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页岩中方解石类型、成因及地质意义

陈泓州[1,2],卢志远[1,2],朱光有[1,2],王嘉宁[1,2],郑凯航[1,2],刘健勇[1,2],张杰志[1,2]
(1. 南方复杂页岩油气地质与开发湖北省重点实验室,湖北 武汉 430100;2. 长江大学 地球科学学院,湖北 武汉 430100)

摘要:方解石作为页岩中广泛分布的脆性矿物,在页岩油气赋存、储层改造及沉积环境指示方面具有重要意义。近年来,围绕页岩中方解石的类型、成因机制及地质意义的研究逐渐增多,但系统性综述较少。首先将页岩中的方解石分为泥晶方解石(分散颗粒状、结核状、纹层状、胶结物和生物成因结构)和亮晶方解石(纤维状、刃状和等轴状)两类。研究表明:方解石脉的形成受流体超压、构造应力与结晶力共同驱动,早期形成过程受微生物还原作用影响,中期与有机质脱羧作用相关,晚期与甲烷热氧化流体活动关联,并在热演化过程中出现方解石的溶蚀窗与逆溶蚀现象。陆相与海相页岩中的方解石形成机制总体相似,差异主要源于沉积环境。泥晶方解石的大量富集、页岩与生物介壳层的组合以及方解石包裹体都对恢复古沉积与成岩环境具有一定指示意义。在页岩脆性方面,方解石作为脆性矿物提升了页岩脆性指数,促进了水力压裂裂缝网络的扩展,但其胶结作用又会降低孔隙度,呈现储层改造的"双刃剑"效果。此外,方解石脉体记录了油气运移过程中的信息,其溶蚀孔与层理缝为页岩油气提供了重要储集空间。目前,古老高演化页岩中的方解石成因机制及其与有机质生烃的耦合关系尚不明确,页岩中方解石U-Pb同位素定年技术的精确度还有很大提升空间,未来应提高检测技术精度,进一步揭示页岩中方解石的复杂形成过程,为富方解石页岩油气的甜点预测与高效开发提供更为有力的理论支撑。

关键词:脆性指数;地质意义;成因机制;沉积环境;方解石脉;页岩
中图分类号:TE121.3 **文献标识码:**A

Types, origin, and geological implications of calcites in shales

CHEN Hongzhou[1,2], LU Zhiyuan[1,2], ZHU Guangyou[1,2], WANG Jianing[1,2], Zheng Kaihang[1,2], LIU Jianyong[1,2], ZHANG Jiezhi[1,2]
(1. *Hubei Key Laboratory of Complex Shale Oil and Gas Geology and Development in Southern China*, *Wuhan*, *Hubei* 430100, *China*;
2. *School of Geosciences*, *Yangtze University*, *Wuhan*, *Hubei* 430100, *China*)

Abstract: Calcites, brittle minerals that are extensively distributed in shales, hold great significance for indicating shale oil and gas occurrence, reservoir stimulation, and sedimentary environments. In recent years, increasing studies have been conducted on the types, genetic mechanisms, and geological significance of calcites in shales. However, systematic reviews on these topics remain limited. In this study, calcites in shales are categorized into two types initially: micritic calcites (including dispersed granular, nodular, and lamellar calcites, calcite cements, and biogenic calcites) and sparry calcites (including fibrous, bladed, and equiaxed calcites). The subsequent results indicate that the formation of calcite veins is jointly driven by fluid overpressure, tectonic stress, and crystallization forces. The formation process is affected by microbial reduction in the early stage, relates to the decarboxylation of organic matter in the middle stage, and is associated with the methane thermochemical oxidation-related fluid activity in the late stage. During the thermal evolution of calcite veins, dissolution windows and retrograde dissolution also occur. Calcites in lacustrine and marine shales generally share similar formation mechanisms, with differences attributed primarily to sedimentary environments. The high degree of enrichment of micritic calcites, the combination of shales and bioclastic layers, and calcite inclusions all provide valuable indications for the reconstruction of paleosedimentary and diagenetic environments. Regarding shale brittleness, calcites both enhance the shales'

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第一作者简介:陈泓州(2001—),男,硕士研究生,地质学。**E-mail:**geochenhongzhou@163.com。
通信作者简介:朱光有(1973—),男,二级教授,深层油气成藏与油气地球化学。**E-mail:**zhuguangyou@yangtzeu.edu.cn。
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brittleness index and promote the propagation of hydraulically stimulated fracture networks as brittle minerals. However, their cementation reduces reservoir porosity, creating dual effects on reservoir stimulation. Additionally, calcite veins preserve records of the hydrocarbon migration process, while their dissolution pores and bedding-parallel fractures offer important reservoir spaces for shale oil and gas. Nevertheless, the genetic mechanisms of calcites in ancient, highly evolved shales and their coupling relationship with the hydrocarbon generation process of organic matter remain poorly understood. Furthermore, the accuracy of U-Pb dating techniques for calcites in shales is yet to be substantially improved, and further improvements in the precision of relevant detection techniques are required to further reveal the complex formation process of the calcites. Advancements in these areas will provide more improved theories for the sweet spot prediction and efficient exploitation of oil and gas from calcite-rich shales.

Key words: brittleness index, geological significance, genetic mechanism, sedimentary environment, calcite vein, shale

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页岩不仅是一种优质烃源岩,同时也是页岩油气的储层,具备自生自储的特点[1-3]。页岩中包含多种矿物,其中矿物类型以及矿物的成岩演化过程控制着页岩的储层特性,主要体现为对页岩孔隙类型和发育程度的控制,以及对页岩力学性质和储层吸附能力的影响[4-6]。方解石、石英、黏土矿物和黄铁矿是页岩的主要矿物成分[7-10],其中方解石在页岩中部分呈泥晶状随机分布,更多则是呈脉状充填于页岩裂缝中,或者以生物成因的介壳形态存在[11]。常用的分析技术手段包括漫反射红外傅里叶变换光谱(DRIFTS)和X射线衍射(XRD),用于研究页岩中的矿物组成,XRD分析可以提供页岩中方解石结晶含量的定量数据[12]。

