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微生物（有机）白云石成因模式研究进展与思考

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摘要： 白云石的成因机制一直是地球科学领域备受争论的议题. 传统研究多认为白云石为次生成因. 新近提出的"微生物（有机）白云石模式"认为该矿物在微生物或有机物作用下可以从溶液中直接沉淀，从而为白云石的成因研究提供了新思路. 总结了该模式近 20 年来取得的重要进展，并指出尚待弥补的环节. 微生物的催化机理主要表现为其代谢作用可提高胞外微环境中白云石的饱和度，同时其携带负电荷的细胞外壁可作为白云石晶体的成核位点. 微生物成因白云石呈球状、哑铃状和花椰菜形等外貌. 高盐度有利于微生物介导低温白云石沉淀，而硫酸根扮演的角色则仍需进一步验证. 最新的研究表明微生物胞外聚合物有助于 Mg^{2+} 摆脱水合作用的束缚，是微生物催化白云石形成的关键. 非微生物源的羧基化合物也可通过与胞外聚合物类似的途径在白云石饱和溶液中促进低温白云石形成. 微生物（有机）成因白云石为原白云石，而非有序白云石. 目前对微生物（有机）成因白云石在成岩改造中的演化过程仍然缺乏认识，是"微生物（有机）白云石模式"的缺失环节，亟待完善.

关键词： 白云石问题；微生物催化作用；胞外聚合物；原白云石；成岩作用；生物地球化学

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Advance and Review on Microbial/Organogenic Dolomite Model

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Abstract: The origin of dolomite has remained an enigmatic issue of earth sciences. Dolomite mineral is generally considered as a replacement phase of pre-exsisting carbonate. However, a microbial/organogenic dolomite model has been recently proposed, suggesting that dolomite can also be directly precipitated from solutions with the aid of microorganisms or organic matters, thus providing new insight into the old enigma. This paper reviews the latest progress on the microbial/organogenic dolomite model and discusses how to improve such model. Microbes facilitate low-temperature dolomite formation primarily by enhancing the saturation state with respect to dolomite. Furthermore, the negatively-charged microbial cell walls can be functioned as template for dolomite nucleation. Microbial dolomite normally displays spherical, dumbbell and cauliflower-like morphologies. Microbially-induced dolomite formation can be facilitated by elevated salinity. The role of sulfate on microbial dolomite formation still awaits resolution. The recent studies have also demonstrated that microbial exopolymeric substances (EPS) play an important role in the dewatering of Mg-H_2O complexes. In the similar manner with microbial EPS, natural organic matters can also catalyze dolomite precipitation. However, growing evidence shows that microbes could only precipitate proto-dolomite rather than ordered-dolomite. More studies are required to improve our understanding of the alteration of microbial/organogenic dolomite during burial diagenesis.

Key words: dolomite problem; microbially catalytic effect; exopolymeric substance; proto-dolomite; burial diagenesis; Biogeochemistry

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0　引言

自 1791 年白云石 [$CaMg(CO_3)_2$] 被发现以来，该矿物的成因机理一直备受学者关注. 广义的白云石泛指一类 Mg/Ca 摩尔比接近于 1 的碳酸盐类质同象系列矿物（Warren, 2000）. 狭义白云石（即理想白云石）除含有以上特征外，其晶体结构中 Ca^{2+} 层和 Mg^{2+} 层沿结晶学 c 轴方向间隔 CO_3^{2-} 层交替排列，因此呈现独特的阳离子有序超结构（Lippmann, 1973; Reeder, 1983）. 完全理想的白云石在自然界很少产出. 特别是全新世以来，在大陆架边缘或碳酸盐岩台地沉积序列中发现的白云石普遍呈现富钙、完全无序或部分有序的特征，故常被称作原白云石或钙白云石（Graf and Goldsmith, 1956; Gaines, 1974, 1977; Arvidson and Mackenzie, 1997; Budd, 1997; Petrash *et al.*, 2017）. 由于现代白云石的晶形往往不明显且晶格缺陷密集，处于热力学亚稳定状态，因而被推测在后续埋藏成岩阶段会趋向转变为理想白云石（Petrash *et al.*, 2017）.

