

Diagenetic History of the Chuping Limestone at Bukit Tungku Lembu, Perlis, Malaysia

(Sejarah Diagenesis Batu Kapur Chuping di Bukit Tungku Lembu, Perlis, Malaysia)

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ABSTRACT

The Early Permian to Late Triassic limestone of Chuping Formation in Bukit Tungku Lembu consists of six microfacies and exhibits various types of diagenetic products, reflecting its complicated diagenetic history. Out of 89 samples collected for petrographic studies, six major microfacies have been identified, namely brecciated limestone, dolomites, mudstones, wackestones, packstones and grainstones. Four major diagenetic processes comprising micritization, cementation, compaction and dolomitization have also been identified. Micritization occurs as the first phase diagenesis after the sediments were deposited on the sea floor, followed by cementation in the marine environment and also in meteoric environments when the shallow marine carbonate at some points in time was subjected to subaerial exposure. Early burial were indicated by fractures and deformation as well as destruction of grains whole structure. Increasing pressure during deeper burial has produced dissolution seams and stylolites. Dolomitization also occurred in the deeper burial environments.

Keywords: Chuping limestone; diagenesis; geochemistry; microfacies

ABSTRAK

Batu Kapur Chuping berusia Perm Awal hingga Trias Akhir di Bukit Tungku Lembu terdiri daripada enam mikrofasis dan memperlihatkan berbagai jenis hasil diagenesis, menggambarkan keragaman sejarahnya. Daripada 89 sampel yang dikutip untuk kajian petrografi, telah dikenal pasti enam mikrofasis iaitu batu kapur terbreksi, dolomit, batu lumpur, batu wak, batu padat dan batu butir. Empat proses diagenesis utama juga telah dikenal pasti seperti pemikritan, penyimenan, pepadatan dan pendolomitan. Pemikritan berlaku pada tahap awal diagenesis selepas sedimen dimendapkan di dasar laut, diikuti oleh penyimenan di sekitaran marin dan juga meteorik apabila sedimen pada suatu ketika terdedah kepada atmosfera. Penimbunan awal ditandai dengan peretakan dan canggaan serta pemusnahan keseluruhan butiran. Peningkatan tekanan semasa penimbunan menghasilkan pelarutan tekanan dan stilolit. Pendolomitan juga berlaku dalam sekitaran penimbunan dalam.

Kata kunci: Batu kapur Chuping; diagenesis; geokimia; mikrofasis

INTRODUCTION

Diagenesis refers to the sum of physical and chemical processes that transforms soft sediment into hard solid rock. The processes normally take place after sediment deposition to bind sediments together and become more stable (Wayne 2008). The process includes stages involving stabilization of carbonate mineral through dissolution of skeletal or any carbonate material to cementation and neomorphism, recrystallization and dolomitization (Flügel 2004) which could take place in any of the major diagenetic environments (Moore 1989).

Diagenesis is a process that takes place out of sight and out of mind since it occurs mostly when sediments are buried deeper in the subsurface. Even arguments and discussions are still being made on how carbonate sediments become hard rock when there is no evidence of cements or cement crystals are found in the rocks. It is even more intriguing when considering how the porosity

of deposited carbonate sediments had been reduced from normally 40-70% to less than 5% in carbonate rocks (Bathurst 1971) without any sign and evidence of compaction. Many researchers have suggested that it was due to cementation. However, the vast amount of cements needed to binds the sediments together need to be answered and most geologists have agreed that dissolution and re-precipitation of the carbonate materials could be the most plausible answer.

Cements are produced by re-precipitation from dissolution of carbonate skeletal materials or from replacement of metastable minerals such as aragonite and Mg-high calcite by more stable calcite cement (Bathurst 1971). Castanier et al. (1999) suggested that precipitation may also occur through carbonatogenous bacterial mediation. Precipitation of cements depends on factors such as chemistry of pore water, mineralogy and crystal chemistry, dissolution and precipitation rates, grain size,

fluid movement and reactions with organic matters (Flügel 2004). Hence, different cement types will be produced in different diagenetic environments from which they are distinguished by their characteristic crystal habits and geochemical components (Tucker & Wright 1990).

This paper is to document the different diagenetic and products that were produced by different diagenetic processes in different diagenetic environments experienced by the Chuping Limestone in Bukit Tungku Lembu area. Their characteristic and diagenetic environments will be discussed and finally its paradiagenetic sequence will be established.