自19世纪末以来,纤维状方解石脉(Beef)与叠锥(cone-in-cone)构造等特殊构造因蕴含流体活动、成岩环境、古应力特征及岩石形变等关键信息而备受重视,地质学家因此对富有机质细粒沉积物内的Beef结构与叠锥构造展开了深入研究[13-18]。页岩方解石脉的物源通常为泥晶方解石和生物来源,而流体超压、挤压构造应力和结晶力等是影响其形成的主要因素[19-21]。在各沉积盆地中方解石极为常见,虽然前人研究发现页岩中方解石不直接参与油气的生成[22-24],但方解石具备丰富的烃、水、岩相互作用信息,又对页岩储层特性和岩石力学性质有着不可忽视的影响,其发育过程和形成机制对油气的示踪和成藏研究具有重要意义[25-26]。陆相页岩中方解石的高富集现象在渤海湾盆地表现显著,尤其以东营凹陷为代表,页岩方解石脉丰富[27-29],其沙河街组页岩裂缝型方解石脉发育,研究程度高,方解石脉对储层的储集空间具有一定的抑制作用[30-32]。海相页岩中方解石的富集现象常见于四川盆地志留系龙马溪组,方解石脉中包含重要的流-岩作用信息[33-34]。综上可见方解石类型、成因及其地质意义的研究对富方解石地区的页岩油气开采具有重要的指导意义[35-36]。

近年来,已有研究综合阐述了页岩中的黄铁矿和石英等矿物类型、特征、成因及意义,以及页岩自生方解石在示踪沉积盆地埋藏过程与成岩流体演化方面的作用[37-42],然而,有关方解石的类型、形成时间、物源以及其与页岩沉积环境、力学性质和储集性之间关系的综合研究未见报道,本文对不同地区页岩中方解石的类型、形成时间和物源进行总结,并归纳对比陆相与海相页岩中方解石的差异,从页岩中方解石对沉积环境的指示作用、对页岩力学性质和储集性能的影响3个方面探究页岩中方解石的地质意义。

1　页岩中方解石的形态特征及类型

1.1　方解石晶体的形态特征

方解石属于三方晶系,空间群(国际符号)为$R\overline{3}c$[43]。方解石常见菱面体晶形(图1a),具六方晶胞结构(图1b),它有3组完全的解理菱面体,活度积常数在25 ℃时为$10^{-8.52}$,莫氏硬度为3,相对密度为2.71[44]。对从四川盆地二叠系吴家坪组取得的页岩样品处理后置于扫描电镜下观察,可看到页岩中方解石呈现规则菱形,棱角尖锐,为自形菱面体(图2a),同时存在团簇状微晶集合体(图2a,b),与伊/蒙混层矿物共生(图2b)。

1.2　页岩中方解石的类型

本文按照晶体粒度大小将页岩中主要的方解石晶

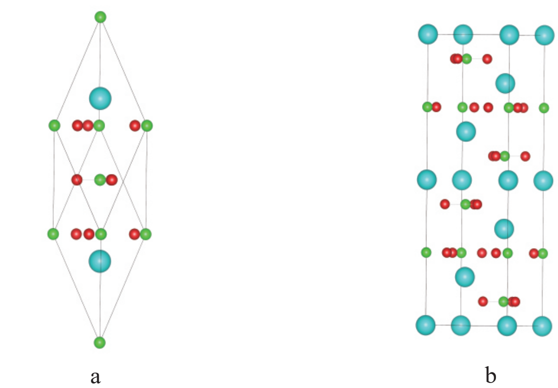

图1 方解石的单胞晶体结构

Fig. 1 Scheme showing the unit-cell crystal structures of calcites

a. 菱面体结构;b. 六方晶胞结构

(蓝色为Ca;绿色为C;红色为O。)

体划分为泥晶方解石与亮晶方解石,泥晶按产出形态分为分散颗粒状、结核状、纹层状、胶结物和生物成因结构,亮晶按长/短轴长度比分为纤维状、刃状和等轴状(表1)。

1.2.1 泥晶方解石

泥晶方解石是一种隐晶至微晶级别的碳酸钙($CaCO_3$)颗粒,粒径通常小于4 μm[52]。泥晶方解石可以在多种沉积环境中形成,在湖泊中常受微生物影响而沉淀,其重结晶是页岩中亮晶方解石脉的重要形成过程[53-54]。泥晶方解石既可以是自生的,即在原地通过物理化学、微生物或生物化学过程形成;也可以是他生的,即通过外源碳酸盐颗粒的破碎作用或沉积作用形成。自生泥晶方解石常见于海底或沉积物表面[55],他可以通过微生物活动、光合作用或其他生物化学过程生成,如在川东北下侏罗统大安寨段发生的文石转化作用形成了大量的泥晶方解石[56]。自生泥晶方解

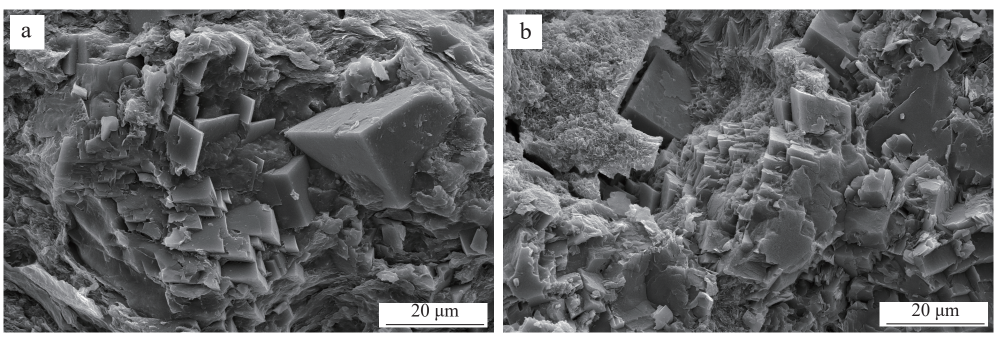

图2 四川盆地二叠系吴家坪组页岩中方解石扫描电镜照片

Fig. 2 Scanning electron microscopy (SEM) images showing the characteristics of calcites in shales from the Permian Wujiaping Formation, Sichuan Basin

a. 菱形方解石,同时可见方解石团簇状微晶集合体,LX1侧眼井,埋深7 040.40~7 040.55 m;b. 方解石团簇状微晶集合体与伊/蒙混层,GS133井,埋深3 909.47~3 909.50 m

表1 页岩中方解石类型

Table 1 Types of calcites in shales

方解石类型	特点	页岩储层意义	典型研究案例(文献)
分散颗粒状泥晶方解石	分散状	反映低能静水环境	[11]
结核状泥晶方解石	呈不规则球状或椭球状	结构致密,原生孔隙少,通常对储集空间贡献有限	[45]
纹层状泥晶方解石	具平行纹层构造,单层厚度毫米-厘米级	烃源岩发育层段 (高*TOC*)	[46]
泥晶方解石胶结物	充填于颗粒间孔隙或裂缝中	破坏原生孔隙 (胶结封堵)	[47]
生物成因(生物介壳)	保留生物结构	生物体腔孔或溶蚀孔可提供储集空间 (改善储集空间)	[48]
纤维状亮晶方解石	长:宽>6.0:1.0	改善脆性 (微裂隙网络)	[49]
刃状亮晶方解石	1.5:1.0<长:宽<6.0:1.0	流体运移通道 (增孔增渗)	[50]
等轴亮晶方解石	长:宽<1.5:1.0	破坏原生孔隙 (胶结封堵)	[51]

