DISSOLUTION OF HIGH-MG CALCITE FOSSILS AND THE FORMATION OF BIOMOLDS DURING MINERALOGICAL STABILIZATION

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ABSTRACT: Biomolds formed by the dissolution of high-Mg calcite (HMC) porcellaneous foraminifera occur in Holocene limestones from the Schooner Cays, Bahamas, and in Pleistocene limestones from the Belmont Formation of Bermuda and the Lucayan Formation of the Bahamas. In all of these cases, the HMC biomolds are associated with either a water table or a subaerial exposure surface. A single foraminiferal test may exhibit both a moldic fabric and a low-Mg calcite (LMC) recrystallization fabric that preserves original texture. Other types of HMC bioclasts, such as red algae and echinoids, do not exhibit biomolds but are mineralogically stabilized.

The fact that foraminifera are more susceptible to dissolution and biomold formation than other types of HMC bioclasts indicates that an intrinsic factor, such as skeletal microstructure, is partially responsible. However, because both mold formation and texture-preserving recrystallization may occur in the same grain, intrinsic properties of the bioclasts alone do not determine the alteration pathway. An extrinsic factor is also implicated. It is proposed that this factor is CO_2 flux across diagenetic interfaces during mineralogical stabilization. Abrupt and large influxes of CO_2 across a subaerial exposure surface or water table will reduce saturation states in bulk pore waters and within skeletal grains. Recrystallization by spatially and temporally synchronous dissolution and precipitation, which is the normal HMC alteration process, will cease and only dissolution of HMC will occur. Bioclasts with high reactive surface areas, such as foraminifera, will be most susceptible to this dissolution. Once the CO_2 influx has been buffered, HMC-to-LMC recrystallization will resume. In this manner, an originally HMC bioclast can exhibit the products of both dissolution and recrystallization pathways. This would be particularly true if recharge, and thus CO_2 flux, is a seasonal phenomenon, as is the case in most carbonate regimes.

INTRODUCTION

Mineralogical stabilization of carbonate sediments involves the conversion of metastable depositional mineralogies to stable, low-Mg calcite (LMC). This process, which is greatly facilitated by subaerial exposure and freshwater influx, involves the dissolution of the metastable phase and the subsequent precipitation of the stable phase. In aragonitic grains, mineralogical stabilization is known to have two endmember pathways. The dissolution of aragonite and precipitation of calcite may be distinctly different events in time and/or space. If so, a mold of the former grain typically develops, resulting in no preservation of the original grain's texture or chemistry (e.g., Friedman, 1964; Land et al., 1967). In the other endmember situation, aragonite dissolution and calcite precipitation will be nearly simultaneous and separated in space by only a thin film of water (e.g., Pingitore, 1976, 1982; Wardlaw et al., 1978). In such a case, there is likely to be varying degrees of textural and geochemical preservation (e.g., Sandberg and Hudson, 1983; Martin et al., 1986; Brand, 1989).

In contrast to aragonite, high-Mg calcite (HMC) grains tend to exhibit but one alteration pathway. The dissolution of HMC and the subsequent precipitation of a calcite phase with less Mg (i.e., incongruent reaction) occurs primarily across thin reaction films, with the two reactions barely separated in time or space (e.g., Land, 1967; Richter, 1979; Brand and Veizer, 1980; Turner *et al.*, 1986). This HMC-to-LMC recrystallization usually results in excellent preservation of textural relationships, at least at the thin-section scale (e.g., Friedman, 1964; Land *et al.*, 1967; Gavish and Friedman, 1969; Sandberg, 1975). Molds formed by the dissolution of the entire HMC phase are rare; Longman (1980) and Harris *et al.* (1985) have both suggested that HMC biomolds never occur.

The objective of this paper is to report on three occurrences of HMC biomolds that either formed or are currently forming as a result of mineralogical stabilization in freshwater. All three examples are of Quaternary age. An hypothesis regarding their significance is then developed based on the stratigraphic and hydrologic occurrences of these HMC biomolds and two additional case examples reported in the literature.