GEOLOGICAL SETTING

This limestone in the study area was first described by Jones (1962) and named after Bukit Chuping in Beseri. The formation is well exposed in both Perlis and Langkawi (Gobbett 1973). In Langkawi, the limestone was found overlying the Devonian-Carboniferous clastic rocks of Singa Formation in Pulau Singa Kecil, Pulau Singa Besar, Pulau Jong and Pulau Jerkom (Jones 1981). However, in Perlis it forms a series of karst hills aligned in two parallel belts in North-South direction stretching from Southern Thailand where the limestone is known as Ratburi Limestone (Fontaine et al. 1994) until Beseri area (Azimah 1998). The limestone is generally massive and unfossiliferous except at the basal part where it consists of well-bedded dark limestone with chert nodules and abundant fossils (Gobbett 1973). The age of the limestone is estimated as Early Permian to Late Triassic by Kobayashi and Tamura (1968) as shown by the presence of fossils includes *Marginifera* sp. and *Hamletella* sp., *Bellerophon* sp., *Euomphalus* sp., *Composita* sp. (Gobbett 1973) and *Sinopora dendroides*. Fontaine et al. (1994) recorded the presence of *Hemigordiopsids* sp., *Sphairionia* sp. and *Lophophyllidium* sp. at Bukit Wang Pisang and *Tubiphytes* sp., *Globivalvulin* sp., *Pachyphloia* sp., *Langella* sp., *Lasiodiscus* sp., Nodosariids, *Pseudovermiporella nipponica*, rare bryozoans, *Sinopora asiatica* (Fontaine et al. 1988) at Bukit Merah. These data suggest that the Chuping Limestone was deposited on a sheltered shelf away from shore line meanwhile isotopic data obtained by Rao (1988) suggested a cool temperate shallow marine setting.

STUDY AREA AND METHODOLOGY

A detailed field study was carried out at a section in Bukit Tungku Lembu (Figure 1) where 89 samples consist of pale to dark grey, pure calcitic laminated limestones were collected at 1 m interval. These samples were made into thin sections and studied under the microscope for petrographic study. The thin sections were stained using a combination of Alizarin Red S and Potassium ferricyanide, dissolved in weak hydrochloric acid as suggested by Friedman and Johnson (1992). The purpose of this staining technique is to differentiate between calcite and dolomite as well as to distinguish between ferroan and non ferroan calcites.

GENERAL MICROFACIES SEQUENCE

A detailed petrographic study based on the carbonate rock classification scheme by proposed by Dunham (1962) as well as Leighton and Pendexter (1962) has distinguished carbonate microfacies from this section including breccia (intraclastic limestone), dolomites, mudstones, wackestones, packstones, grainstones and oolitic limestone as shown in Figure 2. The overall sequence is made up of several facies cycles with each cycle shows a coarsening and shallowing upward trend starting with mudstone at the bottom and end up with brecciated limestone or more precisely can be referred to as intraclastic limestone. Each cycle however, is intercalated with zones of dolomite or dolomitic limestones especially in a zone just below the intraclastic limestone. More prominent zones of dolomite can be seen in the higher part of the section. The brecciated limestone is characterized by the presence of intraclasts cemented by calcite spar which could represent collapse breccia.

DIAGENESIS

Detailed petrographic investigations show that the limestone had experienced various diagenetic processes that took place soon after the sediments were deposited on the seafloor until deep burial. Major diagenetic processes such as micritization, dissolution, cementation, compaction and dolomitization are ubiquitous in this limestone.

MICRITIZATION

Micritization is the first diagenetic phase in formation of carbonate rocks (Adabi 2009). It takes place in the marine environment where the sediments were first deposited. In order for micritisation to happen, the sediments have to remain on the ocean floor for a period of time instead of being quickly buried. Micritization process is evident in this limestone as shown in Figure 3(e). Micrites are formed by microbes which bore small holes along the boundaries of the sediments, giving spaces for micrites to move in (Bathurst 1980). Micritization may take place continuously throughout the rock history, which will eventually destroy the original grain structure (Hashemi Azizi et al. 2013).

CEMENTATION

Cementation processes occurred almost everywhere in the coarse gained facies where four types of cements namely fibrous calcite cement, fine equant calcite cement, drusy mosaic calcite cement and medium-coarse blocky calcite cement have been identified.