注:*TOC*为总有机碳含量。

石通常显示出特定的显微结构，例如光滑的菱面体、哑铃状和晶球状等形态[57]，其荧光和阴极发光显微特征具有很好的对应性，这表明自生泥晶方解石可以通过有机矿化作用形成[58]。他生泥晶方解石则是通过已有的碳酸盐颗粒破碎或异地搬运沉积形成的。这类泥晶方解石没有特定的形成环境限制，可以来自各种不同的地质背景，并且可能包含不同的矿物和化学成分[59]。按照常见产状又可分为分散颗粒状、结核状、纹层状、胶结物和生物成因结构泥晶方解石。分散颗粒状泥晶方解石指微米级泥晶方解石颗粒零星分布于黏土基质中，可能由机械沉积或生物碎屑分解形成[60]；结核状泥晶方解石以不规则斑块或结核形式分散在页岩中，通常与局部成岩作用或生物活动有关[61]；当页岩中泥晶方解石富集到一定程度时可呈现纹层状，在页岩中很常见，可见泥晶方解石层与富有机质黏土层在页岩中的互层现象（图 3a）[62]；泥晶方解石也会以胶结物的形式在孔隙或裂缝中充填，是成岩期化学沉淀的产物[63]；生物成因结构主要是生物介壳（如有孔虫和钙质藻类）经压实作用后形成的泥晶方解石，例如四川盆地大安寨段就发育对岩层脆性有影响的大量钙质介壳层[64]。

1.2.2　纤维状、刃状和等轴状亮晶方解石

通常来说，晶粒粒径大于 10 μm 的方解石晶体被称为亮晶方解石[65]。亮晶方解石广泛分布于全球多个地区的页岩中，尤其在富有机质的深埋页岩中更为常见，常呈脉状发育于裂缝中[66-68]。

纤维状亮晶方解石晶体的轴比为长∶宽 > 6.0∶1.0，以脉状充填于页岩裂缝中，与石灰岩相比其在页岩中更为常见[25,69-71]。Beef结构中存在一条“中线”，由棕色粒状方解石、分散的寄主岩石碎片以及黄铁矿树状体等为界定（图 3b），几个相邻的纤维状亮晶方解石可结合并生长在一起，形成叠锥构造（图 3c）[72]。Beef结构可以作为油气成熟过程中流体超压的构造证据[73]。

刃状亮晶方解石晶体的轴比为 1.5∶1.0 < 长∶宽 < 6.0∶1.0，刃状亮晶方解石脉末端如刀刃（图 3d），在生长过程中与纤维状方解石晶体沿生长方向平行排列或在生长过程中相互竞争（图 3e）。等轴状亮晶方解石晶体的轴比为长∶宽 < 1.5∶1.0，呈现近等轴粒状（图 3f）[74-76]。

2　页岩中方解石的形成时间与物质来源

2.1　形成时间

泥晶方解石，特别是自生泥晶方解石，普遍是同生–早成岩阶段的产物，保留了原始沉积组构（如生物碎屑轮廓），同样是在早成岩阶段形成的还有碳酸盐结核，研究发现碳酸盐结核的形成过程与沉积物甲烷转换带内沉积间歇期间的厌氧甲烷氧化过程相关[79-80]。

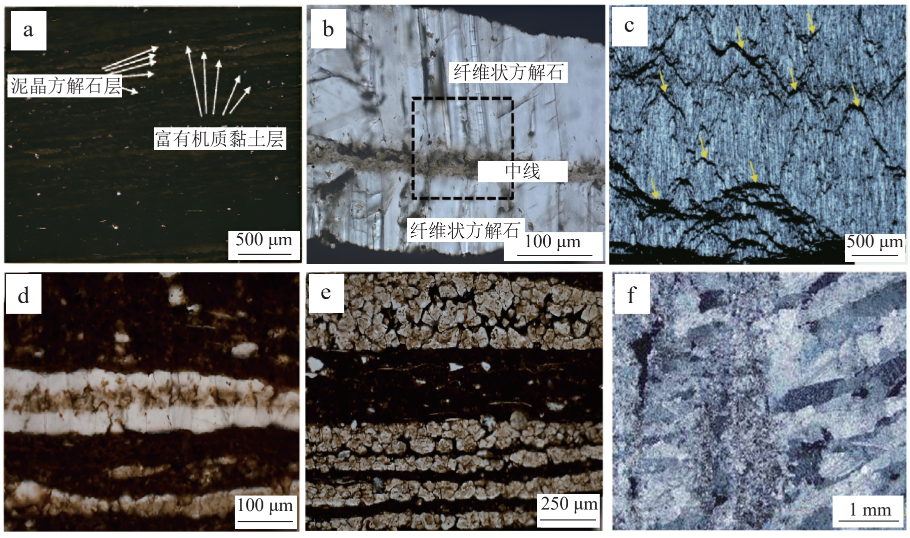

图 3　页岩中不同类型方解石薄片显微特征照片

Fig. 3　Microscopic characteristics of thin sections of various types of calcites in shales

a. 断续纹层，泥晶方解石透镜状断续成层，济阳坳陷沾化凹陷，罗 A 井，沙河街组三段下亚段，单偏光[77]；b. 纤维状方解石脉，中线呈棕色，两侧有清晰的纤维状方解石，渤海湾盆地东营凹陷，N38 井，埋深 3 358.00 m，单偏光[72]；c. 纤维状方解石发育叠锥构造，可见弯曲状包裹体带（黄色箭头所示）及多重嵌套的纤维锥体，松辽盆地，X2HC 井，埋深 2 359.32 m，单偏光[78]；d. 刃状亮晶方解石，单偏光[76]；e. 等轴状亮晶方解石，单偏光[76]；f. 刃状和纤维状亮晶方解石晶体垂直于脉壁竞争排列，阿联酋阿布扎比地区油田，正交偏光[50]

