatic) led to decreasing thickness, increasing long grain contact to point contact and finally pressure dissolution and formation of stylolites (Figs. 2A and 2B). This process significantly reduced the intergranular porosity and permeability, especially in the grainstones.

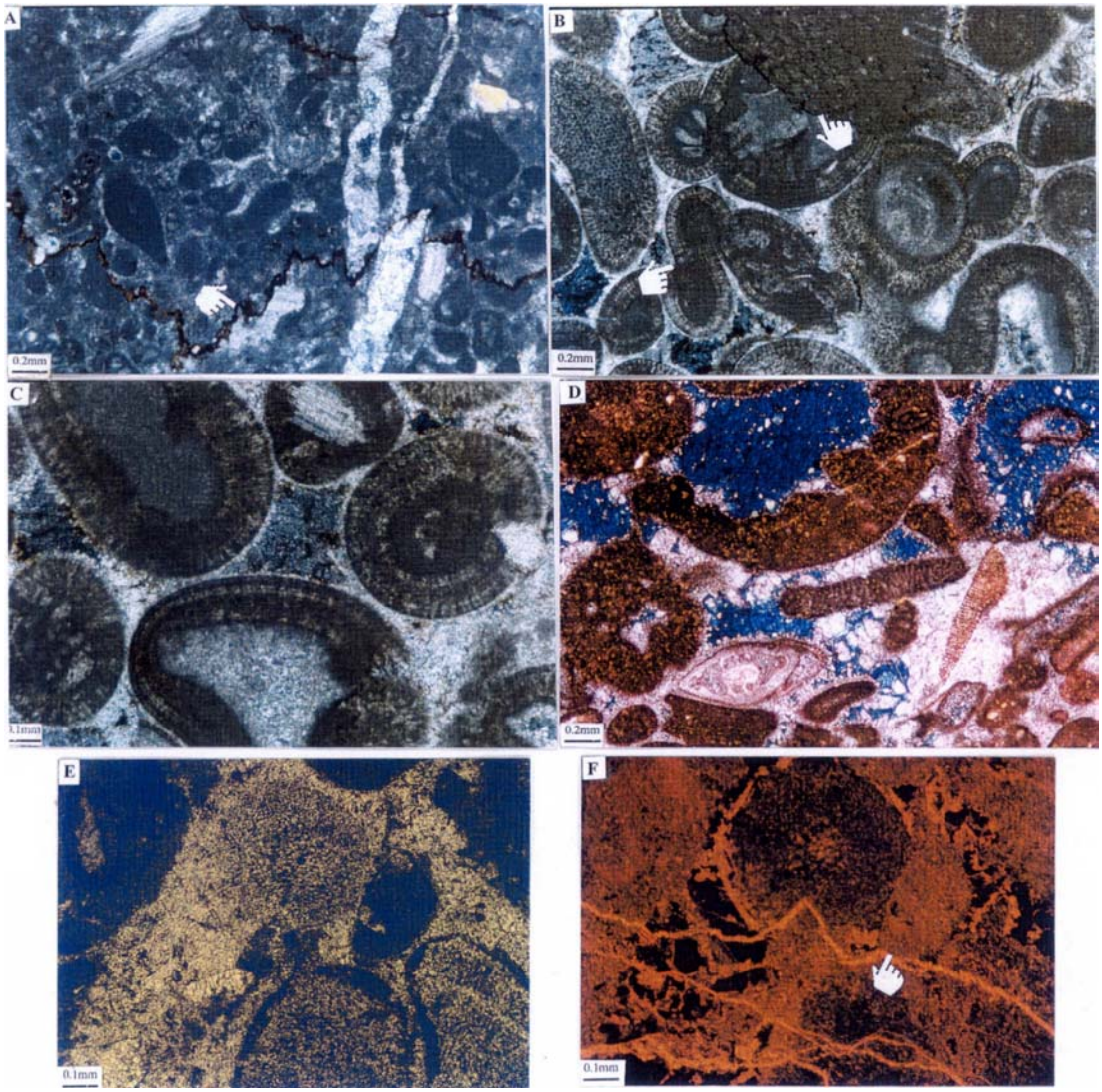

Figure 2. Photomicrographs are showing features of diagenetic processes. (A) Stylolitization in packstone lithofacies (see finger) that formed during the chemical compaction (XPL). (B) A nearly close packing in ooid grainstone lithofacies. It shows spastolith (left side), sutured and concavo-convex contacts (upper left) (XPL). (C) Two generations of cement in ooid grainstone lithofacies. Isopachous fibrous cements is succeeded by cement of blocky fabric (XPL). (D) Skeletal grainstone stained by Alizarin Red and Potassium Ferricyanide mixture solution. It shows non-ferroen red isopachous fibrous cement and ferroen blue equant cement (XPL). (E) Syntaxial overgrowth rim cement in skeletal grainstone lithofacies (PPL). (F) The same view as E under CL. It shows bright luminescence in rim cement and filled microfracture that can not be seen under polarized light.

