



The modern record of “concretionary” carbonate: Reassessing a discrepancy between modern sediments and the geologic record

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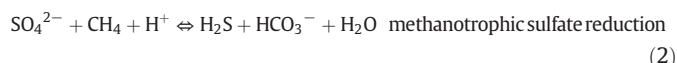
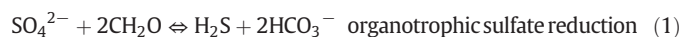
ABSTRACT

Carbonate concretions have long been recognized as common constituents of sedimentary rocks exposed in outcrop. Although formation mechanisms vary, organic matter degradation often plays a role in carbonate concretion authigenesis, providing a source of alkalinity for mineral production. Perhaps surprisingly, modern examples of concretionary carbonates are rarely identified in cores despite ubiquitous organic matter degradation in marine sediments. This has prompted the general notion that the modern record of concretionary carbonate does not mirror that preserved in the geologic record. Here, we present data from continental margin sediments that receive enhanced delivery of organic matter and demonstrate that concretionary carbonates are indeed recovered from many of these sites. In addition, sites where concretionary carbonates have not been recovered exhibit chemical profiles that suggest local authigenesis. The lack of concretion recovery from some locations likely reflects the poor potential for modern coring techniques to adequately capture concretions or provide context for their identification once recovered. Concretionary carbonate recovered from the Peru Margin, Cascadia, the Gulf of California, the Cariaco Basin and the western margin of southern Africa exhibit evidence for modern or very recent formation via methanogenesis or the anaerobic oxidation of methane. Demonstrably modern organotrophic sulfate reduction concretions have not been found.

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1. Introduction

Authigenic carbonates in the form of nodules, concretions and cemented layers comprise a significant lithologic component of some ancient sedimentary rock units (e.g., the Miocene Monterey Formation; Bramlette (1946)). Modern marine concretionary carbonate formation has long been interpreted as non-reflective of the extensive geologic occurrence (Raiswell and Fisher, 2000; Wetzel, 1991). Apparently, true modern concretions have only been documented from non-marine deposits including marsh (Allison and Pye, 1994; Pye, 1981), intertidal (Al-Agha et al., 1995) and fresh water (Ho and Coleman, 1969) environments. Extensive geochemical analyses of concretionary carbonates (both marine and non-marine) have revealed that the microbial degradation of organic matter plays an important role in mineral precipitation (Curtis et al., 1972; Irwin et al., 1977; Loyd et al., 2012; Mozley and Burns, 1993; Raiswell and Fisher, 2000). Specifically, anaerobic reactions involving bacterial sulfate reduction (BSR) and methane production are interpreted to have led to carbonate authigenesis largely because these reactions generate alkalinity (Coleman et al., 1993), as follows:



BSR (Eqs. (1) and (2)) and methanogenesis (Eq. (3)) are common in modern marine sediments, particularly in those under highly productive, upwelling water masses (note, organotrophic sulfate reduction excludes methane here). Therefore, the apparent modern under-representation of marine concretionary carbonate does not match the well-documented extent of concretion-yielding processes.

Here, the modern record of concretionary carbonate is reassessed with specific search criteria applied to Ocean Drilling Program (ODP) and Deep Sea Drilling Program (DSDP) site sediment and pore water data. Sediments along western continental margins experiencing high productivity due to extensive upwelling and sites exhibiting anoxic sediments due to basin restriction are targeted because of the common occurrence of BSR and methanogenesis. It is demonstrated herein that concretionary (and otherwise authigenic) carbonates are not only present in some of these cores, but that they are relatively common. In addition, most cores that do not contain authigenic carbonates exhibit pore water profile characteristics that are consistent with early diagenetic mineral precipitation in sediments, suggesting that coring may have missed the concretionary body and that authigenesis is, nonetheless, ongoing.

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2. Modern sediment search criteria

Deposits forming under highly productive surface waters where the flux of organic matter to the sediments is high characterize multiple ODP and DSDP sites. As a result of this high export, sediments become anoxic relatively soon after deposition. Such anoxic sediments with high organic carbon contents generally exhibit extensive BSR and methanogenesis as evidenced by decreases and increases in pore water sulfate and methane, respectively. Here, BSR includes the microbially mediated reduction of sulfate by methane. Although this process does not strictly involve bacteria (archaea are thought to play a significant role, (Hinrichs et al., 2000; Orphan et al., 2002)), it would influence pore water profiles similar to BSR (decreased sulfate and increased alkalinity with depth, albeit with a different stoichiometry). Therefore whereas the specific electron donors differ, these two distinct processes would influence pore water chemistry similarly.

Sites off the western margin of North America, South America and Africa and the southern margin of Oman are well known for exhibiting relatively high surface ocean productivity. Here, western margin ODP and DSDP records from the Gulf of California (Leg 64), the Oman margin (Leg 117), the Peru margin (Leg 112), the Cascadia margin (Leg 146), the Santa Barbara Basin (Leg 146), the north African western margin (legs 108 and 157), the central African western margin (Leg 159) and the southern Africa western margin (Leg 175) as well as Blake Ridge (Leg 76) and the Cariaco Basin (legs 15 and 165) (Fig. 1) are targeted to assess 1) the presence of anaerobic organic matter degradation, 2) the presence or absence of authigenic carbonates and 3) indirect evidence for carbonate precipitation. As mentioned above, anaerobic organic matter degradation is identified by pore water profiles of

sulfate and methane (Figs. 2–5). Further evidence for organic matter degradation is provided by ammonia contents of pore waters because organic matter degradation represents a major source of sedimentary ammonia in marine systems (King, 1984; Koike and Hattori, 1978). Organic matter remineralization is also identifiable by carbon isotope compositions ($\delta^{13}\text{C}$) of dissolved inorganic carbon or total CO_2 (TCO_2) that differ from marine values (i.e., $\delta^{13}\text{C}_{\text{TCO}_2} \neq 0\text{‰ VPDB}$) (Claypool and Kaplan, 1974; Irwin et al., 1977). The presence of authigenic carbonate is assessed by core descriptions and includes not only concretionary calcites and dolomites but also disseminated dolomite and calcite crystals, perhaps even as overgrowths precipitated on existing carbonate grains. Indirect evidence for carbonate precipitation is inferred by significant pore water depletions in either calcium or magnesium. Carbonate precipitation potential is indicated by elevated pore water alkalinity and the degree of saturation with respect to calcite and dolomite. The degree of saturation is quantified as the saturation index (SI) defined as $\log \Omega$, where Ω is the saturation state (Berner, 1980), calculated as the respective ion activity product divided by the empirically determined solubility product (e.g., for calcite or dolomite). The SI was calculated via PHREEQC using existing pore water chemical constituents to derive electric charge balances consistently better than 7% and generally to within ~4%. Positive SI values indicate supersaturation. It is important to note that recovered sediment cores, especially those rich in carbonate content, exhibit alkalinity uptake as a result of depressurization and that this decrease likely reflects calcium carbonate precipitation (Murray and Wigley, 1998) as an artifact of core recovery. It is unclear whether or not this core retrieval mineral precipitation would be extensive enough to be recognized during initial core description. For this reason and because pressure influences on mineral saturation are not



Fig. 1. Site map.

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