亮晶方解石则大多是中-晚成岩阶段的产物，其形成受到压实/压溶作用、地质构造作用、结晶力、油气生成过程中的流体超压以及有机酸的溶蚀等作用的显著影响（表2）[81]。在中-晚成岩阶段，由于受热液蚀变等因素影响保留下来的泥晶方解石可发生重结晶，形成等轴状亮晶方解石[82]。从早成岩阶段到晚成岩阶段，方解石的化学组分可能产生变化，在构造应力驱动下，富集Fe^{2+}的成岩流体沿岩石裂隙网络进行定向迁移，当此类还原性流体与方解石发生界面反应时，通过Fe^{2+}—Ca^{2+}置换作用触发次生铁质方解石的结晶沉淀。该过程通常伴随着矿物相转化，原始方解石晶格中的Ca^{2+}被Fe^{2+}选择性取代，形成从早期无铁方解石到晚期铁方解石的转变[56,83]。

2.2 物质来源

页岩中他生方解石物源主要为沉积盆地外的碎屑输入，占比相对较少。本文主要讨论自生方解石的物质来源，其Ca^{2+}主要来源于早期钙质物质的溶解、孔隙水、热液以及黏土矿物的转化；CO_3^{2-}来源于有机来源CO_2、无机来源CO_2和原生碳酸盐岩；Fe^{2+}与黏土矿物的运移关系密切（表3）。

2.2.1 Ca^{2+}来源

页岩中自生方解石的Ca^{2+}首先可能来源于早期钙质物质的溶解，如泥晶方解石、白云石、石膏、硬石膏和生物介壳等[88]。在浅埋藏阶段，孔隙水与沉积水体之间的良好连通性促使两者之间发生物质交换，其中，孔隙水中的Ca^{2+}来源于陆源碳酸盐岩的风化产物，因此孔隙水中可能富含Ca^{2+}，为后续方解石的重结晶提供了重要物质来源，高Mg/Ca和Sr/Ca元素含量比值可作为其识别标志[89]。含钙岩浆矿物易溶于热液流体中，因而热液也是方解石中Ca^{2+}的重要来源，同时热液成因的方解石常具有轻稀土元素富集的特征[90-92]。黏土矿物的转化也是一个重要的方解石Ca^{2+}来源——随

表2 页岩中方解石形成时间对比

Table 2 Comparison of the formation time of calcites in shales

成岩阶段	早期成岩阶段（泥晶方解石为主）	中成岩阶段（亮晶方解石为主）	晚成岩阶段（亮晶方解石为主）
形成时间	沉积后数千年内（浅埋藏阶段）	生烃高峰期（有机质成熟阶段）	生烃超压阶段（构造活动期或强生烃增压期）
地质背景	微生物活跃的还原环境	有机酸溶蚀再沉淀作用主导	超压裂缝系统或构造裂隙
岩石学特征	纹层状或结核状	脉状或斑块状充填	纤维状或巨晶簇状
	与有机质纹层互层	切割早期纹层	沿裂隙中心对称分布
储集空间类型	晶间微孔	溶蚀孔	裂隙扩容缝
	层间缝	脉体边缘微裂缝	晶簇间隙孔
地质意义	支撑"源-储一体"岩相	改善页岩渗透率	超压释放通道，深部流体活动指示
	微生物成岩作用标志	生烃增压溶蚀耦合过程的产物	无铁方解石转变为铁方解石
典型研究案例（文献）	［84-85］	［75，82］	［86-87］

表3 页岩中方解石的物质来源

Table 3 Material sources of calcites in shales

离子类型	来源分类	具体来源与机制	识别标志	典型研究案例（文献）
Ca^{2+}	早期钙质溶解物质	泥晶方解石、白云石、石膏、硬石膏和生物介壳等早期钙质物质的溶解	残留的早期钙质物质	［88］
	孔隙水	浅埋藏阶段孔隙水与沉积水体交换物质，陆源碳酸盐岩风化产物进入孔隙水	高Mg/Ca和Sr/Ca元素含量比值	［89］
	热液	含钙岩浆矿物易溶于热液流体	轻稀土元素富集	［90-92］
	黏土矿物转化	蒙脱石伊利石化过程中释放Ca^{2+}：① 一般环境1.0 mol蒙脱石释放2.0 molCa^{2+}；② 富Mg^{2+}环境1.0 mol蒙脱石释放0.1 molCa^{2+}	蒙脱石和伊利石的黏土矿物共生组合	［76］
CO_3^{2-}	有机来源	有机质成岩与热解烃类氧化生成的CO_2	$\delta^{13}C$显著偏负（可达-25‰）	［79，93］
	无机来源	地幔脱气、中下地壳与上地幔楔熔融脱气及变质作用中碳酸盐岩热分解	$\delta^{13}C$通常为2.1‰～4.8‰	［94-95］
	原生碳酸盐岩	早期成岩方解石的溶解再沉淀	$\delta^{13}C$继承早期方解石特征（≈0±3‰）	［96］
Fe^{2+}	黏土矿物携带	①作为黏土矿物本身成分运载；②氧化铁膜吸附固定在黏土矿物表面	黏土矿物化学组分	［97-98］

着埋深和地温增加，蒙脱石发生伊利石化，在此过程中每1.0 mol蒙脱石会释放2.0 mol的Ca^{2+}，但在富Mg^{2+}环境中转化路径有所改变，1.0 mol蒙脱石仅释放0.1 mol的Ca^{2+}，依据是否存在蒙脱石和伊利石的黏土矿物共生组合可识别此物源路径[76]。

2.2.2 CO_3^{2-}来源

碳酸盐来源主要包括3种途径。①有机来源CO_2：有机质在成岩、热解及烃类氧化过程中生成的CO_2，其稳定碳同位素比值($\delta^{13}C$)显著偏负(可达-25‰)[79,93]；②无机来源CO_2：源自地幔脱气、中-下地壳与上地幔楔熔融脱气以及变质过程中碳酸盐岩热分解的CO_2其$\delta^{13}C$通常为2.1‰～4.8‰，与有机来源相比偏正[94-95]；③原生碳酸盐岩：参与方解石溶解-再沉淀的碳酸盐，其形成受有机酸/碳酸控制，碳原子源于早期成岩作用形成的方解石，其$\delta^{13}C$继承早期方解石特征(≈0±3‰)[96]。总体以有机来源CO_2与原生碳酸盐岩为主要CO_3^{2-}物源。

2.2.3 Fe^{2+}来源

Carroll提出黏土矿物组分是Fe元素迁移至沉积体系的关键媒介，其携带Fe元素的途径主要有两种：①将Fe元素作为黏土矿物本身成分运载；②以氧化铁膜形式吸附固定在黏土矿物表面[97-98]。通过黏土组分的携带，方解石可以从环境中获取到Fe^{2+}，完成无铁方解石到铁方解石的转变。