FIBROUS CALCITE CEMENT

Fibrous calcite cement which appears as a rim-like fabric around the surface of the particle occurs quite frequently in this limestone. It consists of radial, needle-like or columnar, fine to medium sized calcite crystals growing outwards

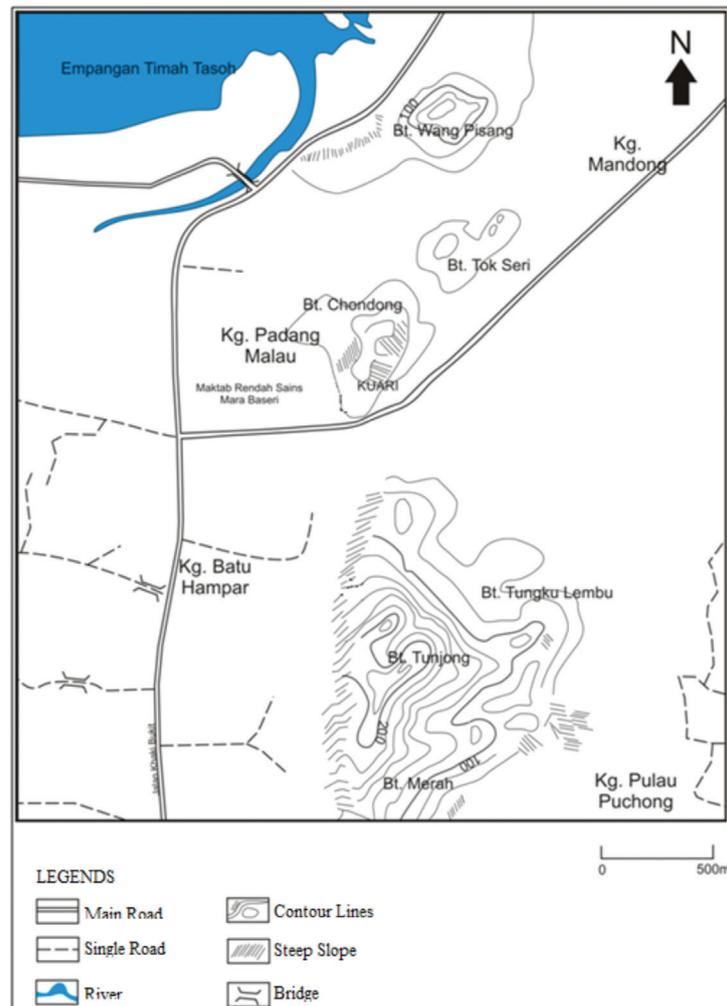


FIGURE 1. Location of Bukit Tungku Lembu in Beseri, Perlis

perpendicular to the particle's surface forming an isopachus or sub-isopachus crusts of about 20-50 μm thick (Figure 3(a)). In some cases, the cement is only concentrated at one side of the particle and in some it appears to be undulating. Isopachus fibrous cement (Figure 3(b)) usually indicates the first generation cement formed on walls of particles as a result of early marine diagenesis precipitation (Flugel 2004; Kendall 1985). It was formed as an early stage of cementing material that turns sediments into hard rocks. The growth of fibrous rim cement formation may also be superseded by growth of other organisms such as algae during deposition or by late diagenetic cements as shown in Figure 3(c).

FINE EQUANT CALCITE CEMENT

Fine equant calcite cements are present mainly within inter-particle pore spaces in the grainstones. The cement crystals are anhedral to subhedral of different sizes range from 5 to 50 μm (Figure 3(d)). This pore-filling cement may be a result of recrystallization of pre-existing cements or it could have represented granular mosaic calcite cements precipitated in a deep burial setting (Flugel 2004).

DRUSY MOSAIC CALCITE CEMENT

This type of cement shows an increasing size towards the centre of inter-particle pores. It is found precipitated inside moulds, intergranular pores and moulds of pre-existing skeletal grains (Figure 3(e)). The calcite crystals are subhedral to anhedral with size ranging from 5 to 50 μm . Drusy mosaic cements usually indicate a near-surface meteoric diagenesis of a later cementation stage during burial (Flugel 2004). In cases in which it fills moulds of skeletal grains, Scholle (2003) interpreted that the grains were completely dissolved, leaving voids and later were filled with sparry calcite cement. The boundaries between the grains and the matrix were distinguished by the micritic pockets preserved from the original sediment. It suggested that the sediments experienced meteoric dissolution and cementation prior to burial diagenesis. Wilkinson et al. (1985) also discovered that the increasing nature of the crystal size towards the centre of pores suggest that instead of aragonite, the cement were precipitated as calcite which normally occur in meteoric water conditions.