ANALYTICAL METHODS

HMC bioclasts were examined in epoxy-impregnated and polished thin sections using transmitted light, back-scatter electron microscopy (BSEM), and electron microprobe (EMP) techniques. Magnesium contents were determined by the EMP analysis; detection limits were 0.03 weight % MgO, which is 0.05 mole % Mg. In most cases, each thin section sample was first examined in transmitted light, then carbon coated and studied by BSEM and EMP. Select sections were then



Figure 1. Scanning electron photomicrograph of pristine rod-like HMC crystals (arrows) in the test of a modern, unaltered porcellaneous foraminifera. Note the random array of the HMC crystals. This foraminifer was hand-picked from a subtidal sand sample that was collected along the barrier-reef tract of Belize. Photograph is of a broken and lightly etched surface; scale bar equals $2 \mu m$.

repolished with 0.25 μ m grit, lightly etched in 0.1% acetic acid for 15 to 25 seconds, gold-coated, and then studied by scanning electron microscopy (SEM).

TYPICAL HIGH-MG CALCITE GRAIN FABRICS

Porcellaneous foraminifera are the dominant grain exhibiting HMC biomolds in all three of the Quaternary occurrences reported in this paper. The unaltered, original fabric of such grains, and the typical fabrics produced with mineralogical stabilization to LMC, are thus reviewed prior to describing the moldic fabrics and their occurrences.

The microstructure of unaltered porcellaneous foraminifera is a three-dimensional array of densely packed, randomly oriented crystals with a surface veneer of tangentially orientated crystals (Towe and Cifelli, 1967; Hemleben, 1969). Individual crystals are rod-like with lengths of 1.5 to 2.0 μ m (Fig. 1). In thin section, the randomly orientated, micron-sized crystals produce the honey-brown color and homogenous porcellaneous texture that is characteristic of such grains. The calcite crystals are HMC with Mg contents that range from 13 to 16 mole % Mg (Chave, 1954; Blackmon and Todd, 1959; Budd, in press). The variation in Mg content is partially related to ambient water temperatures and partially generic (Chave, 1954; Blackmon and Todd, 1959; Milliman, 1974).

The mineralogical stabilization and textural recrystallization of porcellaneous foraminifera is typically a multi-stage process. Initially, the randomly orientated, rod-like crystals recrystallize to LMC without any electron-optically visible alteration in crystal morphology (Towe and Hemleben, 1976; Budd, in press). Subsequently, the rod-like crystals of LMC may recrystallize to equant crystals of minimicrite, micrite, or microspar (Towe and Hemleben, 1976). The products of the second recrystallization are the normal end-product observed in ancient porcellaneous foraminifera.

NEW OCCURRENCES OF HIGH-MG-CALCITE BIOMOLDS Holocene Schooner Cays, Bahamas

Freshwater diagenesis is an active process on small Holocene ooid-sand islands in the Schooner Cays, Bahamas. The location, geology, and hydrology of these islands was described by Budd (1988a) and the dissolution of aragonite and precipitation of LMC cements was documented by Budd (1988a, b) and Budd and Land (1990). The islands have formed atop ooidsand shoals in the last 700 years. Their freshwaterphreatic zones are up to 1.25 m thick and their freshwater-vadose zones range from 0.5 to 1.0 m in thickness. Skeletal grains are minor constituents of the sediments, comprising less than about 5% by volume. The main skeletal grains are Halimeda, mollusks, soritid foraminifera, and echinoids, with the former two grain types being dominant. The HMC skeletal grains average only about 0.8% of the sediment by volume.

Most HMC bioclasts in freshwater-vadose and freshwater-phreatic samples from the Schooner Cays exhibit no textural differences from their unaltered precursors. Foraminifera, for example, reveal a typical porcellaneous texture in thin section (Fig. 2A). Electron microprobe analyses, however, reveal that these foraminifera have average Mg contents that range from 5.9 to 14.1 mole % Mg. Unaltered foraminifera from the adjacent subtidal environment have an average Mg content of 14.8 mole % Mg. The HMC bioclasts in the freshwater diagenetic zones are clearly in the midst of HMC-to-LMC recrystallization, as discussed in detail by Budd (in press).