地质记录中白云石的分布极度不均衡（图1）. 在古代地层中白云石广泛分布，可构成厚达数百米和覆盖面积数百平方千米的白云岩（Warren, 2000）. 理论上，现代海水对白云石处于过饱和状态（Hardie, 1987），甚至以现代海洋的 Mg/Ca 值（Mg/Ca≈5.2）应当发生大规模的白云石化现象（Hsü, 1980）. 与此相悖的是，全新世以来的地层中极少发现白云石沉积记录. 这种巨大的古今差异无疑是对“将今论古”原理的极大挑战. 此外，大量室内模拟实验试图在地表温压条件下合成白云石，却均以失败告终（Fairbridge, 1957; Land, 1998; Tucker, 2001），这使得白云石的成因机制更加扑朔迷离，成为地球科学领域令人着迷的“白云石问题”.

通过合成实验，人们逐渐认识到白云石沉淀反应是一个动力学控制过程（Warren, 2000）. 这些控制因素主要包括 Mg^{2+} 稳定的水合作用（Zhang *et al.*, 2012a, 2012b）、SO_4^{2-} 对白云石结晶的抑制效应（Baker and Kastner, 1981）、现代海水中低活度状态的 CO_3^{2-} 等（Lippmann, 1973）.

尽管现生白云石沉积非常罕见，一些半深海-深海富有机质沉积物、超盐度潟湖和内陆盐湖的沉积物中陆续发现了少量白云石（Kelts and Mckenzie, 1982; Baker and Burns, 1985; Mitchell *et al.*, 1987; Vasconcelos *et al.*, 1995）. 这些局限地区的现生白云石沉积，极大地激发了研究人员的兴趣，并提出多种模式尝试刻画其成因机制. 据统计，自 20 世纪 60 年代以来，已有 20 余种模式见于诸刊（王茂林等, 2013）. 依据其建立的时间，可将它们分为以次生白云石化为核心思想的传统模式（20 世纪 90 年代之前）和新近的微生物（有机）白云石模式. 本文在回顾各类白云石模式的基础上，重点介绍微生物（有机）白云石模式的发展历程以及近年来获得的一些新认识.

1　传统白云石成因模式及其局限性

尽管现代海水对白云石过饱和，但沉淀的主要碳酸盐矿物是文石. 加之学者们已认识到白云石直接沉淀反应受动力学因素所束缚（Land, 1998; Arvidson and Mackenzie, 1999），因此白云石化作用（dolomitization）一度被认为是白云石形成的唯一途径. 这类次生成因模式认为白云石是富镁流体与富钙碳酸盐矿物发生交代反应的产物，其反应方程可表述为 $2CaCO_3 + Mg^{2+} \rightarrow CaMg(CO_3)_2 + Ca^{2+}$（Hsü, 1980）.

最为人们所熟知的白云石化作用模式有蒸发泵吸、回流渗透和混合水白云石化模式等. 其中，蒸发泵吸模式最早用于解释中东萨勃哈等地区蒸发潮坪中的白云石成因. 该模式认为，海水可为白云石化反应提供充足 Mg^{2+} 来源，强烈的蒸发作用持续浓缩孔隙水，进而显著提高卤水的 Mg/Ca 比，最终卤水与早期文石或方解石长期接触发生交代反应(Hsü, 1980）. 与蒸发泵吸模式不同的是，回流渗透模式常被用于解释碳酸盐台地或沉积盆地中广泛的白云化作用. 这一模式认为高镁盐水的密度较大，因此会向下回流，促使下伏地层中的碳酸盐沉积物或石灰岩发生白云石化（Adams and Rhodes, 1960）. 混合水白云石化模式则认为，当海水以 10%~40% 的比例与淡水混合时，会形成一种对方解石不饱和但对白云石过饱和的流体，从而导致方解石发生白云石化转变. 这种模式较为成功地回答了美国威斯康星中奥陶统白云石的成因（Badiozamani, 1973）. 总体而言，以上次生成因模式均强调了特定地质背景中稳定的 Mg^{2+} 来源以及水文循环等因素的重要性(王茂林等, 2013）. 经过数十年的发展，次生白云石模式被大部分沉积学家所接受，同时也受到了各方质疑. 有研究认为这些模式没有充分考虑海水中基本的化学动力学