Cementation: Cements in the grainstone lithofacies are a variety of types, as a result of precipitation in several different diagenetic environments. Fabrics observed are as follows:

I) Isopachous fibrous calcite represents the first generation of cement around the skeletal and non-skeletal grains (Fig. 2C). Length-to-width ratios of crystals were generally greater than 6/1 but occasionally less than this values.

II) A blocky calcite cement of single crystal is commonly occupies an entire pore and contains no inclusions (Fig. 2C). These poikilotopic cements range in size from 0.04 to 0.35 mm.

III) The equant calcite is a second generation of cement precipitated after the fibrous calcite (Fig. 2D). Where the first generation fibrous cement is absent, the equant cement precipitated directly around the skeletal and non-skeletal grains.

IV) Syntaxial overgrowth rim cement formed on unmicritized surfaces of echinoderm fragments and is in optical continuity with the monocrystal-substrate (Figs. 2E and 2F).

Micritization: Micritization is common in bioclastic grain-supported lithofacies. Identification of micrite envelopes is very easy in bioclastic grainstones (Fig. 3A) but very difficult in mud-supported lithofacies.

Aggrading neomorphism: Aggrading neomorphism is observed in lime mudstone lithofacies. As a result of this process, lime-mud matrix changed gradationally to microspar and spar (Fig.3B).

Replacement: Silicification is the only type of replacement observed. Chert and microcrystalline quartz replaced part of the bioclasts such as bivalves and brachiopods (Fig. 3C). Interbedded shales within the limestones may be the source of silica for replacement as has been suggested by several authors (McBride, 1989; Hesse, 1989; Bjorlykke and Egeber, 1993).

Fracturing: Postdepositional events caused fracturing in some of the limestone beds. Later the fractures were filled with calcite (Fig. 3D). These fracture fills were examined with cathodoluminescence and showed several thin zones of bright-dull-dark luminescence (Figs. 3E and 3F), indicating fluctuation in the redox conditions of pore fluid during precipitation of calcite.