A second HMC stabilization pathway also occurs in the Schooner Cay samples. Foraminifera collected at water-table sites on one of the islands exhibit dissolution of the HMC crystals and the formation of biomolds. A single occurrence of a partially dissolved echinoid fragment was also noted in one of these watertable samples. Dissolution fabrics include complete foraminiferal biomolds (Fig. 2B), partial biomolds (Fig. 2C), and delicate intercrystal micropores within shell walls. The latter two fabrics are most common with most foraminifera exhibiting a combination of partial molds, microporous shell wall, and undissolved shell wall (Fig. 2C). Mg contents of the calcite in the undissolved portion of the tests range from 6 to 8 mole % Mg, indicating that these portions of the tests are undergoing texture-preserving recrystallization (Budd, in press).

No relationship was observed between the presence or absence of foraminiferal biomolds and the presence or absence of either oolitic coatings around foraminiferal tests, interparticle calcite cements around the tests, or cement or carbonate mud in intraparticle pores



Figure 2. Dissolution fabrics in Holocene HMC foraminifera from the Schooner Cays, Bahamas. A) The foraminiferal grain in this thinsection photomicrograph shows no visible sign of dissolution or recrystallization; the test exhibits the typical porcellaneous texture of soritid foraminifera. Average Mg content of this test, however, is now only about 7.0 mole %, as determined by electron microprobe analysis. Primary intraparticle pores are filled with LMC cement (c) and the

within tests. For a minifer a collected in the vadose zone, or in the phreatic zone at depths greater than 20 cm below the water table, were never observed to exhibit either moldic or partial moldic pores.

Pleistocene Belmont Formation, Bermuda

The second example of HMC biomolds occurs in the Pleistocene Belmont Formation of Bermuda. The geology and diagenetic history of these rocks at Devonshire Bay, Bermuda, were described by Land (1970). A paleo water table at this locality subdivides an ancient eolianite; its upper portion has experienced only vadose-zone diagenesis whereas its lower portion experienced a period of phreatic-zone diagenesis during Belmont time. A subaerial exposure surface caps the entire eolianite deposit, and in turn is overlain by younger Belmont beach or eolian deposits. The freshwater-phreatic diagenesis of the older eolianite presumably occurred in a coastal freshwater lens formed during deposition of the younger Belmont deposits (Land, 1970).

Samples of the older eolianite that were collected above the paleo water table and within one meter of the intra-Belmont exposure surface (site 65 of Land, 1970) contain many foraminifera that are complete biomolds (Fig. 3A). In thin section, these foraminifera are outlined by interparticle LMC cements and their internal architecture is defined by LMC cement in the foraminiferal chambers (Fig. 3B, C). Not all foraminifera in these samples, however, occur as biomolds. Some exhibit a preliminary state of dissolution, namely a combination of partial molds and delicate micropores between undissolved crystals (Fig. 3D). Others exhibit complete and excellent preservation of their porcellaneous microstructure (Fig. 3C). The latter tests are LMC, indicating that they have followed the recrystallization pathway.

No relationship was observed between the presence or absence of foraminiferal biomolds and the presence or absence of either interparticle calcite cements around the tests, or cement or carbonate mud in intraparticle pores within tests. Other HMC bioclasts, such as echinoids and red algae, exhibit no biomoldic pores; they apparently stabilized only by texture-preserving recrystallization.

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test has an ooid coating (arrow). Scale bar is 200 μ m. **B**) Thin-section photomicrograph of a foraminiferal grain in which the original skeletal material has dissolved to create a moldic fabric (m). Intraparticle and surrounding interparticle pores are filled with LMC cement (c). Scale bar equals 50 μ m. **C**) Back-scattered electron photomicrograph of an epoxy-impregnated and polished foraminiferal grain that exhibits partial dissolution. Within the test, black is epoxy-impregnated mold (m) and light gray is undissolved shell wall (s). Darker gray shades (arrows) represent microporous areas where epoxy-filled crystal molds (micropores) are intercalated with the undissolved calcite crystals. Scale bar is 50 μ m.