2.2.4 成岩演化过程中的碳、氧同位素变化

方解石形成过程中其稳定碳、氧同位素比值($\delta^{13}C$与$\delta^{18}O$)总体呈现出从早成岩阶段到晚成岩阶段逐渐偏负的趋势。①早成岩阶段受微生物活动(如细菌硫酸盐还原和厌氧甲烷氧化)影响，碳源以无机碳和早期有机质分解为主，无机碳相对富集^{13}C，总体$\delta^{13}C$值偏正(2.1‰～4.8‰)，$\delta^{18}O$偏负(-11.25‰～-7.13‰)；②中成岩阶段受有机质热演化影响，碳源以有机质分解产生的CO_2为主，$\delta^{13}C$逐渐偏负(-7.01‰～-0.83‰)，$\delta^{18}O$进一步降低(-15.69‰～-10.23‰)；③晚成岩阶段受有机质高成熟演化(如原油裂解和甲烷生成)影响，碳源以高温裂解生成的甲烷及相关氧化产物为主，$\delta^{13}C$进一步偏负(-17.21‰～-13.49‰)，$\delta^{18}O$持续偏负(-9.55‰～-5.26‰)[50,95,99-100]。海相页岩中的方解石脉锶同位素含量比值($^{87}Sr/^{86}Sr$)从早成岩到晚成岩阶段整体保持高值特征(0.719 366～0.721 342)[100]。

3 页岩中方解石的形成过程

3.1 方解石脉形成过程的三要素与模型

流体超压、构造应力和结晶力被普遍认为是方解石脉形成的主要影响因素，三者中流体超压是方解石脉最重要的形成基础；构造应力则在活动断裂带或褶皱翼部影响显著；结晶力则是方解石脉拓展过程中的主要控制因素[101-103]。

流体超压为方解石脉的形成提供了空间基础。在静岩压力环境中，垂向主应力(S_v)一般大于最小水平主应力(S_h)[104]。但当发育平行层理的裂缝时，垂向主应力需被抑制至低于最小水平主应力($S_v < S_h$)[105]。为实现这一应力反转，需产生地层超压抵消上覆岩层重力(即S_v的垂向载荷)，并确保超压至少超过上覆岩层压力与岩石抗拉强度的叠加量。此过程通过超压弱化垂向应力主导状态，为水平应力主导的裂缝扩展提供必要条件[106]。在川中侏罗系大安寨段的富有机质页岩中层状方解石脉及介壳灰岩夹层广泛发育，它们均是流体超压作用的结果[87]。有机质生烃过程产生的超压以及该过程产生的副产品有机酸共同对介壳的演化进程以及方解石的沉淀过程施加影响，历经了十分强烈的有机酸溶解作用和方解石再沉淀作用后，最终使得方解石与沥青共存，除沥青外共生矿物还有高岭石和石英等[107-109]。除流体超压因素外，Zanella等[110]推测挤压构造应力促使岩层压裂，进而对方解石脉的形成起到了辅助作用。如在松辽盆地古龙凹陷区广泛发育层间方解石脉体，其中层状方解石脉主要由垂直于层理的纤维状方解石晶体构成[111]。这些方解石脉体普遍存在共轭破裂系统和构造挤压变形，记录了古构造应力场反转过程，为古应力重建提供了关键地质证据。通过构造应力反演分析发现三向应力差异显著，有效解释了广泛发育的层理裂隙系统及沿层理面生长的纤维状方解石脉的形成机制[112-113]。结晶力既引起方解石脉体拓展，也可能控制着纤维状方解石的优先生长，因此同样是方解石脉形成不可忽视的一个重要因素[21]。方解石结晶力源于过饱和溶液内的晶体析出过程，从而引发方解石在周边介质中的结晶生长[103]，在纤维状亮晶方解石脉内部普遍存在的围岩包裹体U型或V型排列结构就被认为是由结晶力持续驱动矿物生长所致[114]。

典型纤维状亮晶方解石脉的形成模型可分为3期动态过程：①首先，泥晶方解石纹层内发育初始孤立方

解石晶核(图4a);②随后进入增生扩展期,晶体群密度显著提升,粒径同步增大导致相邻晶面发生横向连生现象,形成网状晶体结构(图4b);③最终阶段呈现异向性生长模式,晶体的水平扩展作用终止,转而通过沿轴向持续延伸形成定向排列的纤维状方解石集合体(图4c)。该序列化结晶过程揭示了矿物生长方向与空间约束条件的动态耦合关系[115]。按方解石脉的生长方向可分为同轴脉和反轴脉(同轴脉由单一界面向另一界面生长,反轴脉则从中线向两侧围岩壁面扩展)[116],页岩裂隙扩展速率与脉状结晶状态呈现显著相关性:在开放空间环境中,刃状亮晶方解石更易发育(图5a,a_1);反之,在受限空间条件下则会促进纤维状亮晶方解石的形成(图5b,b_1,c,c_1)[78]。

3.2 陆相页岩与海相页岩中方解石的对比

3.2.1 陆相页岩的中方解石

陆相页岩中的碳酸盐矿物主要以纹层状-结核状方解石和泥晶团块为主,具有显纹层结构(毫米级韵律层),可能存在结核状类型(如干旱湖盆的钙结层)[117-118]。陆相页岩所在湖盆体系较为封闭,水体滞留(蒸发量 > 补给量),盐度波动大(淡水—超盐),干-湿季气候驱动特征明显,相较于全球气候变化,雨季淡水输入的稀释作用和旱季蒸发浓缩作用等区域气候控制因素占绝对主导地位,方解石纹层反映了季节性化学沉淀过程[119-120]。例如四川盆地下侏罗统大安寨段中碳酸盐矿物多为泥晶方解石胶结;而在渤海湾盆地东营凹陷古近系沙河街组四段上亚段(沙四上亚段)页岩中则发育粒状方解石纹层+混合纹层+有机质纹层组合[121-122]。

3.2.2 海相页岩中的方解石

海相页岩中方解石主要以分散状微晶方解石和生物碎屑方解石为主,颗粒细小、均匀分散于黏土基质中,其主要是生物成因(如有孔虫和颗石藻碎片)或化学沉淀作用形成的。方解石含量较高,含少量的镁,同时海相深水页岩也可能存在薄层状方解石条带和灰岩夹层等[45,123]。海相页岩沉积环境偏开放水体,水体循环相对通畅,盐度稳定(约为35‰),存在富有机质缺氧底水,受海平面变化等全球气候波动控制,由于深水压力稳定,早期成岩作用相对较弱[124-125]。以五峰组-龙马溪组海相页岩为例,除了观音桥介壳灰岩,大多数页岩是由分散状微晶方解石和生物碎屑方解石组成,且方解石含量较低,整体为黑色富有机质页岩。在龙马溪组一段(龙一段)中部(早期高位体系域)受广西运动影响,粗碎屑含量增加;龙一段顶部(晚期高位体系域)受化学风化及物质输入的影响,碳酸盐矿物含量显著增加,沉积灰质泥岩或泥质灰岩[126-127]。

