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Review Article

New Advances and Existed Problems for the Forming Mechanism of the Microbial Dolomite

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Abstract: In the recent years, the study on the formation mechanism of microbial dolomite broadens the concept of "dolomite problem" perspective in sedimentology. The microbial dolomite model discusses sulfate reduction reaction, methane production, organic molecular hydrolysis and many other related topics under the same heading, which help in explaining the microbial metabolic mechanism of precipitated dolomite. Recent research on the dissolved sulfide precipitation of dolomite combined with the reduction reaction mechanism of sulfate provides a new understanding in promoting the mechanism of dolomite precipitation reduction reaction. Studies of the modern sedimentary environments highlighting the precipitation of primary dolomite induced by microbial accretion also represent the progress towards defining microbial dolomite. The striking results achieved in fixing the “dolomite problem” pointed out that the study of microbial processes contributing towards environment of primary dolomite precipitation and its mechanism may provide more thoughtful explanations for the microbial dolomite in the stratigraphic record. This study highlights that despite of the advancement in dolomite studies, adoption of microbial dolomite model only to explain the complex phenomenon of dolomite in geological record is limited and still need further research.

Keywords: Dolomite Problem, Microbial Dolomite, Sulfate Reduction, Microbial Mat

1. Introduction

Dolomite is a common carbonate mineral throughout the geological history. In contrast to its mass presence in the ancient strata, dolomite is rarely seen in modern strata in a series of studies represented by the famous experiments of Land. It is demonstrated that the precipitation of ordered dolomite cannot be carried out by non-biological means underground [1]. It is further explained that there is a chemical dynamic obstacle in the precipitation of dolomite under the surface condition, which is the famous dolomite problem in sedimentology [2-5].

In Vasconcelos' experiment in 1990s, the microbiotic process was introduced into the study of dolomite precipitation in the surface environment, so that the metabolism of microbiophytes was involved in the redox reaction. The phenomenon that dolomite can be precipitated under the condition of surface temperature and pressure has been revealed [6, 7]. Thus, the concept of "microbial dolomite" is derived, which represents a conceptual progress in the study of dolomite problem in recent years [8]. A series of microbial metabolic processes such as sulfate reduction, methanogenesis, organic molecular hydrolysis etc, which can promote dolomite precipitation, have produced a "microbial dolomite (rock)" model. At the same time, it also provides a new research idea for further exploring the mechanism of micro-organism action to promote dolomite precipitation. Furthermore, the experimental study on the catalytic precipitation of dolomite by soluble sulfides deepens the understanding of the chemical kinetic barrier to the reaction of dolomite, and complements the mechanism of influence on the precipitation of dolomite by the reversion reaction of sulfate. The discovery of primary dolomite deposits within microbial mats in the modern environment can provide a new approach for understanding whether there are primary dolomite records in geological records. In pursuit of surface environment, the progress of microbes involved in dolomite precipitation is of great significance to further understand the dolomite problem in sedimentology.

2. The Mechanism of Microorganism Metabolism in Dolomite Formation

2.1. Sulfate Reduction

Sulfate-reducing bacteria were first found to be involved in the precipitation of dolomite, which can overcome the obstacles in chemical kinetic reactions and promote the microbial metabolism of dolomite precipitation. As early as 1903, Russian biologist Nadson observed that a small amount of fine dolomite could be precipitated by sulfate reducing bacteria isolated from salt lakes under anaerobic conditions, which did not attract much attention at that time. Vasconselos has discovered through experiments that dolomite minerals precipitate where sulfide reducing bacteria accumulate, however precipitation could not happen without strains or with dead strains [6, 7]. The simulated experimental results are very similar to those of dolomite crystal precipitation in sediments in modern high-salinity environments [9-12]. Similar reactions exist in modern deep-sea oxygen-rich organic-rich environments: SRB metabolism converts organic matter into raw materials for sulfate reduction by sulfate reducing bacteria. And release the Mg^{2+} in the strong ion $MgSO_4$ pair through reduction reaction, thus satisfying the preparation for dolomite reaction in the ion environment around the SRB fine cell [13]. These phenomena related to the precipitation of dolomite mean that, sulfate reduction is a special microbial metabolism related to dolomite precipitation mechanism. In view of the abundant research achievements of the predecessors, the general assumption of the principle of sulfate reduction is summarized below [3]:

$$Ca^{2+} + Mg^{2+} + 2SO_4^{2-} + 2CH_3COOH \rightarrow CaMg(CO_3)_2 + 2H_2S + 2H_2O + 2CO_2 \quad (1)$$

2.2. Methanogenesis

Methanogenic archaea is a kind of anaerobic bacteria with special cell composition and methane-producing function. In fresh water, methanogenic bacteria improve the alkalinity of environment by separating organic components to promote dolomite precipitation. In seawater, methanogenic bacteria cooperate with SRB and undergo the process of organic mineralization. Oxidation of methane to promote dolomite deposition [14].

Based on the geochemical observation of the methane formation process affecting carbonate rock production in the Nyegga Pockmark area of the Norwegian Sea, the results are as follows: Methanogenic bacteria use low molecular organic acid to form some authigenic carbonates in areas where microbiological reactions are active [15]. The study of the process of methane leakage under the sea has obtained the recognition that the methane-rich fluid transported to the sea floor and SO_4^{2-} brought by the upper sea water diffusion into the sediments would cause anaerobic oxidation methane through methanogenic bacteria (AOM: Anaerobic Oxidation of Methane). In this process, the synergistic metabolism of sulfate reducing bacteria and methanogenic bacteria (AOM) increases the pH and alkalinity of water. HCO_3^- formed by ionization binds to Mg^{2+} Ca^{2+} and cause the precipitation of dolomite in the sulfate-methane transiton zone (SMTZ: Sulfate-Methane Transiton Zone) [16, 17].

2.3. Hydrolysis of Organic Molecules

Previous studies have found that either the sulfate reduction of SRB or the AOM with methane producing bacteria is the result of the microbiological cultivation experiments under anaerobic conditions, while the discovery of moderate halophilic aerobic bacteria further improves the involvement of microbes in the Dolomite Precipitation in the "microbial dolomite" system [14]. Snchez-Romn et al showed that heterotrophic moderately halophilic aerobic bacteria hydrolyze organic molecules to produce CO_2 and NH_4^+ through aerobic respiration metabolism. To increase ph value of culture medium and increase alkalinity in water environment to promote dolomite precipitation [9, 18, 19].

Extracellular polysaccharides and other organic molecules may promote the formation of disordered dolomite crystals by weakening the complexation between Mg^{2+} and water molecules. This conclusion introduces organic molecules into the theoretical model of microbial metabolism to promote dolomite precipitation [20]. The discovery of extracellular polysaccharides to promote dolomite precipitation has a new understanding and puts forward the idea that microbial extracellular polysaccharides participate in dolomite precipitation by polymerizing form [21]. *Lysinibacillus Sphaericus* and *Sporosarcina Psychrophila*, two microbes with urea hydrolysis activity can promote the discovery and study of dolomite precipitation in the experimental simulation environment [22] which broadens the field of study on dolomite primary precipitation. In particular, the hydrolysis of urea promoted the primary precipitation of dolomite. It was observed that Bacillus Sphaeraceus decomposed urea into CO_2 and NH_4^+, which increased the basicity and ph value of surrounding environment and reduced the activation energy of magnesium complexation, promoting Ca^{2+} and Mg^{2+} to accumulate near microbial cells, and thus promoting the crystallization of dolomite. The reaction equation of urea hydrolysis is:

$$CO(NH_2)_2 + 2H_2O \rightarrow 2NH_4^+ + CO_3^2 \quad (2)$$

Therefore, the results of this experiment confirm the kenward's idea in 2013 [21] that organic molecular hydrolysis is mainly located in cell wall (Figure 1), and obtained rhombohedral dolomite crystals, adding another way for microbes to promote the precipitation of primary dolomite. It also represents an important development in the exploration of microbial dolomite precipitation by Chinese scientists.