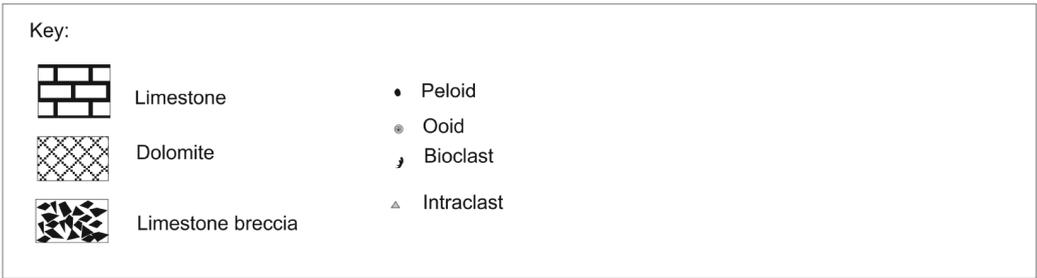
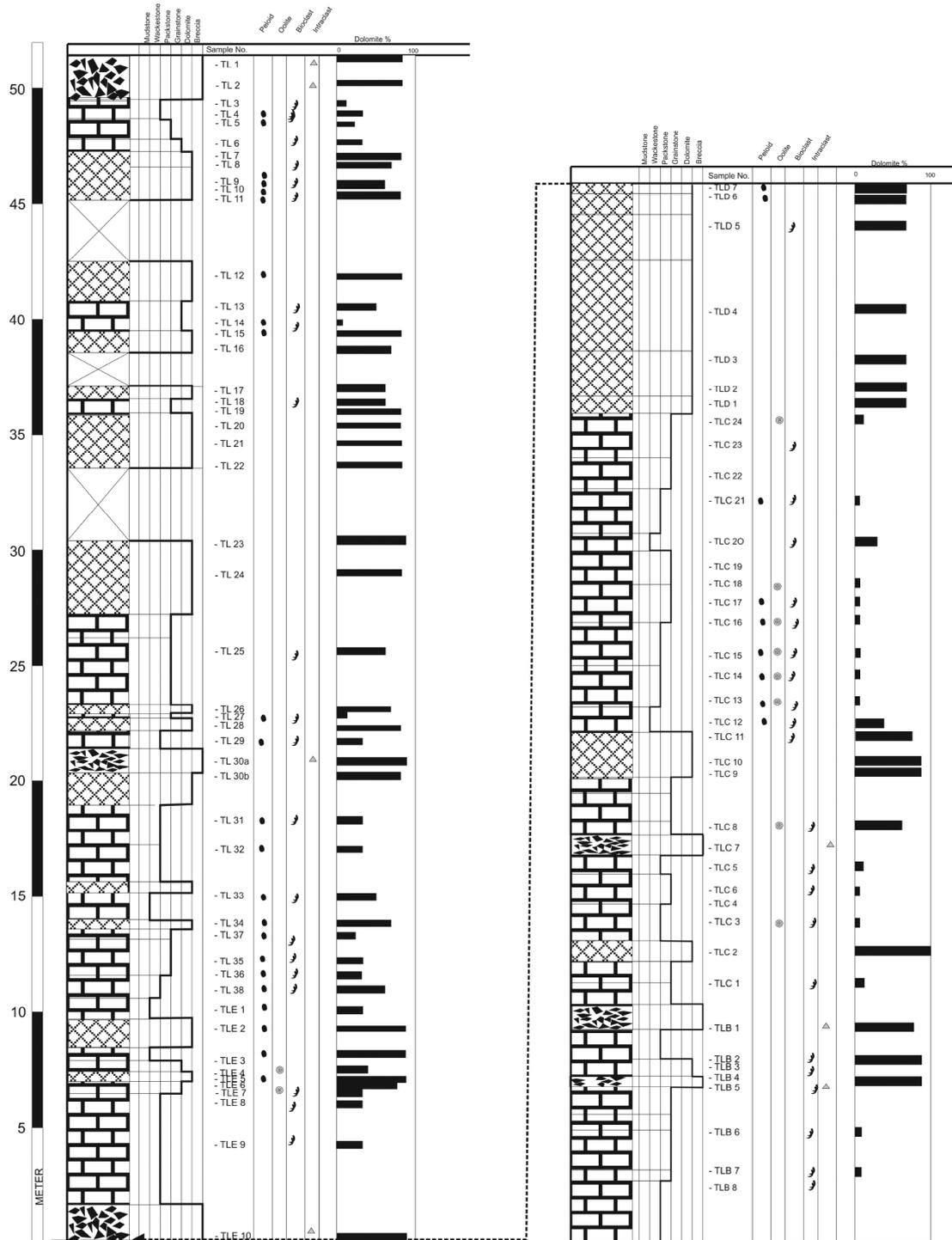


FIGURE 2. Sedimentary log showing Chuping Limestone's microfossils in Bukit Tungku Lembu

MEDIUM-COARSE GRAINED BLOCKY CALCITE CEMENT

The medium to coarse grained cement are found in sizes more than 50 μm in moulds of particles and in intergranular pores between rock fragments in breccias. It does not possess any defined shape, showing distinct boundaries between each crystal grains. They are made of non-ferroan calcite, with well-developed cleavage and extinction.