Figure 3. Dissolution fabrics in the Pleistocene Belmont Formation, Bermuda. A) Back-scattered electron photomicrograph of an epoxy-impregnated and polished foraminiferal grain. Calcite cement in interparticle, intraparticle, and microboring pores is white, epoxy-filled pores are black. Complete dissolution of the HMC foraminiferal test has produced a biomoldic pore. Scale bar equals $150 \mu m$. B) Thin-section photomicrograph of extensive moldic porosity (m) in a foraminiferal grain. The identity of this grain as a foraminifer is possible only because of the LMC cement and micrite in original intraparticle pores. Scale bar is $50 \mu m$. C) Adjacent foraminiferal tests in this thin section photomicrograph exhibit distinctly different alteration pathways. The grain to the left has been extensively dissolved, producing moldic porosity (m) within the original shell wall. In contrast, the grain to the right is now 100% LMC and exhibits no moldic porosity. Scale bar is $100 \mu m$. D) Scanning electron photomicrograph of an epoxy-impregnated, polished, and lightly etched foraminiferal grain. The grain exhibits LMC cement in intraparticle pores (c), moldic pores (dark epoxy, m), and zones of delicate intracrystal microporosity (light gray shell walls, mp). The microporous zones consist of both epoxy-filled crystal molds and undissolved calcite crystals. Moldic pores form as micropores coalesce (arrows) with additional dissolution. Scale bar equals $50 \mu m$

Pleistocene Lucayan Formation, Bahamas

The third example of HMC biomolds comes from the Pleistocene Lucayan Limestone (Beach and Ginsburg, 1979) of the Bahamas. The shallow-subtidal deposits examined were recovered in engineering borings taken in Bell Channel near Freeport, Grand Bahama Island. One boring extends to 120 ft (36.6 m) and contains numerous subaerial exposure surfaces, as de-



Figure 4. Thin-section photomicrographs of dissolution fabrics in the Pleistocene Lucayan Formation, Grand Bahama Island, Bahamas. A) Foraminiferal grain that exhibits partial moldic pores (arrows) separated by micritic LMC (darker gray, s). Scale bar is 100 μ m. B) Close-up of grain shown in A. Photo was taken with a red filter so that pores filled with blue epoxy are black (Tapp and Prezbindowski, 1990). The grain is clearly part mold (m) and part micrite (s); there is no microporosity in this example. Scale bar is 50 μ m.

fined by the criteria of Perkins (1977). Thin sections cut from core samples 5 ft (1.5 m) or less below one such surface at about 60 ft (18.3 m) exhibit partial biomolds of foraminiferal grains. Echinoid and red algae grains exhibit excellent textural preservation and obviously experienced the HMC-to-LMC recrystallization pathway.

Only partial molds of foraminiferal grains occur in this case example. Individual patches of moldic porosity within grains tend to be about 10 to 40 μ m in length with a width equal to that of the foraminiferal shell wall (Fig. 4A). Interspersed between these partial dissolution molds is micritic LMC (Fig. 4B). Presumably this micritic LMC is the recrystallization successor to the original HMC. The very small crystal size of the LMC argues against it being a mold-filling cement.

As in the Schooner Cays and Bermuda examples, the Bahamian Lucayan foraminifera exhibited no relationship between the presence, absence, or distribution of biomolds and the presence, absence, or distribution of either interparticle calcite cements around the tests, or cement or carbonate mud in intraparticle pores within tests. In contrast to these other case examples, however, no microporous intracrystalline dissolution fabric between undissolved HMC crystals was observed in any of the Lucayan foraminifera.

DISCUSSION

The formation of biomolds in HMC skeletal grains is an atypical reaction path in Quaternary limestones (James and Choquette, 1984); to my knowledge, the only previous reports are those of Schroeder (1979) and Saller (1984a). As in this study, both of these authors described numerous types of HMC fossils, including foraminifera, that exhibited two stabilization pathways: dissolution to form biomolds and texturepreserving recrystallization to LMC grains. Schroeder's (1979) examples are from Kenya, and Saller's (1984a) examples are from Enewetak. The three examples described in this paper, combined with the work of Schroeder (1979) and Saller (1984a), clearly indicate that molds of HMC skeletal grains do form during mineralogical stabilization, and that their occurrence is geographically widespread.

Two observations are critical to understanding the origin and significance of HMC biomolds. The first is that porcellaneous for aminifer appear to be more susceptible to dissolution and biomold formation. Biomolds of other types of HMC bioclasts can occur as evidenced by the observations of Schroeder (1979) and Saller (1984a), but the data presented in this paper clearly indicate that foraminiferal grains are the most likely type of HMC bioclast to form biomolds. This observation suggests that factors intrinsic to each type of HMC skeletal grain play a role in the formation of HMC biomolds. Such factors might include skeletal microstructure (Walter, 1985), crystallographic heterogeneity (Bischoff et al., 1983), compositional variability (e.g., Schroeder, 1969; Moberly, 1970), or bound water (Gaffey, 1988).