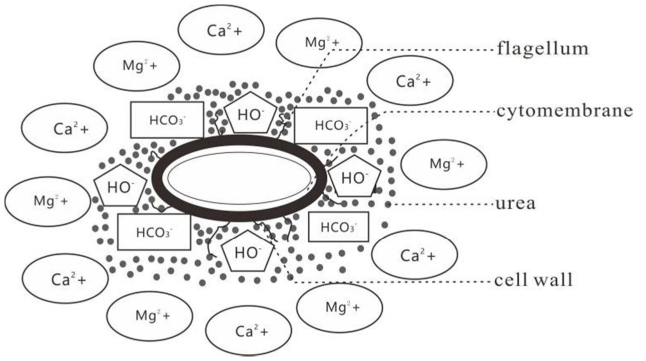

Figure 1. *Concept of promoting dolomite precipitation by urea hydrolyzed active bacteria.*

3. Dolomite Precipitation Catalysis of Dissolved Sulphide

Although the mechanism of SRB involved in dolomite precipitation-sulfate reduction has been widely concerned and recognized. However, there are still some difficulties in explaining the reaction principle of dolomite precipitation. Firstly, the presence of SO_4^{2-} may inhibit the dolomite reaction, and Mg^{2+} in the sedimentary environment will be preferentially captured by SO_4^{2-} to form $MgSO_4$. The formation of dolomite crystals or the progress of dolomitization are hindered by the adhesion to the deposition surface during the post-deposition process [23]. Secondly, the results of the study on the effect of Mg^{2+} on the crystallization of calcium magnesium carbonate in aqueous depositional environments are discussed. It is considered that the contact mode of Mg^{2+} on the surface of carbonate crystal and the shadow response of crystal axis growth remain in doubt [24, 25]. The adhesion of Mg^{2+} to the formation of dolomite crystals are mainly in two forms: hydrated ions bound with water molecules are one of the most stable binding ionic bonds. The amount of Mg^{2+} involved in the formation of calcium magnesium carbonate is limited [26, 27, 28], and the growth of dolomite crystal is inhibited by the adsorption of hydrated Mg^{2+} on the surface of the original crystal [29, 30, 31].

According to Astilleros' observation in 2010 that in the process of biogenic high magnesium calcite production, polysaccharide molecules are produced by microbial metabolism on the crystal surface and dissolved sulfides, dewatering the magnesium ions attached to the crystal production site. Although no evidence has been observed in previous studies to explain how dissolved sulphide exerts its catalytic effect, the adsorption of dissolved sulfides on the surface of calcite crystals [25-30] can be observed obviously in the precipitation of high-magnesium calcite. In its supersaturated aqueous solution, the participation rate of magnesium ions in the growth of calcium magnesium carbonate crystals corresponds to the mere presence of only millimeters of magnesium ions which can also affect the participation of magnesium ions in the growth of calcium magnesium carbonate [32]. This means that the dissolved sulfide adsorbed on the surface of the disordered dolomite crystal reduces the dissociation energy to stabilize the formation of the crystal nucleus, in other words, the existence of dissolved sulphide, the time of maintaining stable state of carbonate ions on crystal surface is prolonged. At the same time, due to the existence of dissolved sulfides, the activation entropy of the reaction ring increases, which changes the electrochemical properties of the aqueous solution on the crystal surface and weakens the adhesion of the magnesium ion hydration group. The dielectric constant of the group precipitated from the calcium magnesium carbonate crystal on the disordered dolomite crystal nucleus formed by the first step is changed. It is proved that the sulfide dissolved in the sedimentary water environment weakens the adhesion of magnesium hydrate with octahedral groups on the crystal and weakens the blocking effect of water and magnesium ions on the formation of dolomite crystals.