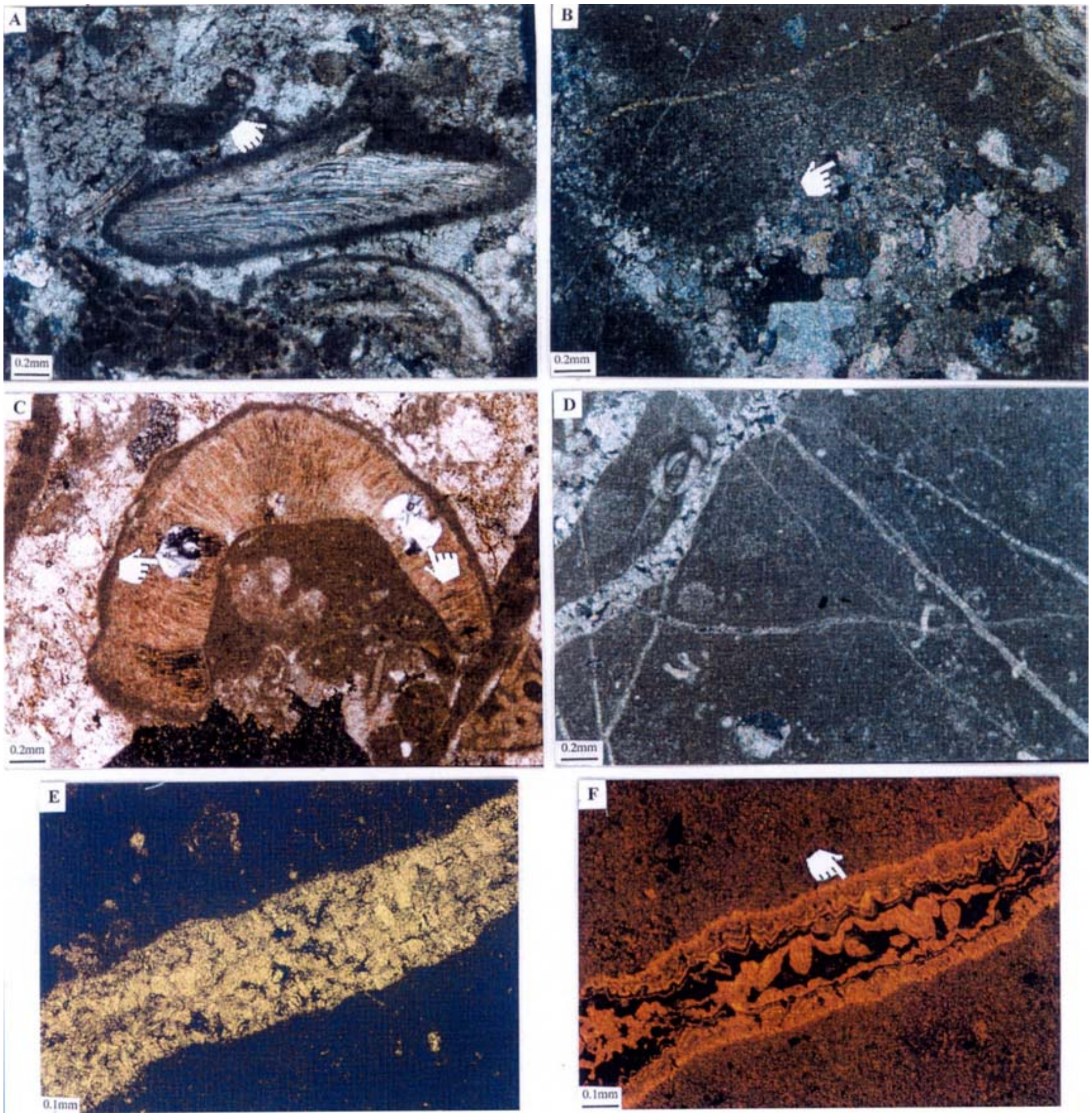

Figure 3. A) Micrite envelope around brachiopod debris in a skeletal grainstone lithofacies (XPL). B) Lime mudstone lithofacies that changed gradationally to microspar and spar (aggrading neomorphism) (XPL). C) Silicification of a large bioclast in skeletal grainstone lithofacies (XPL). D) Fracturing in wackestone-lime mudstone lithofacies. These fractures are filled by secondary calcite cement (XPL). E) Close up view of a fracture filled by calcite in lime mudstone lithofacies (PPL). F) The same view as E under CL. Several thin zonations of bright-dull-dark luminescence reveal that they have formed under different oxidation and reduction conditions.

Geochemical evidence

The Lower Cretaceous limestones in the study area were analyzed for their isotopic and trace elemental contents. These are very useful methods for the interpretation of diagenetic history and are used by many workers (e.g. Brand and Veizer, 1980; Morse and Mackenzie, 1990; Tucker, 1993; Rao, 1996 and 1997; Simo and Lohmann, 2000; Rasser and Fenninger, 2002; Mahboubi et al., 2002).

Results

Isotopic data

The oxygen and carbon isotopic data from constituents are shown in Table 1. The ranges in oxygen and carbon isotopic composition are -2.1 to -11.2‰ PDB and 0.2 to 4.3‰PDB in average respectively. Oxygen isotope ratios of constituents are: for oyster shells ranges from -2.1 to -2.4‰, for ooids from -5.0 to -5.7‰, for early cements from -6.3 to-11.2‰ and for micrite from -4.2 to 6.9‰ PDB. Carbon isotope ratios for these components range from 3.8 to 4.3‰, 2.85 to 2.9‰, 0.2 to 2.7‰ and 0.5 to 2.7‰ PDB respectively.

Trace elemental data

The Na, Fe, Mn and Sr data for some constituents are shown in Table 2 and can be summarized as follows:

a) Oyster shells: The average Na, Fe, Mn and Sr contents are 10000, 3630, 700 and 1383 ppm respectively.

b) Micrite: The Na content ranges from 390 to 7400 ppm (average ~ 3400 ppm); the Fe ranges from 3115 to 6990 ppm (average ~ 5000 ppm); the Mn ranges from 450 to 1300 ppm (average ~ 700 ppm); and the Sr ranges from 355 to 1014 ppm (average ~ 700 ppm).

c) Ooids: the Na content ranges from 5100 to 8500 ppm (average ~ 6400 ppm); the Fe ranges from 5350 to 12900 ppm (average ~ 7500 ppm); the Mn ranges from 700 to 1200 ppm (average ~ 950 ppm); and Sr ranges from 920 to 1096 ppm (average ~1000 ppm).