Blocky cements are typical of meteoric phreatic and burial environment as a late pore-filling stage. Medium-sized cement may also be found in mouldic pores of ooid grains. This was caused by leaching of the ooids leaving voids to be filled by calcite cements. The cements are different from intergranular cement as it is lighter in colour, granular with no distinct crystal faces and fractured. Coarser blocky calcite cements are common in brecciated facies (Figure 3(f)) which might represents collapse breccia.

COMPACTION AND PRESSURE SOLUTION

Compactions are quite dominant in Chuping Limestone as shown by fractures, broken and deformed grains, stylolites and pressure solutions. These indicate that the overburden pressure from burial had caused both mechanical and chemical compaction to occur. Mechanical compaction happened after few meters of burial (during early burial settings) causing fractures, broken and deformed grains. As the overburden pressure increase, it gives rise to closer grains packing, forming pressure solution (Halimeh et al. 2013).

DOLOMITIZATION

Dolomite occurrences are very common in Chuping Limestone of Bukit Tungku Lembu area. The dolomite occurs as individual rhomb replacing certain grains or as pervasive dolomite. The textural fabrics of dolomites in the Chuping Formation are in the forms of planar-e and planar-s. Most of them have well-cleavage, cloudy and inclusion-free. It replaces certain grains and in other cases it replaced the whole fabrics completely.

Grain-replacing dolomites made about 1 to 30% of the total rock volume, selectively replacing grains especially in ooids, pellets and skeletal allochems (Figure 4(b)). Most of them are cloudy in nature, some also possessed a clear rim in a perfect euhedral rhomb shape. Meanwhile the pervasive dolomite as shown in Figure 4(c) usually has a subhedral to anhedral texture with curvy to straight boundaries (Figure 4(d)) with well-developed cleavages.

GEOCHEMISTRY

The results of geochemical analysis of this sedimentary rock (Table 1) show big fluctuations of the elements values that may reflect the variation of subsurface water in which dolomitization had taken place. The concentrations of trace elements other than that of Ca and Mg are normally less than 1%. The Ca/Mg ratios however, are fluctuating very significantly, ranging from 1.681 mol to as high as

106.942 mol, showing that the limestones may have been precipitated from a high-Ca fluid. This data might also suggest that there are dedolomitization taking place in some of the samples in which dolomites crystal are being replaced by calcite.

Fe and Mn concentrations are fairly low suggesting that the dolomites are not of a hydrothermal origin (Zhang et al. 2009). The Mn concentration is less than 1 ppm while Sr concentrations range from 0.008-0.031% (80-310 ppm). Meanwhile Na concentrations vary from 0.010-0.034% (100-340 ppm) showing that the dolomites were precipitated from Na-rich fluids but with relatively low Sr concentration (Veizer 1983). It suggests that it is not of a marine origin as Na concentration in marine dolomite is normally recorded as between 110 and 160 ppm.

DIAGENETIC HISTORY

Based on the detailed petrographic study, diagenetic history of Chuping Limestone at Bukit Tungku Lembu can be summarised as follows:

EARLY MARINE DIAGENESIS

An early process of diagenesis is characterised by micritization and precipitation of early fibrous circumvoid rim cements. Micritization which normally occurs in quiet environments helps to fills pore spaces in between grains aided by microbial activities (Kim & Lee 1996). Meanwhile, early precipitation of calcite cements acts as the first generation cement that binds sediments together. This process is assisted by water movement such as wave forcing, tidal pumping, thermal convection and diffusive transport (Scholle 2003). They transport and force water from the surface into the interior of the sediments and fill the pores between particles.

METEORIC ENVIRONMENT

Meteoric environment can be divided into phreatic (saturated) and vadose (unsaturated) zones (Flugel 2004; Scholle 2003). A vadose zone is located nearest to the surface of the earth, overlies the phreatic zone. Here, dissolution of carbonate materials is common and the cement fabrics usually reflect the way how water was distributed through the particles. It includes formation of pendant-like cement, meniscus cement in between grains, whisker crystals, microspar and some other features (Scholle 2003). Figure 3(c) shows pendant cements in grainstone. A phreatic zone occurs below water table where the pore spaces are completely filled with freshwater (Flugel 2004). In this zone, intrgranular, mouldic and vuggy porosities are recemented by calcite. Figure 4(a) shows formation of mouldic vugs in ooids, destroying the internal structures of ooids completely and replaced it with medium grained granular calcite cement. It is also shown by the complete precipitation of equant calcite in intergranular pore spaces (Figure 3(a)).