Based on these two experimental results, it can be concluded that the overall effect of precipitating the precipitated dolomite precipitated by dissolved sulphide is twofold: to promote the dehydration of magnesium ions on the long surface of the crystal nucleus, by changing the electrochemical properties of the crystal surface, the adsorption of magnesium ions on the crystal growth was removed.

4. Primary Dolomite Precipitation in Microbial Mats

Microbial mats represent one of the most diverse ecosystems in the modern earth's surface environment. The special microbial communities themselves represent the earliest and longest lasting ecosystems on earth. In answer to the question of "what is the microbial mat", the answers presented are diverse, but this is the source of diversity discussion in different positions, which provides a theoretical basis for a young sedimentology branch of the earth's Biological Science - "microbial mat sedimentology". However, through diagenetic "filters", analogies are made through the study of modern microbes, the understanding of the growth mechanism and development process of microbial mat construction through geological record remains difficult to solve. However, the study of the microbial mat construction with modern and ancient stromatolites as a substitute shows that its complex microbiological metabolism is often used as a reaction for carbonate precipitation [33].

Dolomite deposits formed in the modern tidal flat sedimentary environment are closely related to microbiological mats. Extracellular polymers within microbes complexate calcium and magnesium and regulate the alkalinity of deposition environment through organic carbon respiration to promote the crystallization of disordered dolomite [34]. In 2010, Tomaso et al studied carbonate deposition in Abu Dhabi, United Arab Emirates. Geochemical analysis and sediment composition of three sites with longitudinal depth were carried out in this experiment. It is demonstrated that microbiological metabolism promotes carbonate mineralization [35]. The precipitation of microbial dolomite and microbial activity are closely related to the rising alkalinity of the pH in the water environment, which can be used to explain the occurrence of the microbe in the microbe between the high-tide zone and the subtidal zone in the depositional environment of the sea. The disordered Dolomite Precipitation in microbial mat. Tomaso and others believe that "the Dolomite Precipitation in this area is closely related to the burial process of the microbial mat", and the Dolomite Precipitation in the buried microbial mat and EPS: exopolymeric substances in microbial mat are closely related [35]. The absence of apparent biological activity in a highly evaporated burial microbial mat does not mean that the process of dolomitization here is an "abiotic" process. Instead, dolomite was precipitated on the basis of the supersaturation of carbonate in the environment and by the inactive microorganism in the "production workshop" [36, 37, 38].

However, the mechanism of dolomite production and the process of microbes participating in mineralization in tidal flat environment are not clear. In order to study the formation of dolomite in high salinity microbial mats in Lagos, Brazil, we inferred that the active carbonate was undersaturated caused by the oxidation of sulfides during the formation of dolomite [39]. There may be reactions between sulfur ions or some organic molecules that facilitate the dehydration of magnesium ions in sedimentary environments, In order to ensure that the nucleation stage of mineral genesis. In 2015, Petrash fully affirmed that the formation of dolomite in the modern tidal flat is closely related to buried microbes. The mechanism of extracellular polymers complexing calcium and magnesium and regulating the alkalinity of depositional environment to promote the nucleation of disordered dolomite by organic carbon respiration were also discussed. The observation of the burial of dolomite deposits in microbes in the high-salinity lagoon of the Lothrox Islands, Venezuela was carried out by high resolution chemical stratigraphic analysis, combined with synchrotron radiation analysis and microbial surface characterization analysis (electrical potential and FTIR), analyzing the electrical potential of electron receptors besides sulphides during dolomite formation. The study found that there is a tightly bound redox metabolic cycle of manganese ions and sulfides during the decay of bioactivity in rapidly buried mats (Figure 2). In addition, there are considerable dolomite precipitates in manganese ion redox sites in the sedimentary environment [40].