Table 1- Results of carbon and oxygen stable isotope analysis for the Lower Cretaceous limestone samples. Results are expressed in ‰ relative to the PDB standard.

Sample No.	Constituent	$\delta^{18}O$	$\delta^{13}C$
5a.01	micrite	-5	2.1
5c.01	micrite	-5.3	2.2
C8.01	micrite	-4.9	2.4
C9.01	micrite	-6.9	0. 5
5d.01	micrite	-4.55	2.1
10b.01	micrite	-4.2	2.6
15f.01	micrite	-5.6	2.7
D1.01	micrite	-6.3	1.8
5b.01	micrite	-5.2	2.4
5e.01	micrite	-4.7	1.8
7a.01	micrite	-5.65	2
10a.01	micrite	-6.5	1.7
C10.01	micrite	-4.6	2.7
5d.02	oyster	-2.1	4.3
10b.02	oyster	-2.4	3.8
15a.01	early cement	-6.3	2.7
15b.01	early cement	-10.05	1.5
15c.01	early cement	-11.2	0.2
15a.02	ooid	-5.7	2.85
15b.02	ooid	-4.7	2.80
15c.02	ooid	-5	2.9

Discussion

Oxygen and carbon isotopes variations

Diagenesis often results in a small but variable many to more negative $\delta^{18}O$ values of marine carbonate (e.g. Land, 1970; Allan and Matthews, 1977; Morse and Mackenzie, 1990; Rao, 1997). This shift occurs because cementation and recrystallization often takes place in fluids depleted in ^{18}O with respect to sea water (e.g. meteoric water) or at elevated temperatures (burial conditions). Early marine diagenesis should result in little if any oxygen isotope shifts in carbonate sediments which precipitated at or near equilibrium with sea water.

The variations in $\delta^{18}O$ of constituents in the studied limestones show a more to quite negative values (-2.1 to -11.2‰ PDB). The skeletal and non-skeletal grains, as well as cements, are depleted in ^{18}O with respect to $CaCO_3$ in equilibrium with sea water. This shift could be the result of meteoric or burial diagenesis. In petrographic studies, we have seen both cements that are formed in meteoric conditions as well as compactional features that formed in burial stage. Therefore, we have concluded that these limestones may have been exposed to the meteoric and burial diagenetic environments.

Table 2 - Results of elemental analysis for limestone samples of the study area.

Sample No.	Constitute	Sr(ppm)	Mn(ppm)	Fe(ppm)	Na(ppm)
15f.01	micrite	355	800	5085	1600
5d.01	micrite	898	700	3115	4300
5b.01	micrite	610	700	6670	3900
5c.01	micrite	620	800	6610	4400
10a.01	micrite	598	1300	4210	800
5a.01	micrite	585	700	6990	420
C9.01	micrite	715	600	6500	630
C8.01	micrite	556	500	3470	1075
C10.01	micrite	746	450	3220	7400
D1.01	micrite	937	700	5530	390
5e.01	micrite	749	600	5790	6000
7a.01	micrite	802	700	4510	6000
10b.01	micrite	1014	600	3610	7400
15b.02	ooid	920	1000	5375	8500
15c.02	ooid	920	700	5350	5100
15a.02	ooid	1096	1200	12900	5600
5d.02	oyster	1383	700	3630	10000

Significant shifts in the carbon isotopic composition of marine carbonates during diagenesis can occur when the degradation of organic matter is the dominant process supplying dissolved carbonate species to diagenetic fluids (Allan and Wiggins, 1993; Rao, 1996 and others). The ranges in carbon isotopic composition for all components are between 0.2‰ to 4.3‰ PDB. This range is similar to Phanerozoic marine limestones that have been proposed by Veizer and Hoefs (1976), Veizer (1983) and Hoefs (1987). Therefore, there is no indication of biogenic source for carbon in these limestones because of the range of carbon isotope is mostly positive.