The second observation critical to understanding HMC biomold formation is the fact that both dissolution to a biomold and texture-preserving recrystallization pathways can occur in the same foraminiferal grain (e.g., Figs. 2, 3, and 4). Each of the two different products may occupy significant portions of a grain with areal dimensions of tens of microns to millimeters. Such dimensions are far greater than the dimensions of known structural and chemical elements of HMC bioclasts (e.g., Bischoff *et al.*, 1983). Therefore, the determinative factor between dissolution and recrystallization pathways of HMC stabilization cannot be an intrinsic factor such as fluid chemistry and/or flux are also implicated.

The spatial distribution of HMC biomolds in the Holocene Schooner Cays and Pleistocene limestones suggests that they preferentially form in the vadose zone, particularly at its upper and lower diagenetic interfaces: the subaerial exposure surface and water table. The Schooner Cay samples that exhibited foraminiferal biomolds were all collected at the water table. The Pleistocene examples from Bermuda and the Bahamas were from within 1.5 m of paleo subaerial exposure surfaces. Schroeder's (1979) samples consisted of marine-cemented Holocene beachrock that had subsequently undergone meteoric diagenesis, presumably implicating a subaerial exposure surface. Saller's (1984a) data on the stratigraphic distribution of HMC biomolds and cement fabrics (his Figs. 10-12, 40 and 42) indicate that HMC biomolds in Pleistocene limestones on Enewetak are more likely to occur in samples dominated by vadose-zone alteration at or slightly below subaerial exposure surfaces.

Given the association between meteoric diagenetic interfaces and HMC biomolds, it is proposed that large and abrupt influxes of CO₂ at or near these interfaces is the extrinsic driving factor in the formation of HMC biomolds during mineralogical stabilization. An abundance of metastable mineralogies will normally mean that pore waters are saturated with respect to LMC (e.g., Budd, 1988b). However, an abrupt influx of CO₂ could create a "sudden" undersaturation with respect to LMC. The decrease in saturation state would cause an exponential increase in the dissolution rate of HMC (Walter, 1985) but preclude LMC precipitation. The HMC alteration path would thus change from the normal recrystallization reaction (i.e., incongruent dissolution + precipitation) to merely a dissolution reaction (i.e., congruent dissolution). Once intragranular fluids achieved equilibrium with respect to LMC, mineralogical stabilization by incongruent reactions (i.e., recrystallization) would be resumed. Although the duration of congruent HMC dissolution would be short. the amount of dissolution would only be dependent on the amount of CO₂ introduced. HMC bioclasts with relatively large reactive surface areas, such as foraminifera, would be more prone to dissolution and mold formation (Walter, 1985), but all types of HMC could

dissolve if the degree of undersaturation was sufficiently large.

The range in dissolution fabrics observed, particularly in foraminifera from the Schooner Cays and Bermuda, probably represents various stages of HMC dissolution and biomold formation. The tests of porcellaneous foraminifera are initially composed of densely packed, randomly orientated arrays of micronsized rods of HMC crystals (Towe and Cifelli, 1967; Hemleben, 1969). Dissolution of HMC within a foraminiferal test would thus involve the dissolution of these crystals and the formation of a small micropore in its place. Initially this would yield the microporous dissolution fabric (Figs. 2D and 3D). Repeated CO₂ influxes and the resultant dissolution of HMC crystals over long periods of time would eventually result in the coalescence of these micropores into partial biomolds (Figs. 2C and 3D), and eventually into complete biomolds (Figs. 2B and 3). According to this interpretation, the microporous dissolution fabric of HMC foraminifera would have certain similarities to partial, delicate intrafabric aragonite dissolution fabrics described by Gvirtzman and Friedman (1977), Goter and Friedman (1987), Budd (1988a), and Saller and Moore (1989).