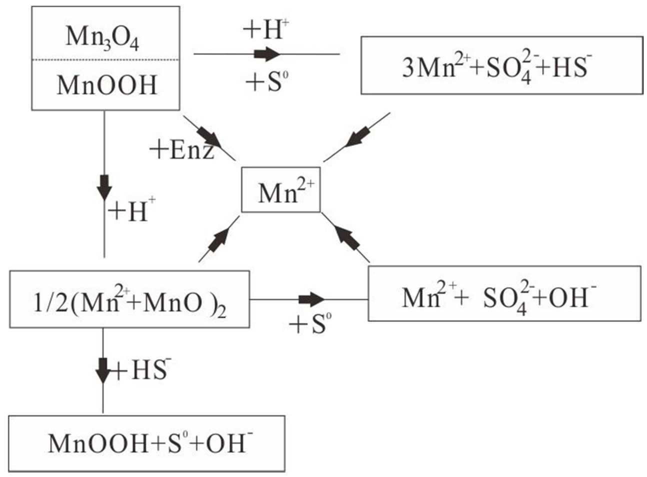

Figure 2. *Sketch diagram of Mn-S redox cycling reaction.*

The results are in agreement with the results of some studies on controlling the mineralization rate of organic carbon by affecting the activity of fine bacteria in some marine sedimentary environments [36]. Petrash's experimental observations provide sufficient evidence for the theory that the bioactivity decline of buried microbial mat produces a metabolic cycle of nonequilibrium geochemical metal ions. The oxygen reduction of manganese ion and the interaction between organic molecules have positive significance to the reduction of sulfate, which not only enhances the degradation of organic matter but also maintains the basicity and ph value of dolomite precipitation. This conclusion is undoubtedly an improvement to the theory of microbe mats precipitating dolomite, and the Mn-S redox cycle complements the reaction between sulphide and organic molecules.

It can be concluded that the primary dolomite precipitated by Mn-S redox cycle can be summarized in three aspects: (1) the microbial mat weakens the water molecule to magnesium ions and manganese ion by extracting biocatalytic action with high reactive activity. (2) the imbedded microorganism passed through the influence of organic molecules to control the non-autotrophic reaction. At the same time, elevating alkalinity promotes magnesium ions into carbonate to construct lattice. (3) On the basis of fast burial, the rich manganese ion and diffusion-limited biofilm substrate was brought into the reduced sulfate region and the alkalinity of the surrounding environment was improved by the autocatalysis of the Mn-S redox reaction cycle. The effect of the redox cycle of Mn-S on the pH and alkalinity in the local area is obvious during the burial of the microsheet, and the high ratio of mg and Ca is observed in the lagoon environment. It can be used as a prerequisite for the early formation of dolomite in shallow diagenetic environments (Figure 3).