Trace elemental variations

Trace element contents in marine carbonate rocks are often used to estimate the extent of diagenetic alteration. For example, Veizer (1977), Brand and Veizer (1980), Veizer (1983) and Morse and Mackenzie (1990) have argued that Sr and Na contents of limestone should decrease and Mn and Fe content increase during progressive diagenetic alteration. These trends result (1) from large differences in the Sr/Ca, Na/Ca, Fe/Ca and Mn/Ca ratios between sea water and diagenetic fluids and (2) in the calcite-water distribution coefficients of these ions (Sr and Na have $D_c<1$ while Fe and Mn have $D_c>15$).

Variations in trace element concentrations of our samples revealed that with increasing effect of diagenetic waters, the amounts of trace elements (Mn, Fe, Sr and Na) have changed. These variations are similar to meteoric conditions, because with increasing infiltration of meteoric waters (with relatively high Fe and Mn and low Sr and Na concentrations), Fe and Mn are enriched but Sr and Na are depleted. These variations are correlated with $\delta^{18}O$ and $\delta^{13}C$ variations of shown in Figures 4 and 5. Fe and Mn are generally incorporated into carbonate minerals in a reduced state. This condition in the meteoric environment is probably related to the sustained reduction of abundant organic matter that is supported by positive correlation between Fe and Mn (Fig. 6). The CL zonation in the calcite cements (Fig. 7 and also see Figs. 2E and 2F) indicate fluctuation in redox conditions during cement precipitation.

Water temperature

For calculation of the temperature of the ambient water from which the calcite was precipitated, we have used formulas published by Shackleton and Kennet (1975), Friedman and O'Neil (1977) and Anderson and Arthur (1983). However, we found that the formula which has been given by Shackleton and Kennet (1975) can be fitted with our data.

$$T= 16.9-4.38 (\delta c-\delta w) + 0.10 (\delta c-\delta w)^2$$

Where T is temperature (in $^{\circ}C$), δc is the oxygen isotope value of calcite relative to PDB, and δw is the oxygen isotope value of water relative to SMOW.

By using -2.1‰ PDB as the least negative value, the temperature of the ambient water for the Lower Cretaceous limestone is about 26 $^{\circ}C$.

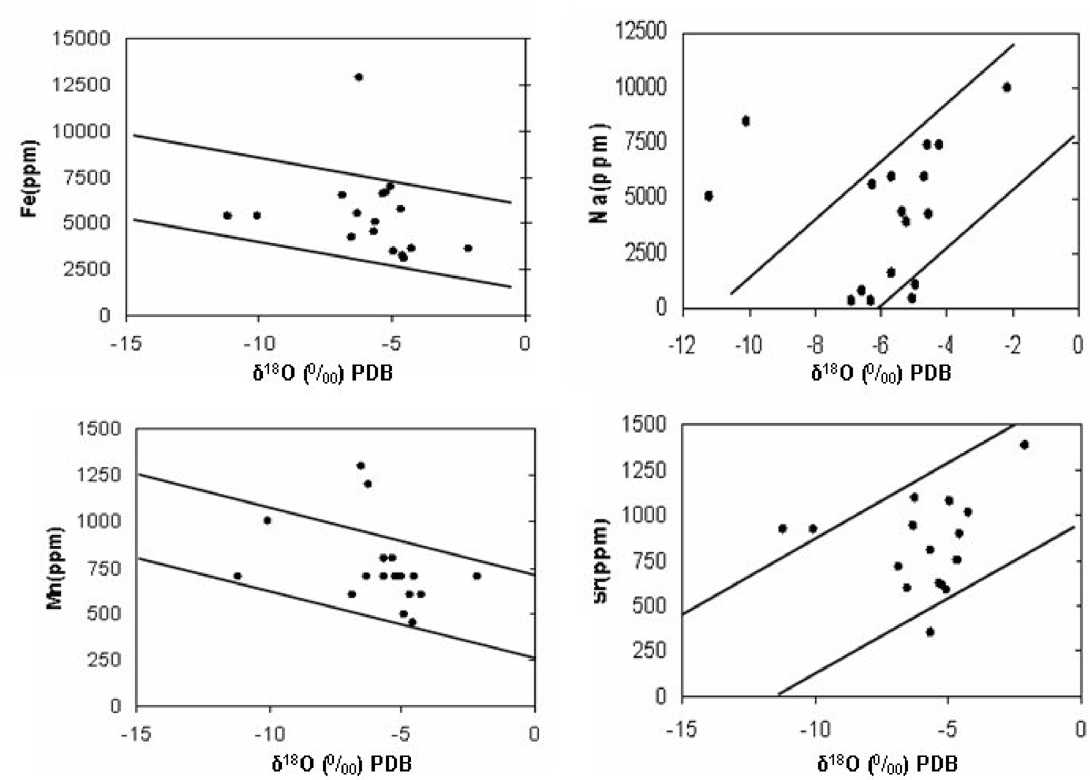

Figure 4 - Plots of $\delta^{18}O$ versus Mn, Na, Fe and Sr concentrations in the limestones of the study area indicating the influence of meteoric fluids.