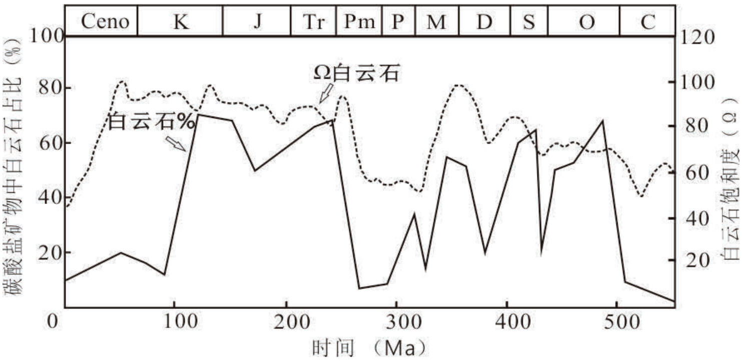

图 1 显生宙白云石丰度和海水饱和度变化曲线

Fig.1 Phanerozoic record of dolomite abundance and dolomite saturation state

修改自 Burns *et al.*(2000), Riding and Liang(2005); 实线代表白云石饱和度, 虚线代表碳酸盐矿物中白云石占比

制约(Oren, 2005), 并指出蒸发泵吸模式中伴随着蒸发浓缩作用, CO_3^{2-} 活度会明显降低, 进而抑制了白云石形成(Lippmann, 1973). 而被认为是回流渗透模式典型实例的加勒比 Pekelmeer 潟湖甚至没有在石膏层下发现回流的卤水(Lucia, 1968). 混合水模式也因为混淆了有序白云石和无序白云石的溶度积、野外实例与理论预测有较大出入而陷入争议(Hardie, 1987). 另一方面, 发育现生白云石沉积的海滨潟湖和萨勃哈潮坪的水体和沉积物中微生物较为繁盛. 电子显微镜也验证了曾被认为是文石或方解石交代成因的白云石实际上是直接从孔隙水中沉淀的产物(Wenk, 1993). 以上发现暗示了还有其他白云石成因模式可以解释现生白云石沉积.

2 “微生物(有机)白云石模式”的提出与发展

微生物白云石模式的正式提出始于 20 世纪90 年代, 为解答“白云石问题”开辟了新思路(李波等, 2010). 该模式的创立归因于以 Judith McKenzie 为代表的学者们多年野外观察和室内模拟研究. 早期, 他们在总结巴西 Lagoa Vermelha 和 Brejo de Espinho 等潟湖的沉积序列和地球化学等数据的基础上, 大胆推测微生物可能是该地区白云石形成的幕后推手. 随后, 室内微生物培养实验直接证明缺氧条件下硫酸盐还原菌确实能够促进低温白云石沉淀(Vasconcelos *et al.*, 1995; Vasconcelos and McKenzie, 1997). 综合已有研究, 硫酸盐还原菌催化低温白云石的形成主要通过以下途径:(1)它们能够氧化小分子有机质生成 CO_3^{2-}/HCO_3^-, 进而提高胞外微环境的碱度和 CO_3^{2-} 活度;(2)该功能微生物还能消耗沉积物孔隙水中的 SO_4^{2-}, 从而破坏 $MgSO_4^0$ 离子对, 提高 Mg^{2+} 的活性;(3)硫酸盐还原菌的细胞壁还能充当白云石晶体的成核模板(Vasconcelos *et al.*, 1995; Vasconcelos and McKenzie, 1997; Castanier *et al.*, 1999; Wright, 1999; Warthmann *et al.*, 2000).