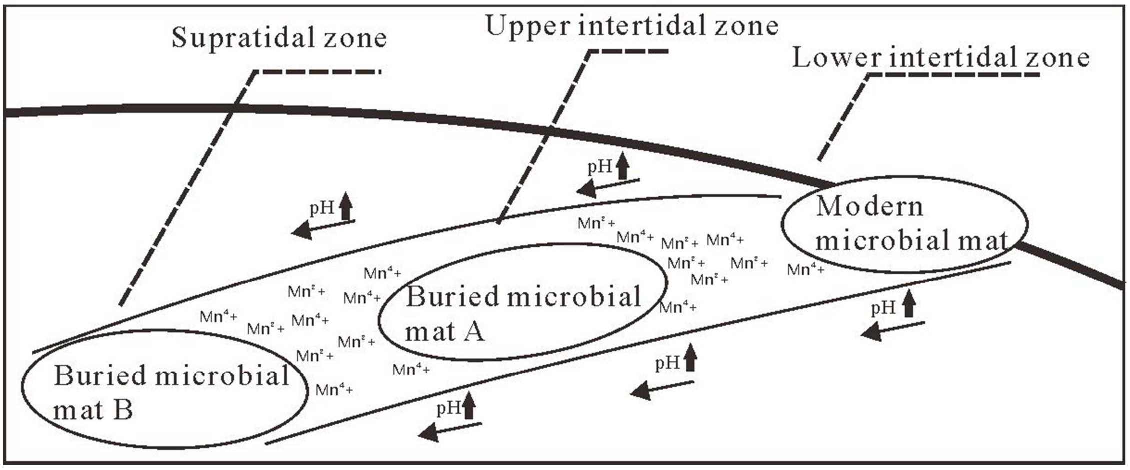

Figure 3. *Mn-S redox cycling in the buried microbial mat.*

5. Discussion

Since the first confirmation of dolomite by Van Tuyl in 1916, to the famous experiment of Land [1], to the introduction of the concept of microbial dolomite to Vasconselos et al. [6, 7], the research on dolomite has been studied for 100 years. For decades, SRB-led sulfate reduction, methanogenic bacteria in the marine environment, organic molecular hydrolysis to promote dolomite precipitation which provides a clue and thinking way for exploring the possible deposit of primary dolomite in the surface environment. The dissolution of sulfides promotes dolomite deposition through a dual mechanism, although it complements the understanding of the mechanism of sulfate reduction, but the following problems arise: it is the sulfate reduction itself that promotes dolomite precipitation. The reaction products promote dolomite precipitation. Although these studies also approximately answer the question that there may be primary dolomite precipitation in the surface environment, But a variety of microbial metabolism, such as sulfate-reducing reaction [3, 6, 7], methanogenesis, sulfide oxygenation [38], urea hydrolysis [22], And the discovery of the precipitation of primary dolomite in this special environment [34] and the Mn-S redox cycle [40] in its burial process, and even the effect of dissolving sulfides on the precipitation of primary dolomite, indicating the complex nature of the microbial process and its connection, and some questions that need to be studied. It is still the most important problem to determine and identify the mechanism of microbial action for the formation of primary dolomite in geological records.

Although the research of microbial dolomite model has made great progress, the primary dolomite produced by the simulated precipitation experiment in the laboratory still faces two questions: 1) mineralogical query of microbial dolomite; 2) sulfate reduction promote calcium carbonate precipitation or not. Mineralogical study reports that dolomite ($CaMg(CO_3)_2$) performed as a rhombohedral carbonate mineral having a structure that consists of an ordered alternation of layers of Ca^{2+} and Mg^{2+} ions interspersed with CO_3^{2-} anion layers, all normal to the c-axis, giving the mineral R-3 symmetry and display some degree of cation disorder (Ca2+ on Mg2+ sites and vice versa). It means that these laboratory products show no evidence of cation ordering and appear to be very high-magnesium calcite [41]. Beyond that, sulfate reduction has been suggested as a mechanism to induce precipitation of calcium and magnesium carbonates in marine sediments and microbial mats through most of Earth's history. However, sulfate reduction also causes a drop in pH that favors dissolution rather than precipitation of carbonates [42]. With increasing salinity, sulfate reduction becomes even less efficient to induce carbonate precipitation. Especially in an alkaline, like Precambrian ocean, where large amounts of carbonate were formed, induction through sulfate reduction was entirely ineffective [42]. Although this view has been refuted in experimental methods and the types of organic matter in the metabolism of sulfate reduction reaction [43]. However, the simulation results based on laboratory environment still can not fully represent the complex conditions of sulfate reduction in natural environment.