Paragenetic Sequence

The interpretation of the paragenesis of the Lower Cretaceous limestone is based mainly on the evidence of diagenetic processes. These processes have taken place in four different stages including marine, meteoric, burial and uplift diagenetic environments. They are summarized in Figure 8 and discussed below.

Stage 1. *Marine phreatic environment*

Micrite envelope formation micritization and precipitation of isopachous cement are the most important processes that have taken place at this stage. Micritization mostly occurs in grainstone lithofacies. This process is followed by the precipitation of non-ferroen fibrous cement.

Stage 2. *Meteoric phreatic environment*

Cementation (including blocky, equant mosaic and syntaxial overgrowth cements) is the most common process in the meteoric phreatic environment. Blocky cement is commonly ferroan which is related to reducing conditions and higher Mn/Fe ratio. We have seen a bright-dull-dark zonation on this kind of cement under CL (see Fig. 7). Syntaxial overgrowth rim cement is formed around echinoderm bioclasts. Although these cement fabrics also form in burial conditions but we separated these two types of cement generation by compactional features in the study area. The second generation of cement is characterized by ^{18}O depletion, decreasing the amounts of Na and Sr as well as increasing Fe and Mn. Some dissolution and neomorphism are the other processes that are acted in most of the lime mudstone lithofacies.

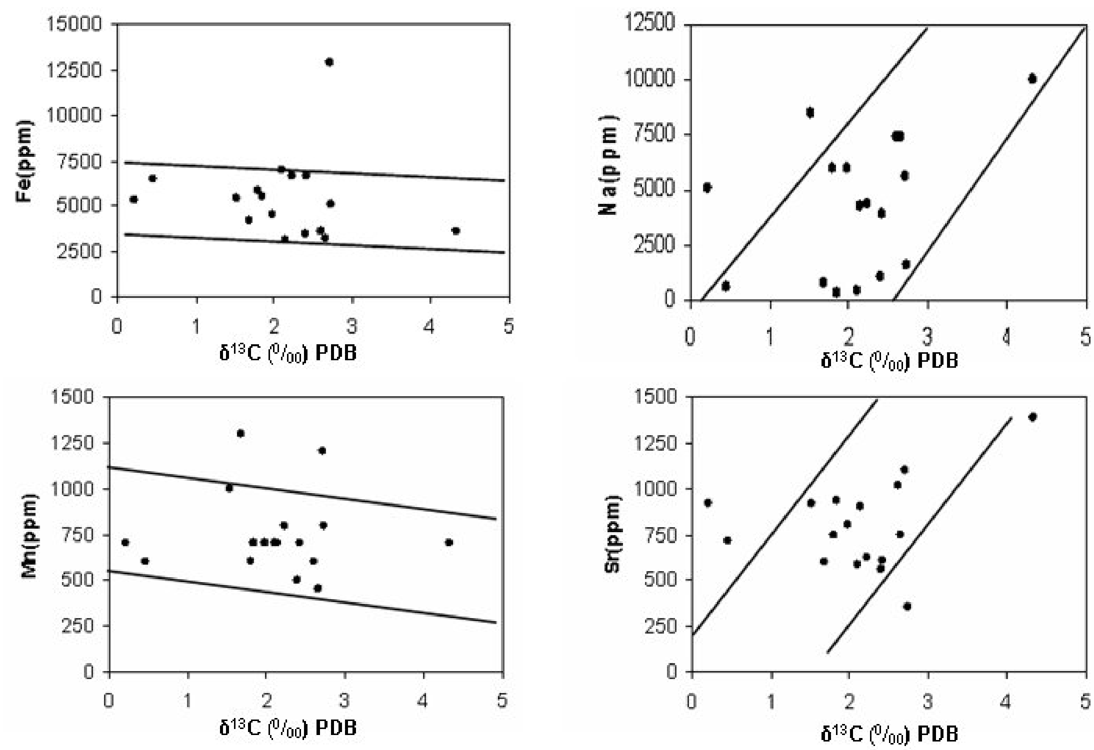

Figure 5 - Plots of δ^{13}C versus Mn, Na, Fe and Sr concentrations in the limestone of the study area. These data are similar to variation of δ^{18}O (see Fig. 5).