除硫酸盐还原菌外, 深海白云石的形成可能与微生物参与的甲烷厌氧氧化过程密切相关(Moore *et al.*, 2004). 也有报道表明一些产甲烷古菌或菌群也能通过自身的代谢过程显著提高微环境的碱度和白云石饱和度, 即使在 Mg/Ca<1 的溶液中也能促进白云石生成(Roberts *et al.*, 2004; Kenward *et al.*, 2009).

原生白云石的形成并非局限于缺氧环境中, 在 Brejo de Espinho 潟湖表层沉积物等有氧带中同样发现其踪迹(Morse and Casey, 1988). 随后, 室内培养实验显示好氧嗜盐微生物可以促进白云石等碳酸盐矿物的沉淀(Rivadeneyra *et al.*, 2006; Sánchez-Román *et al.*, 2008, 2009, 2011a, 2011b; Deng *et al.*, 2010; Balci and Demirel, 2016; Disi *et al.*, 2017; Qiu *et al.*, 2017; 段勇等, 2017). 据现有报道, 好氧微生物通过分解有机物尤其是含氮化合物(蛋白质等), 生成 NH_4^+ 和 CO_3^{2-}, 显著提高溶液的碱度(Sánchez-Román *et al.*, 2008; 宋泉颖等, 2014). 同时, 呈负电荷的细胞壁高效地吸附阳离子并为白云石提供成核位点, 最终突破低温白云石的化学动力学屏障, 在室温条件下催化白云石形成(Sánchez-Román *et al.*, 2008, 2011b; Qiu *et al.*, 2017).

微生物成因白云石的形貌与高温合成的菱面体白云石截然不同(图2). 生物成因的颗粒大小多为微米或亚微米尺度, 形态可呈刺球、椭球或圆球状. 此外, 这些近球形白云石往往由更小的颗粒聚

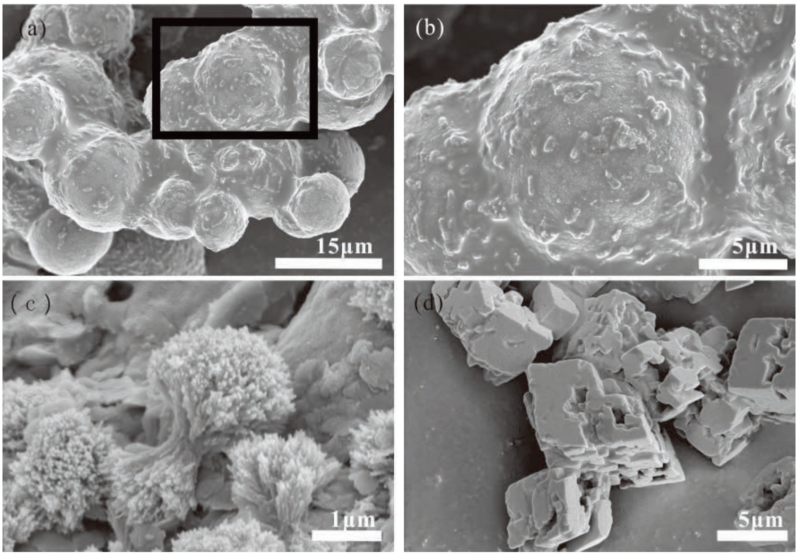

图 2 生物成因与无机成因白云石形貌对比

Fig.2 SEM images of biogenic dolomite and abiotic dolomite

a. 球状生物成因白云石;b. 图 A 的局部放大图(黑框部分),显示钙化微生物菌体;c. 花椰菜形的生物成因白云石;d. 高温合成的菱面体白云石

集形成的集合体，因此其表面在电镜下呈现凹凸不平、粗糙毛刺状的特征(Sánchez-Román *et al*., 2009; 由雪莲等, 2011). 除了亚微米级的球状，一些报道还揭示了微生物成因白云石的典型形态还包括哑铃状和花椰菜状等形态的集合体(Morse and Casey, 1988; van Lith *et al*., 2003; Sánchez-Román *et al*., 2008, 2009; Bontognali *et al*., 2012, 2013)，且这些集合体的内部还具有规则的显微结构，如放射状的纤晶(Sánchez-Román *et al*., 2009)和亚微米级别的空隙(Deng *et al*., 2010).