TABLE 1. XRF data of different elements from selected samples of Chuping limestones

Sample/ Conc.	CaO (%)	MgO (%)	Al ₂ O ₃ (%)	Fe ₂ O ₃ (%)	K ₂ O (%)	MnO (%)	Na ₂ O (%)	P ₂ O ₅ (%)	SiO ₂ (%)	SrO (%)	TiO ₂ (%)	Ca/Mg (mol)
TL4	40.263	9.635	0.266	0.051	0.014	0.005	0.021	0.004	0.354	0.010	0.009	4.179
TL6	44.292	4.321	0.161	0.029	0.012	0.006	0.014	0.005	0.200	0.009	0.008	10.250
TL7	37.587	13.197	0.147	0.092	0.014	0.005	0.027	0.003	0.302	0.010	0.012	2.848
TL12	43.870	5.117	0.341	0.065	0.018	0.007	0.016	0.005	0.414	0.010	0.011	8.573
TL15	40.207	10.265	0.066	0.039	0.012	0.005	0.022	0.005	0.112	0.010	0.008	3.917
TL20	35.445	16.552	0.079	0.037	0.010	0.004	0.029	0.003	0.094	0.011	0.009	2.141
TL22	41.898	7.805	0.082	0.047	0.014	0.005	0.018	0.004	0.126	0.010	0.008	5.368
TL23	40.940	9.011	0.092	0.037	0.013	0.004	0.020	0.006	0.204	0.010	0.008	4.543
TL26	34.121	17.724	0.089	0.049	0.012	0.008	0.033	0.004	0.147	0.012	0.009	1.925
TL28	46.800	1.449	0.083	0.028	0.014	0.005	0.011	0.004	0.134	0.010	0.008	32.298
TL29	43.785	5.610	0.062	0.040	0.012	0.005	0.015	0.005	0.111	0.008	0.008	7.805
TL33	42.771	6.903	0.095	0.026	0.010	0.004	0.018	0.005	0.115	0.008	0.008	6.196
TL37	44.433	4.441	0.046	0.030	0.011	0.004	0.015	0.010	0.073	0.009	0.007	10.005
TLB5	47.589	0.445	0.061	0.018	0.010	0.004	0.010	0.004	0.077	0.021	0.007	106.942
TLB8	47.476	0.535	0.050	0.024	0.010	0.004	0.010	0.004	0.078	0.010	0.007	88.740
TLC1	47.166	0.950	0.071	0.026	0.011	0.003	0.011	0.004	0.086	0.035	0.007	49.648
TLC2	33.980	18.382	0.044	0.030	0.010	0.005	0.034	0.005	0.052	0.011	0.008	1.849
TLC3	47.279	0.655	0.035	0.017	0.010	0.004	0.010	0.006	0.054	0.025	0.007	72.182
TLC5	47.251	1.016	0.173	0.021	0.010	0.003	0.011	0.004	0.192	0.015	0.007	46.507
TLC6	47.251	0.889	0.202	0.025	0.011	0.003	0.010	0.004	0.221	0.018	0.007	53.151
TLC9	34.318	17.107	0.144	0.053	0.013	0.006	0.030	0.004	0.148	0.011	0.010	2.006
TLC18	47.364	0.464	0.046	0.018	0.011	0.005	0.010	0.004	0.063	0.028	0.007	102.078
TLC21	47.279	0.546	0.248	0.021	0.012	0.004	0.011	0.004	0.260	0.031	0.008	86.592
TLC24	46.406	1.911	0.059	0.033	0.013	0.004	0.011	0.004	0.084	0.012	0.007	24.284
TLD1	33.642	18.181	0.153	0.063	0.011	0.006	0.033	0.007	0.217	0.011	0.010	1.850
TLD4	33.501	18.431	0.088	0.065	0.011	0.005	0.032	0.004	0.105	0.011	0.011	1.818
TLD5	32.853	19.547	0.038	0.054	0.010	0.006	0.033	0.004	0.045	0.011	0.009	1.681
TLE1	43.842	5.250	0.068	0.034	0.014	0.004	0.016	0.007	0.136	0.010	0.009	8.351
TLE4	44.631	4.303	0.047	0.030	0.012	0.004	0.014	0.007	0.083	0.010	0.008	10.372
TLE6	42.010	7.590	0.047	0.031	0.011	0.005	0.020	0.006	0.109	0.011	0.008	5355.