3.2.3 陆相与海相页岩中方解石脉形成过程的对比

对比陆相与海相页岩方解石脉形成过程发现其具有极高的相似性(图6a,b)。以泌阳凹陷陆相页岩中方解石脉的形成过程为例,可分为3个阶段:①生物成因气的生成引发超压现象,这种超压导致水平水力压裂的发生,从而形成裂缝,这些裂缝作为气体排出通道;②气体释放完成后,张开的裂缝会因上覆载荷而收缩,但不会完全闭合;③随后宿主岩石中方解石组分通过压溶作用提供的物质会扩散到未愈合的水平裂缝中。在结晶力的作用下,方解石反轴生长,形成Beef结构,同时裂缝扩张(图6a)[128-130]。海相页岩中

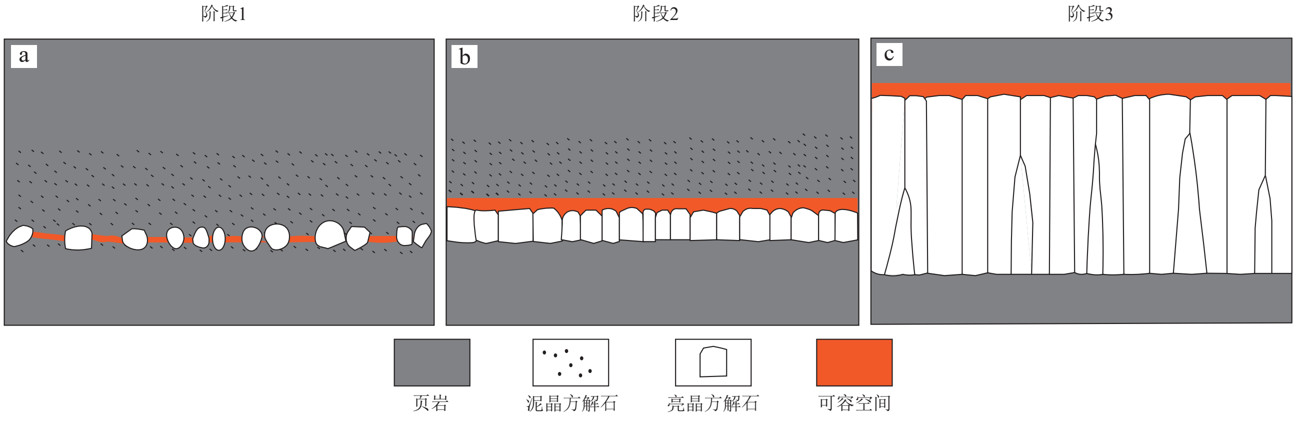

图4 纤维状亮晶方解石脉演化过程示意图(据文献[115]修改)

Fig. 4 Evolution of fibrous sparry calcite veins (modified after reference [115])

a. 形成初期;b. 扩展时期;c. 纤维状方解石脉形成

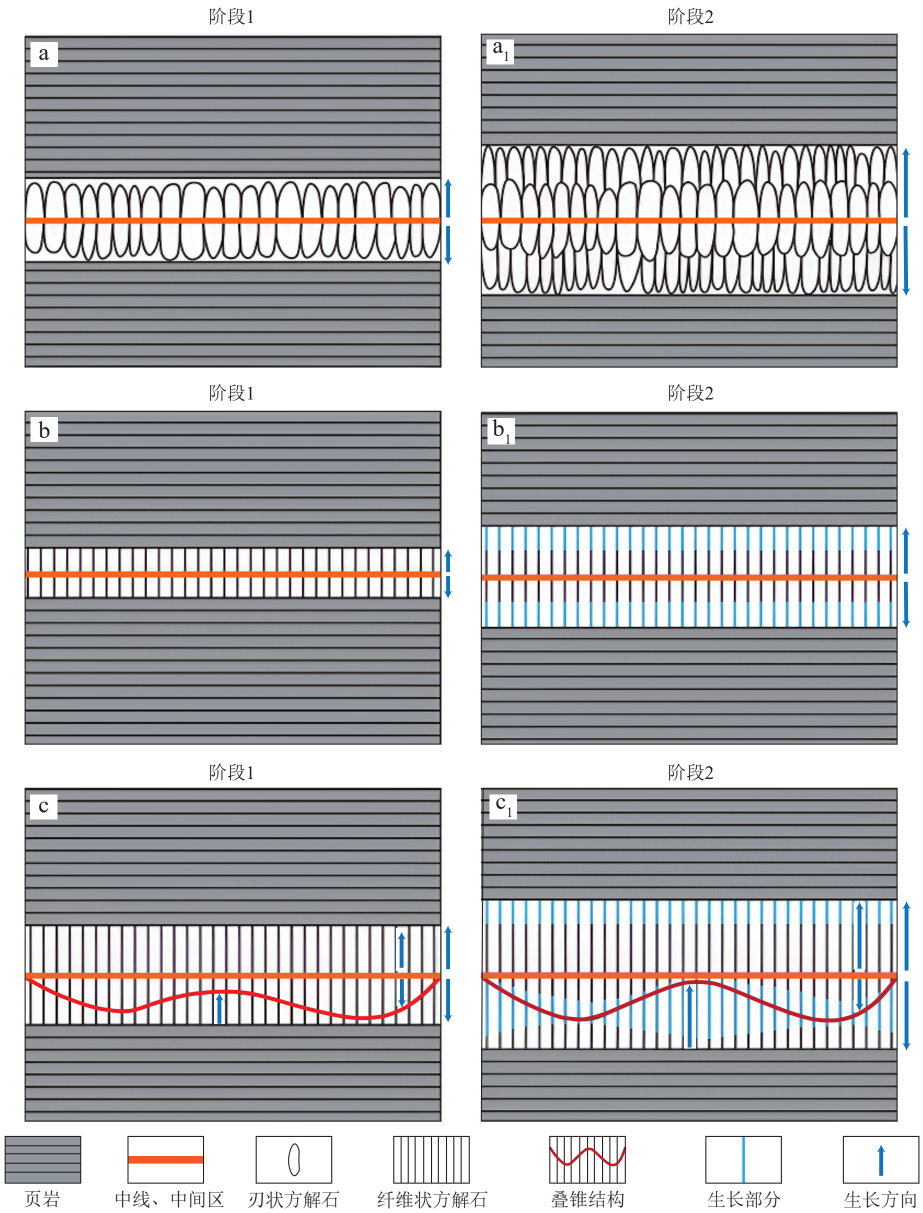

图5　页岩方解石脉的反轴生长模型(据文献[78]修改)