Stage 3. *Burial environment*

Mechanical and chemical compaction at this stage is characterized by closer packing in less cemented lithofacies, reduction of the primary intergranular porosity, resulting the sutured and concavo-convex grain contacts, broken bioclasts and deformation of ooids (spastolith - Flugel, 1982). Stylolitization and dissolution in some mud-supported lithofacies also formed at this stage. Silicification of some skeletal grains can also be observed. Although precipitation of cement is insignificant volumetrically at this stage. However, these cements are present in some packed grainstone lithofacies with similar fabric of meteoric cement, burial evidence and bright luminescence. It is noted that high overburden pressure at this stage created microfractures that filled by high Mn/Fe ratio calcite cement and bright luminescence (see Figs. 2E and 3E).

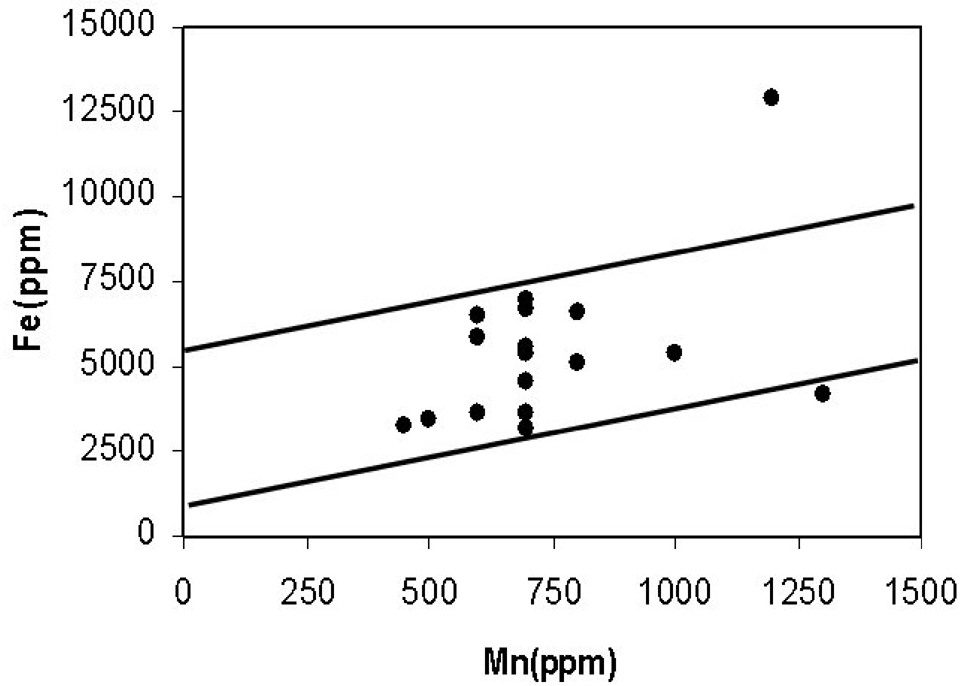

Figure 6 - Positive correlation of Fe versus Mn, showing the influence of meteoric fluids.

Stage 4. *Uplift*

The Lower Cretaceous limestones in the study area were folded and faulted during the Alpine Orogeny (Alavi, 1992 and 1996) and a lot of fractures were formed. At this stage, dissolution by meteoric waters occurred along these fractures.