EARLY TO LATE BURIAL DIAGENESIS

Burial diagenesis took place in the subsurface, shown by evidence of physical alteration such as compaction, pressure solution (existence of stylolites) and late stage cementation (Flügel 2004). Evidents of burial diagenesis are shown by the presence of medium to coarse, blocky, sparry calcite cements as well as broken fragments of allochems. Late stage dolomitization is another indication of burial diagenesis. Petrographic studies of this limestone show that most of the dolomite crystals are of medium to coarse crystal size, light to dark brown in colour. Individual rhombus dolomite crystals that are not affected by any compaction or pressure indicate a replacement by dolomite crystal growth in a shallow burial environment. However, dolomites that

are tightly packed showing xenotopic texture suggested a late stage burial environment in which the rock experienced compaction. This is also supported by the presence of saddle dolomite. There is also an indication of a possible dedolomitization process occurred in the Chuping Limestone. Referring to the petrophysical and geochemical data of TL12, the high Ca/Mg ratio does not reflect the 80% dolomite crystal observed in the thin section. It is believed that the dolomite crystals have been replaced by calcite at a very late stage.

PARADIAGENETIC SEQUENCE

Petrographic evidences show that the first generation cement appeared in the form of fibrous rimmed cement

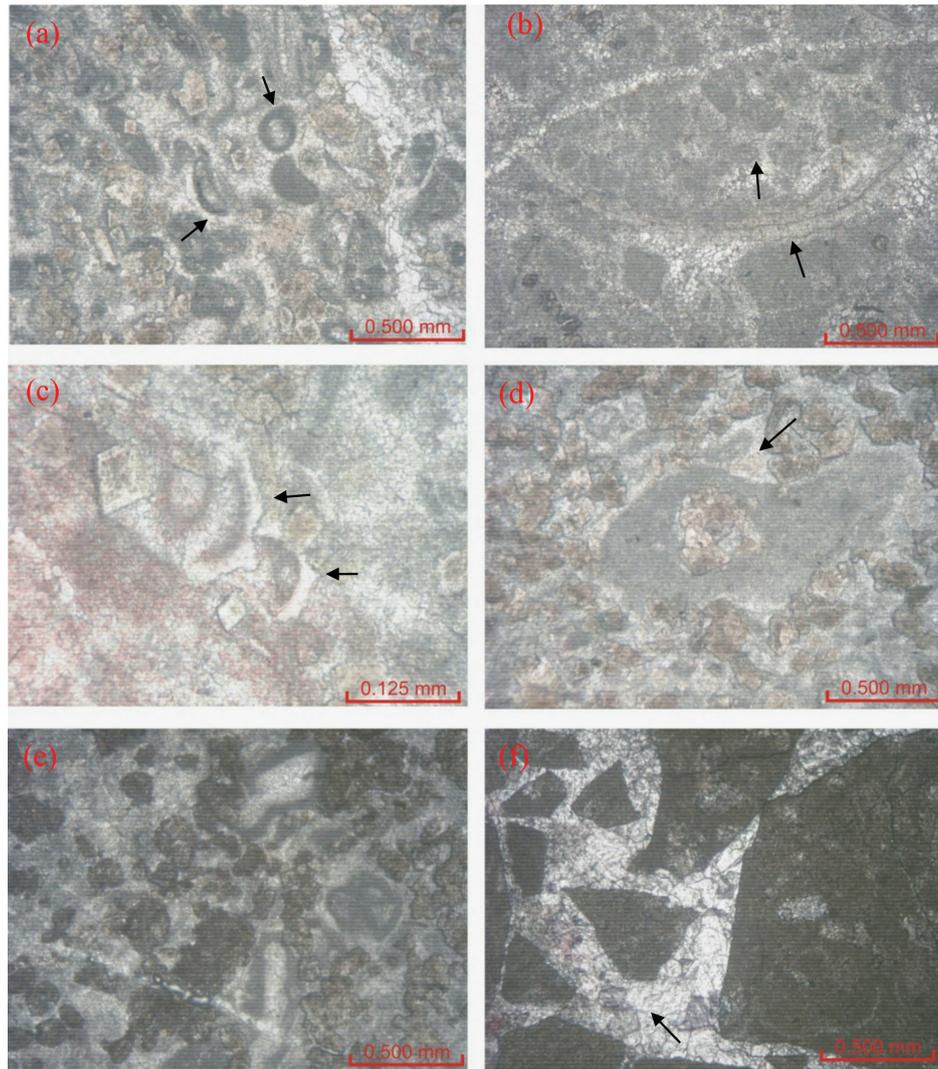


FIGURE 3. (a) Showing fibrous rim cements, (b) Fibrous rim cement on the surface of a brachiopod clast, (c) Growth of pendant cements indicating a vadose meteoric environment, (d) inter-granular fine equant calcite cement (e) Drusy mosaic calcite cement and (f) Medium to coarse grained blocky calcite cement with micritic pockets