微生物催化白云石沉淀不仅与其代谢活动息息相关，同时还受控于水化学条件. 溶液盐度被广泛认为能影响白云石沉淀. 高盐度意味着溶液中含有高浓度的 Mg^{2+}、Ca^{2+} 和 CO_3^{2-} 等离子，这对提高离子间的碰撞次数至关重要(Machel and Mountjoy, 1986). 同时,高盐度也能降低 Mg^{2+} 和 Ca^{2+} 去水合化作用所需要的能量(Lippmann, 1973; Gaines, 1980; Machel and Mountjoy, 1986). van Lith *et al*.(2002)等人的沉积学证据证明其他水化学条件相似的两个相邻潟湖(Lagoa Vermelha 和 Brejo do Espinho),盐度较高的 Brejo do Espinho 中微生物沉淀白云石的作用更旺盛. 除以上作用外，最近的研究也揭示高盐度可刺激微生物合成更多的羧基化合物(Qiu *et al*., 2017),而羧基官能团有利于促进阳离子去水合作用进行(Wang *et al*., 2009; Zhang *et al*., 2012b; Roberts *et al*., 2013; Shen *et al*., 2015). 以上发现可与现生白云石多发现于潟湖、萨勃哈潮坪和内陆盐湖等超盐度环境这一事实相互佐证. 与盐度相比,SO_4^{2-} 在白云石形成过程中扮演的角色耐人寻味. Baker and Kastner(1981)通过室内高温无机实验证明白云石化的程度随 SO_4^{2-} 浓度增加而急剧减弱，从而提出硫酸盐抑制白云石沉淀这一学说. 由于该学说影响深远，不少学者相信降低硫酸盐浓度是微生物催化白云石形成过程中必不可少的步骤(van Lith *et al*., 2002)，并由此推论微生物白云石模式只能发生在低 SO_4^{2-} 环境中. 但 Baker and Kastner(1981)的实验操作温度为 200 °C，这个结论可能对低温下微生物催化白云石并不适用(Warren, 2000). 值得一提的是，古代蒸发岩沉积序列中微晶白云石常与石膏和硬石膏共存(Warren, 1991, 1999),表明在高硫酸盐环境中白云石依然可以形成，这与硫酸盐的抑制假说明显不符. 目前,已有研究报道指出不具备消耗硫酸盐能力的好氧嗜盐菌可在高 SO_4^{2-} 溶液中(56 mol/L)诱导白云石沉淀(Sánchez-Román *et al*., 2009),这对硫酸盐抑制假说提出了挑战，但仍需更多的研究加以验证.

微生物菌体可作为白云石的成核位点的观念已逐渐被认可. 近年来的研究还指出微生物胞外附属物 (如胞外聚合物) 在白云石的形成中也至关重要. 除了电镜下常常观察到微生物成因白云石与胞外聚合物伴生外，现代海相沉积物中白云石晶体也多发现于微生物分泌的生物膜中(Krause *et al*.,

2012）. 此外, 在地质记录中也发现了白云石晶粒孔中残留有疑似胞外聚合物矿化碎片（Mastandrea *et al.*, 2006; Perri and Tucker, 2007; You *et al.*, 2013）. 据推测, 微生物的胞外聚合物的催化机理体现在其携带的负电荷基团, 这些以羧基为代表的官能团能够有效地螯合阳离子, 有助于打破 $[Mg(H_2O)_6]^{2+}$ 的水解能量屏障, 是调控低温白云石结晶的关键因子（Roberts *et al.*, 2004; Vasconcelos *et al.*, 2006; Sánchez-Román *et al.*, 2009）.

最新的研究显示非微生物源的羧基化合物也具备催化白云石沉淀的效果, 从而将“微生物白云石模式”进一步拓展为“微生物（有机）白云石模式”. 例如, Zhang *et al.*(2012b) 及合作者向白云石饱和体系中添加羧甲基纤维素, 同样能观察到白云石的生成. Roberts *et al.*(2013) 利用表面羧基化处理的微米球在人工合成的志留纪和现代海水溶液也得到了低温白云石. 值得注意的是,这些有机物催化而得的白云石的形貌与微生物成因白云石非常相似.