Fig. 5　Growth models of anti-axial calcite veins in shales (modified after reference [78])

a,a_1. 分别为反轴刃状方解石脉早期和晚期阶段;b,b_1. 分别为反轴纤维状方解石脉早期和晚期阶段;c,c_1. 分别为反轴方解石脉早期和结晶扩展晚期叠锥结构阶段

方解石脉的形成以四川盆地志留系龙马溪组为例,也可分为3个阶段:①有机质尚未成熟时,由微生物活动占主导地位,由其引发的超压效应促使裂缝形成;②随着热演化程度加深,有机酸大量生成,方解石受烃源流体改造;③达到过成熟状态后,方解石脉受到甲烷改造影响。在海相第二和第三阶段中结晶力持续促进方解石脉的扩展(图6b)[100,131-132]。对比二者发现流体超压、构造应力和结晶力都是其形成过程中的主要影响因素,展现出高相似性,而二者的差异主要归因于陆相与海相沉积环境的不同,见前两小节。

3.3　方解石脉形成过程中的地球化学差异与逆溶蚀现象

根据地球化学特征的差异,方解石脉的形成可分为三期[134]。①第一期形成于同生-准同生成岩阶段,方解石单独产出或沿围岩界面生长,溶蚀现象较弱,富Fe,Mg和Al而贫Mn,碳同位素(^{13}C)富集而氧同位素(^{18}O)亏损,其形成受微生物(如甲烷菌)还原作用影响;②第二期形成于早成岩阶段,方解石溶蚀改造现象明显,相对富Fe,Mg,Mn和Al元素,^{13}C和^{18}O同位素均亏损,受有机质脱羧作用改造;③第三期形成于晚成岩

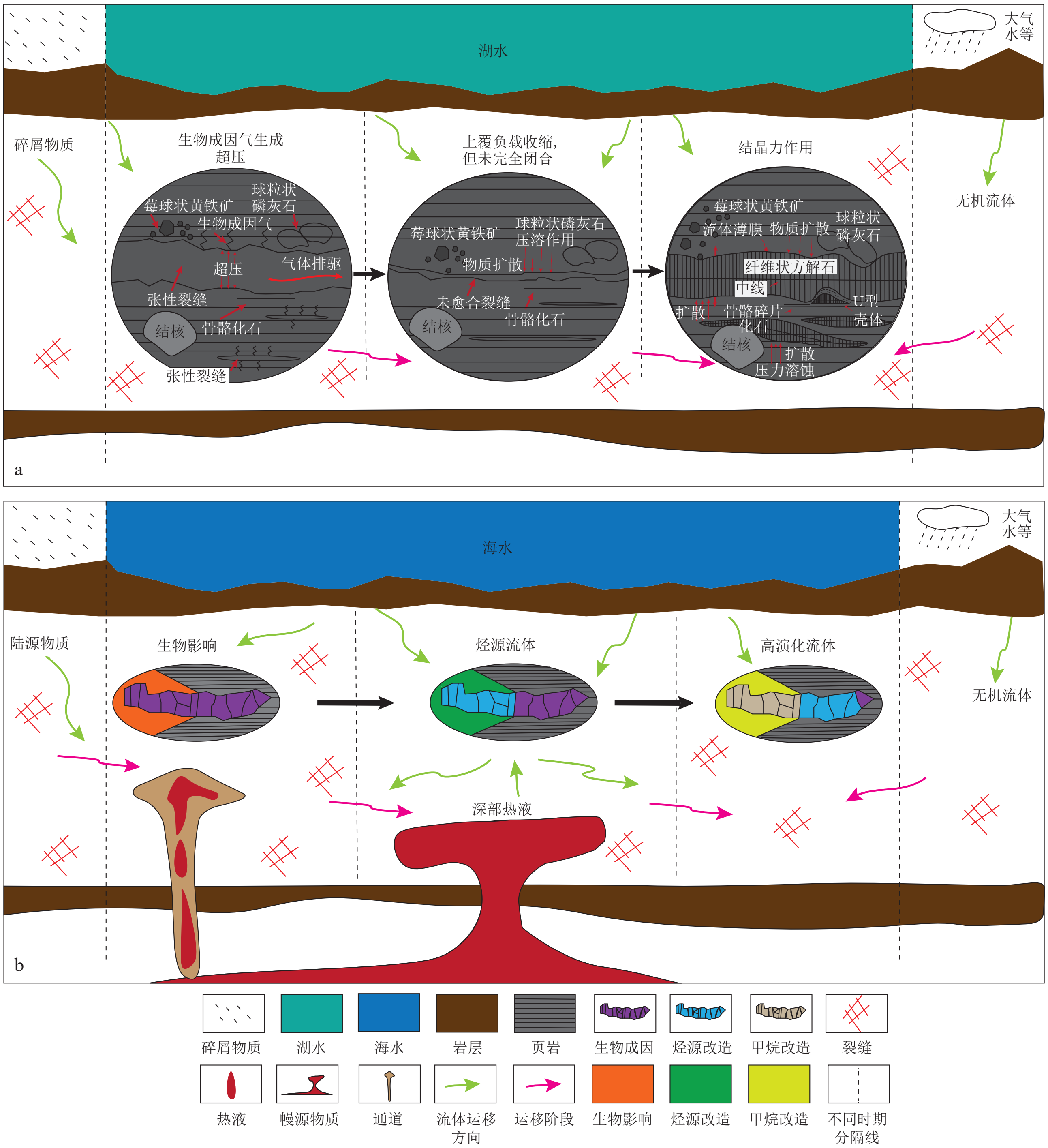

图6 陆相页岩(a)和海相页岩(b)中方解石脉的形成过程

(a和b图分别据文献[130]和[133]修改。)

Fig. 6 Comparison of the formation processes of calcite veins in lacustrine and marine shales

阶段,方解石晶型完整,无溶蚀现象,富Fe,Mg和Mn而贫Al元素,^{13}C亏损而^{18}O同位素相对富集,成岩流体可能与甲烷热氧化作用有关[87,133]。