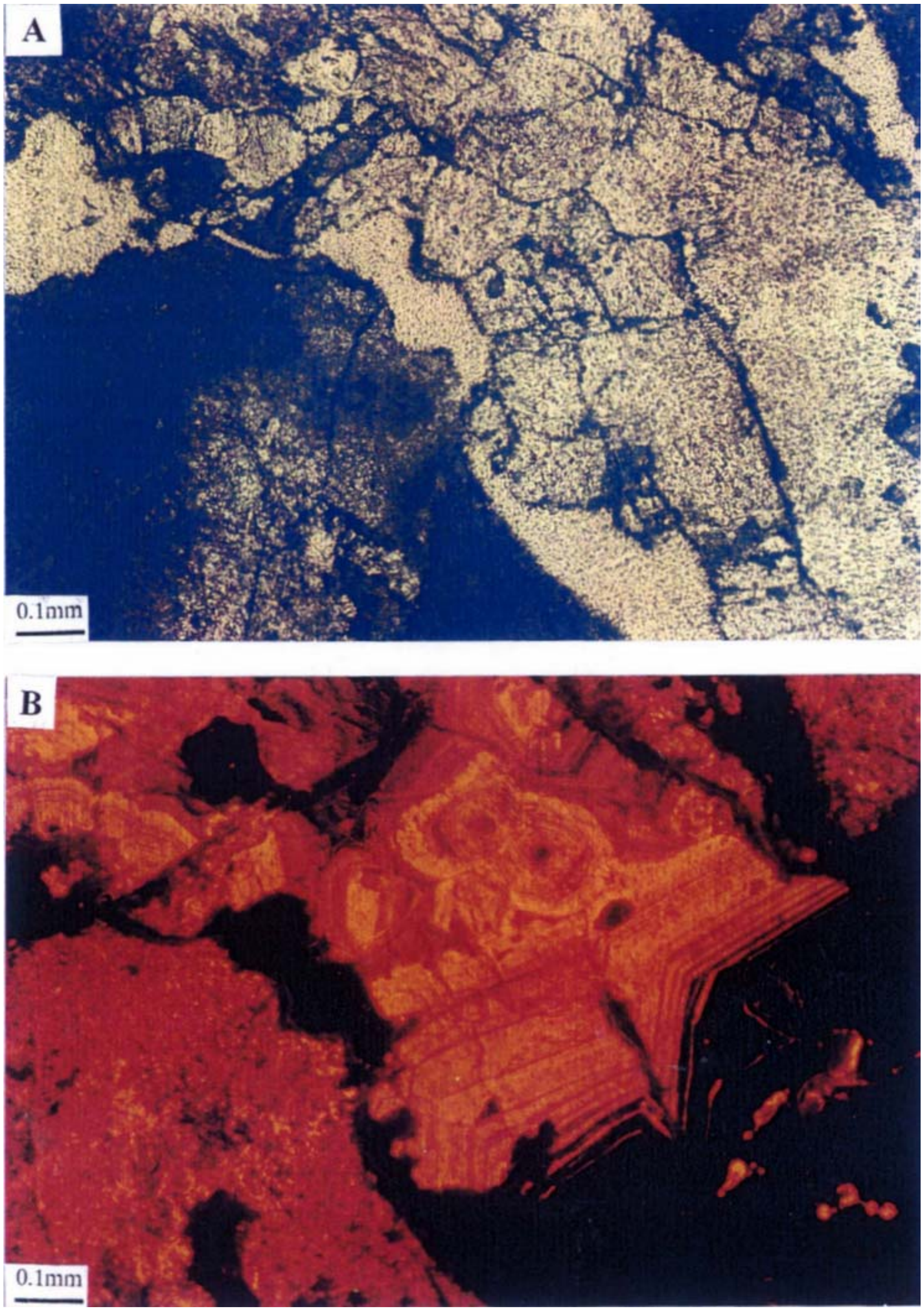

Figure 7. A) Meteoric cement in grainstone lithofacies (PPL). B) The same view as A under CL. It shows zonation that formed under variety of oxidation (dark) and reduction (bright) conditions.

Time / Diagenetic Processes	Early (Marine Phreatic)	Middle (Meteoric Phreatic)	Late	
			Burial	Uplift
Micritizitation	——			
Isopachous Fibrous Cement	——			
Blocky Cement		——		
Syntaxial Overgrowth Rim Cement		——		
Equant Cement		——		
Aggrading Neomorphism		——		
Physical Compaction	- - - -	- - - -	——	
Stylolitization			——	
Silicification			——	
Fracturing				——

Figure 8 - Paragenetic sequence for the Lower Cretaceous limestones in the Binalood region.

Conclusion

The Lower Cretaceous limestones exposed in the Binalood Mountain Range south of Mashhad in NE Iran were deposited in open-marine, sandy-shoal, lagoon and tidal flat facies belts on a platform of ramp type. Compaction, cementation, micritization, silicification, neomorphism and fracturing are the most important diagenetic processes that have affected these rocks in different diagenetic environments. Depletion of ^{18}O (ranges from -2.1 to -11.0‰ PDB) and decreasing amounts of Na and Sr as well as increasing of Fe and Mn are attributed to diagenetic alteration in the meteoric diagenetic environment. Interpretation of paragenetic sequence revealed that diagenetic processes have operated in four different stages including marine, meteoric, burial and uplift. The calculated temperature of the ambient water in which marine calcite was deposited is about 26°C.

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