3　“微生物（有机）白云石模式”的缺失环节

历经二十余载的研究,“微生物（有机）白云石模式”得到不断修订和补充,俨然成为解决“白云石”问题的一把钥匙. 然而,近年来学者逐渐认识到微生物（有机）成因的白云石缺失阳离子有序度, 更接近现代沉积环境中原生胶结物形式的原白云石, 而非有序白云石（Zhang *et al.*, 2012a, 2012b; Gregg *et al.*, 2015; 张亦凡等, 2015）. 由于在高温合成实验中, 原白云石作为一种重要的中间产物, 会遵循 Ostwald 法则经溶解—再沉淀过程向有序白云石演化（Sibley *et al.*, 1994; Malone *et al.*, 1996）. 故而, 学者推测微生物成因的高镁方解石或原白云石可能是有序白云石的前驱体, 在地层中经历埋藏熟化—稳定的改造, 最终成为沉积序列中的理想白云石（Vasconcelos and McKenzie, 1997）. 这种猜想尽管解释了微生物成因白云石与理想白云石的差异, 却没有提供严密的实验论证, 可以称得上是“微生物（有机）白云石模式”的缺失环节.

沉积学观察显示白云石在成岩过程中会历经递进式的调整作用（Petrash *et al.*, 2017）, 在此过程中微生物成因白云石所携带的“生物信息”的保存状况尚不得而知. 一方面, 这种溶解-重结晶的过程是反复进行的, 只要沉积物保持一定的渗透性且环境中有 Mg 的来源,新的白云石化作用便会发生（Warren, 2000）. 这意味着早期原白云石蕴含的地球化学特征（微量元素和同位素信息）在成岩演化中会被多次重新分配并达到平衡（Land, 1980）. 类似的, 部分研究者认为球状、哑铃状或花椰菜状的外观以及纤维状的内部构造是识别地质体中微生物成因白云石的重要标志. 虽然有研究表明, 将沉积物中部分有序的白云石表面用稀酸处理后可以观察到球形的内核（Vasconcelos and McKenzie, 1997）, 但这种特殊的形貌特征到底在什么条件下能够保存? 在成岩改造过程中如何发生转变? 甚至将这种特殊形态用于鉴别生物成因白云石是否可靠? 目前的研究也为数寥寥, 亟待突破.

4　结语与展望

自 20 世纪 60 年代以来, 多种模式已被陆续提出试图解答“白云石问题”. 其中,“微生物白云石模式”能够成功地解释原生白云石沉积现象. 从前期关注发现具有催化效能的功能微生物, 到近年来深入刻画其催化机制,“微生物白云石模式”已逐渐扩展为“微生物（有机）白云石模式”. 微生物通过其代谢活动可以提高胞外微环境白云石的饱和度. 此外, 由于微生物细胞壁及胞外聚合物富含负电荷基团, 可以有效地螯合镁钙离子, 从而促进镁钙离子摆脱水分子束缚, 进入生长的碳酸盐晶格中, 最终可能导致白云石的生成. 与微生物胞外聚合物类似, 自然界其他非微生物源的有机物可能也在低温白云石沉淀过程中起着不容忽视的作用. 微生物诱导白云石沉淀的行为受水化学条件控制, 其中溶液盐度是重要的一类影响因子. 微生物成因的白云石多呈球状、哑铃状和花椰菜状等形态. 需要指出的是, 越来越多的证据显示微生物或有机成因的白云石为原白云石, 而非有序白云石. 层出不穷的沉积学记录显示古代碳酸盐岩中白云石有序度高, 而全新世以来形成的白云石则多为原白云石或钙白云石, 可能意味着这些无序或低有序度白云石会经埋藏成岩改造, 趋向转变为理想符合化学计量比的有序白云石. 然而, 目前尚缺乏追踪埋藏成岩过程中微生物或有机成因的原白云石演化历程的相关研究, 这也是“微生物（有机）白云石模式”的缺失环节.

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