Just as Dupraz emphasizes, how to traverse "diagenetic filter" to find evidence related to microbes in geological records, understanding the interaction between microbes and minerals is the key to explain rock records [44]. The study of microorganism dolomite model still needs to be further explored. Any single dolomite genetic model for the interpretation of the common dolomite strata shows a certain local limit. The stratigraphic sequence of dolomite in the natural environment, which is thousands of meters thick, shows that the formation of dolomite is indeed a complex sedimentological problem, for example: dolomite of Cambrian massive dolomite in thousands of meters Loushanguan Group [45], massive dolomitic limestone from Wumishan formation of Proterozoic whose depth could be of dozen thousands, North China Platform [46], Selective dolomitization in the central part of the Cambrian Fengshan formation along the microbe crypt in the Xiaweidian section of the western suburb of Beijing (Figure 4 (a)), and in this section, the lithology at the top of Cambrian changes gradually from limestone to dolomite, which shows that dolomitization is closely related to exposure (Figure 4 (b), further proves the complexity of diagenetic and sedimentary environments of dolomitization. Therefore, the microbial dolomite (rock) model is used to analyze the ancient massive dolomite strata, especially those that have been demonstrated to be related to the shallowness of the sedimentary environment. Further exploration is needed to give a more reasonable explanation.

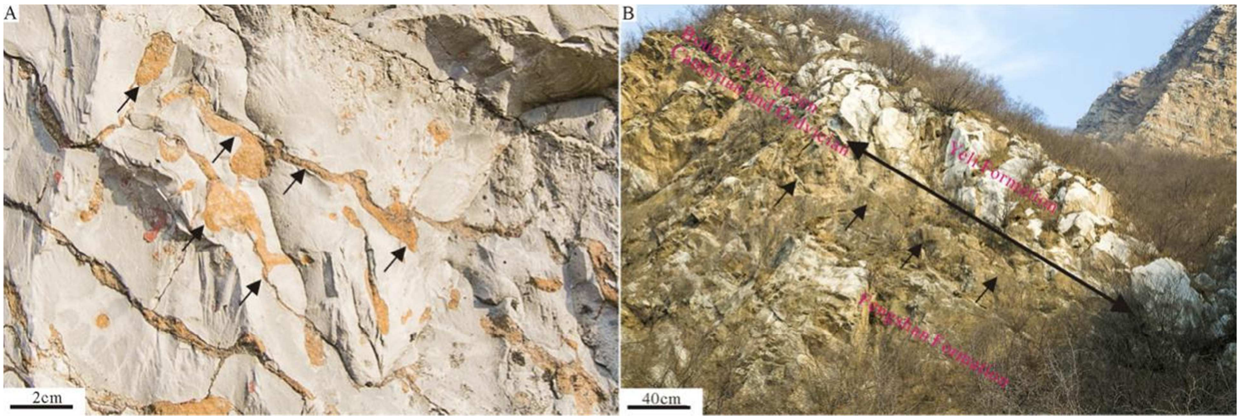

Figure 4. *Two special dolomitized phenomena in Cambrian Fengshan formation in the western suburb of Beijing.*

A-Black arrows show the dolomitization in dwelling burrow; B-Black arrow show the depositional environment tends (From deep to shallow), dolomitization increasing.

6. Conclusion

The introduction of microbial processes into the study of sedimentary primary dolomite in the surface environment and the establishment of microbial dolomite (rock) model represent an important advance in the study of "dolomite problems in sedimentology". These fruitful studies fully show that the microbial metabolism involved in promoting dolomite precipitation has the characteristics of diversity. However, the formation of dolomite has complex and multiple mechanisms, and most of the dolomites recorded in the stratigraphic records are the product of early diagenetic metasomatism, and buried hydrothermal dolomitization is also more common. More geological phenomena also illustrate the complexity of the dolomite problem. This means that any single dolomite genetic model to interpret complex phenomena in geological records has limitations. Therefore, in order to give a reasonable answer to the phenomenon of dolomite attenuation in geological history, it is necessary to interpret different dolomite action mechanisms in combination with different geological ages, different sedimentary and diagenetic environments, therefore achieving a thorough solution to the "Dolomite problem".

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