Diagenetic interfaces would be the most likely sites of large CO₂ fluxes in the meteoric realm, and recharge may be the factor that makes the fluxes "sudden". At the subaerial exposure surface (or anywhere else in the vadose zone) recharge might emplace a CO₂-rich fluid where previously the pores had been dry. At the water table, O₂-rich recharge could cause a rapid increase in the oxidation of organic matter within the sediment, and thus a sharp increase in CO₂. Alternately, if rainwater reached the water table without attaining equilibrium with respect to the CO₂ content of the soil or vadose zone, then the subsequent transfer of CO₂ across the water table could also create a rapid and large CO₂ influx. Such a phenomenon could easily occur in settings similar to that in the Schooner Cays where the vadose zone is thin and the sediments are highly permeable (Budd, 1988a). Fluid infiltration rates to the water table would easily exceed the relatively slow kinetics of CO₂ transfer from a gas to a liquid (Matthews, 1974).

Seasonal recharge could explain why an individual foraminifer can exhibit both biomoldic pores and incongruent alteration to LMC. Schooner Cay pore waters were undersaturated with respect to LMC only in association with meteoric recharge during the fall rainy season; pore waters were saturated to slightly oversaturated with respect to LMC the remainder of the hydrologic cycle (Budd, 1988b). Biomoldic porosity would form in association with fall recharge. During the remainder of the hydrologic cycle, however, dissolution of the HMC phase would be accompanied by the precipitation of LMC, thus yielding a preservation of texture and no biomold. Individual foraminiferal grains at diagenetic interfaces could thus evolve through the two different diagenetic pathways during the course of multiple hydrologic cycles. As stressed by Matthews (1974) and Schroeder (1979), seasonal variation allows different solution and precipitation phenomena to be superimposed one on the other while the sediment remains in essentially the same diagenetic environment.

If the proposed explanation for the formation of HMC biomolds during mineralogical stabilization is correct, then the rarity of HMC biomolds in the rock record may actually reflect the rarity of sampling not only a hydrologic interface, but one in which CO₂ additions were sufficiently "extreme". Conversely, the occurrence of HMC biomolds in the rock record may be an indirect proximity indicator of an ancient water table, subaerial exposure surface, or vadose zone. Caution must be exercised however, because originally HMC allochems that are stabilized to LMC may still undergo fabric-selective dissolution in other diagenetic situations (James and Choquette, 1984), such as with convecting cold seawater (Saller, 1984b) or in the deep burial environment (e.g., Donath et al., 1980; Mazzullo, 1981; James and Klappa, 1983).

A final point is the implication of HMC biomolds to the determination of ancient skeletal mineralogies. Sandberg (1983, 1984) proposed that the occurrence of skeletal molds or calcite-filled molds was one of six criterion for recognition of ancient aragonite. Sandberg (1983, 1984) acknowledged that it was an equivocal criterion because molds could form from calcitic components, even in undolomitized rocks. The observations of this study strongly support such a qualification, and in fact, suggest that molds or calcite-filled molds should be removed from any list of criteria for the recognition of ancient aragonite. HMC biomolds do occur and are, in fact, more abundant in Quaternary carbonates than implied in the literature. Thus, a moldic fabric alone does not mean an originally aragonite mineralogy.

CONCLUSIONS

HMC bioclasts, like aragonite bioclasts, can experience two types of alteration pathways during mineralogical stabilization. The most common pathway is the well-documented microscale dissolution and precipitation pathway (i.e., recrystallization) that preserves texture at the macroscale. Dissolution of HMC crystals, yielding microporous fabrics and/or complete biomolds is the second pathway. Although rare, the dissolution pathway has now been documented in Pleistocene limestones from Bermuda, the Bahamas, and Enewetak, and in Holocene limestones from Kenya and the Schooner Cays, Bahamas.

Porcellaneous foraminifera appear to be more susceptible to dissolution and biomold formation than other types of HMC bioclasts. This suggests that a factor intrinsic to HMC skeletal grain types effects the dissolution process. Reactive surface area, a function of skeletal microstructure, is the most likely intrinsic factor to effect biomold formation.

The evidence presented herein also indicates that

formation of HMC biomolds is related to meteoric interfaces, such as subaerial exposure surfaces and water tables. These interfaces could be the loci of sudden CO_2 influxes related to meteoric recharge. Abrupt and sudden CO_2 influxes would temporarily cause dissolution of HMC. Texture-preserving recrystallization would resume once the CO_2 charge was buffered. Microporous fabrics are the initial dissolution product; repeated dissolution events, however, eventually lead to the coalescence of micropores and the formation of complete biomolds. Seasonal recharge would result in the juxtaposition of both biomolds and texture-preserving stabilization products.

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