on the surface of allochems. Its formation is assisted by water movement and also from secretions of bacteria and other multicellular organism in the earlier marine diagenesis. This binds the sediments together and formed a hard rock. This is followed by dissolution of allochems and recrystallisation forming secondary calcite spar cements that fills the pores in between the sediments. This happened in the meteoric environment in which dissolution is most possible. Then a series of physical and chemical compaction takes place as the sediments entered burial diagenetic phase. Stylolites, pressure solution and deformation of clasts occur due to the pressure. Dolomitisation serves as the final diagenetic process as it replaces matrix and molds of skeletal clasts with euhedral to subhedral dolomites crystals. Table 2 shows a paradiagenetic sequence which consisted of the various diagenetic processes experienced by the Chuping Limestone in their approximate order of occurrence.

CONCLUSION

These diagenetic processes play a really important role in the history of the Chuping Limestones, Overall the Chuping Limestone had undergone 3 diagenetic processes. It started with early marine diagenesis which is indicated by the process of micritization and formation of fibrous rim calcite cements. Marine phreatic environment then took place in shallow marine water involving the stabilizing of metastable minerals into stable calcite cements. Then the sediment came into a meteoric phreatic environment in which voids and pore spaces created by dissolution were recemented. This is followed by diagenesis in the subsurface environments where mechanical compaction took place after few meters of burial forming fractures, deformation and destruction of grain structures. As the overburden pressure increases, dissolution occurs forming pressure solutions, stylolites as well as new cement generations. Replacement by

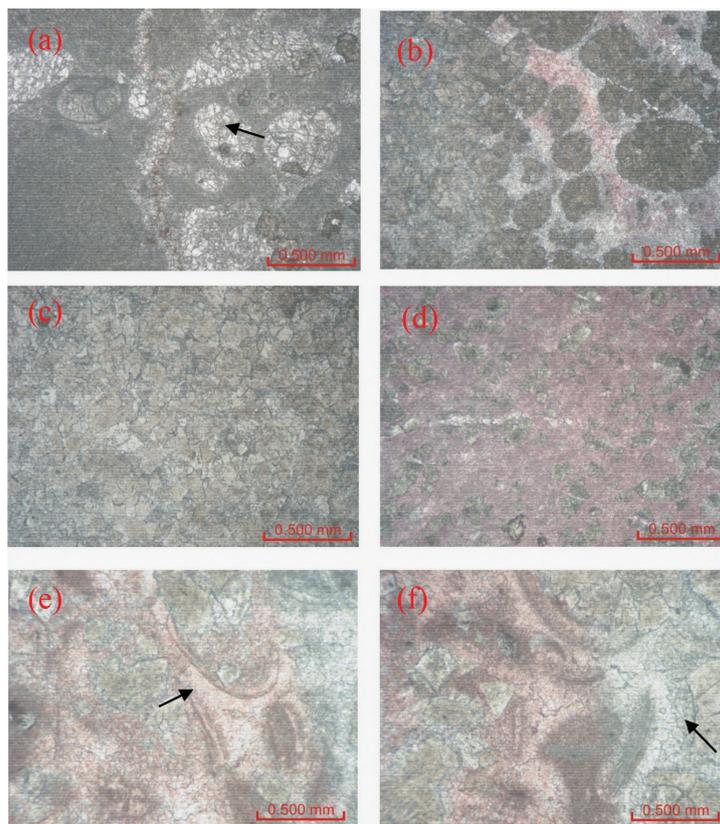


FIGURE 4. (a) Mouldic voids formed through leaching and dissolution in a meteoric phreatic environment, (b) Dolomitization happened as replacement in ooids grains, (c) Complete dolomitization, (d) Partially dolomitized, the stained in red are calcite while the unstained minerals are dolomites, (e) and (f) showing the occurrence of first (indicated by the arrow) and second generation cement (in the box)

TABLE 2. Paradiagenetic sequence of Chuping Formation

	Early marine diagenesis	Meteoric diagenesis	Shallow burial	Late burial diagenesis
Marine cementation	██████████			
Micritization	██████████			
Dissolution		██████████		
Meteoric cementation			██████████	
Compaction			██████████	██████████
Late stage cementation				██████████
Stylolitization				██████████
Dolomitization				██████████

dolomites follows and re-changed the whole limestone fabrics and structures.

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