成岩热演化过程中除了地球化学差异,方解石还存在溶蚀窗和逆溶蚀现象(溶蚀窗指达到特定温度区间内方解石溶蚀效果显著增强;逆溶蚀现象指方解石受热演化过程中化学反应的影响发生重结晶的现象)。在温度达到200 ℃之前主要表现为重结晶作用及方解石对钾长石的交代;当温度超过200 ℃阈值时,矿物溶蚀效应显著增强;在温度至250 ~ 300 ℃区间时局部区域出现重结晶特征[135]。上述现象表明方解石溶蚀并非温度越高溶蚀效果越强,而是在200 ~ 250℃范围内存在溶蚀窗,方解石溶蚀效果最佳,超过这个范围继续升温反而引发逆溶蚀现象(图7)。这可能与热化学硫

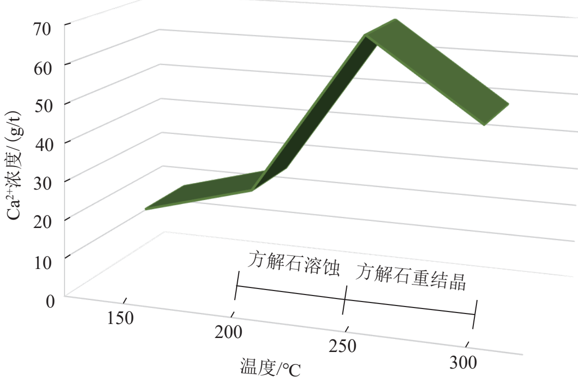

图 7 方解石沉积过程中溶液中钙离子浓度随温度的变化（数据来自文献[77]）

Fig. 7 Ca^{2+} concentration in a solution (data from reference [77])

酸盐还原反应）(TSR)释放的 Ca^{2+} 和 CO_2 及钙蒙脱石伊利石化过程产生的 Ca^{2+} 和 Mg^{2+} 等离子干扰水-岩平衡，从而诱发矿物二次结晶有关[77]。TSR 反应[136]与伊利石化[136-138]产生离子干扰的公式如下：

$$\text{硫酸盐} + \text{烃类} \rightarrow CaCO_3(CO_2) + H_2S + H_2O + \text{其他产物} \quad (1)$$

$$\text{钙蒙脱石} + K^+ \rightarrow \text{伊利石} + \text{硅质}(SiO_2) + \text{释放阳离子}(Ca^{2+}, Mg^{2+}, Na^+) + H_2O \quad (2)$$

4 方解石对古沉积与成岩环境的指示意义

页岩中方解石的含量对沉积环境具有指示意义[139-140]。页岩中发育保存完好的泥晶方解石，常指示低能静水的沉积条件，结合页岩自生黄铁矿、高有机质含量及氧化-还原敏感元素(Re，Mo 和 U 等)指标可进一步判别缺氧环境，如 Re 含量(> 1 ng/g)、元素富集因子比值 Mo_{EF}/U_{EF}(> 现代海水中的比值)和 Th/U 元素含量比值(0 ~ 2)等[141]。同时，页岩中较高的方解石含量表明陆源碎屑输入速率较低，在区域古地理背景下，这常常与离岸较远的深水-半深水沉积环境相关联[142]。

介壳作为页岩中方解石的一种生物骨骼赋存形式也可指示沉积环境[143]。块状介壳灰岩、页岩与介壳互层、页岩夹介壳层及纯页岩的不同组合对应不同的沉积单元环境，进而可指示古气候、古生产力、沉积速率和水体环境等，评估有机质富集的有利组合，这对评估不同层段的页岩油气资源潜力具有重要价值[144-145]。从表 4 各项数据可以看到，块状介壳灰岩段、页岩与介壳层互层段和页岩夹介壳层段和纯页岩段都基本处于缺氧环境，而古气候从干旱逐渐变潮湿（气候指数 *C* 值越大潮湿度越高），再结合 *TOC* 与古生产力指标分析可知，页岩夹介壳层段与纯页岩段明显展示出更大的生烃潜力。

通过分析页岩中方解石包裹体信息可以反演古沉积与成岩环境[147-149]。方解石包裹体的形成机制、成分及分布特征可反映沉积物的来源、搬运路径及沉积环境的物理-化学条件[24,150]。方解石包裹体的同位素组成($\delta^{18}O$ 和 $\delta^{13}C$)和微量元素含量可指示流体的性质、来源及与围岩的相互作用过程。如在生油窗内大量生气过程中方解石捕获的富 CH_4 包裹体，其 CH_4 的 $\delta^{13}C$ 可用于计算成熟度（镜质体反射率计算值 R_c）（图 8a，b），成熟度大幅增加的方解石包裹体流体中，除了 CH_4 外还含有 CO_2，推测是该沉积岩段的热液蚀变作用造成[151]。在成岩过程中，方解石包裹体的形成与演化记录了流体的迁移路径、压力变化及温度条件，为重建古沉积环境和成岩过程提供了重要信息[152]。同时，页岩中方解石包裹体的荧光特性可指示有机质的成熟度和热演化程度（蓝色—黄绿色—橙黄色—微弱褐光、无荧

表 4 不同岩性段有机质丰度及古沉积环境指标（数据来自文献[146]）

Table 4 Statistics of organic matter abundance and palaeosedimentary environment indicators across lithological units (data from reference [146])

岩性段类型	古生产力	古盐度	沉积速率	氧化-还原环境	古气候	有机质丰度
	$Ba_{bio}/10^{-6}$	Sr/Ba	$(La/Yb)_N$	V/(V + Ni)	气候指数(*C*)	*TOC*/%
块状介壳灰岩段	204.48	4.90	1.34	0.376 ~ 0.724 0.612	0.014 ~ 0.044 0.031	0.03 ~ 1.18 0.35
页岩与介壳层互层段	530.10	1.00	1.22	0.466 ~ 0.814 0.718	0.027 ~ 0.848 0.489	0.27 ~ 2.25 0.72
页岩夹介壳层段	625.40	0.40	1.23	0.557 ~ 0.811 0.728	0.136 ~ 0.950 0.609	0.42 ~ 3.11 1.34
纯页岩段	596.84	0.37	1.18	0.584 ~ 0.827 0.748	0.265 ~ 1.008 0.640	0.32 ~ 2.40 1.17

注：表中氧化-还原环境、古气候和有机质丰度的数据格式为“$\frac{\text{最小值} \sim \text{最大值}}{\text{平均值}}$”。元素符号代表该元素的含量；$Ba_{bio}$ 为生物 Ba 含量；$(La/Yb)_N$ 为标准化比值。

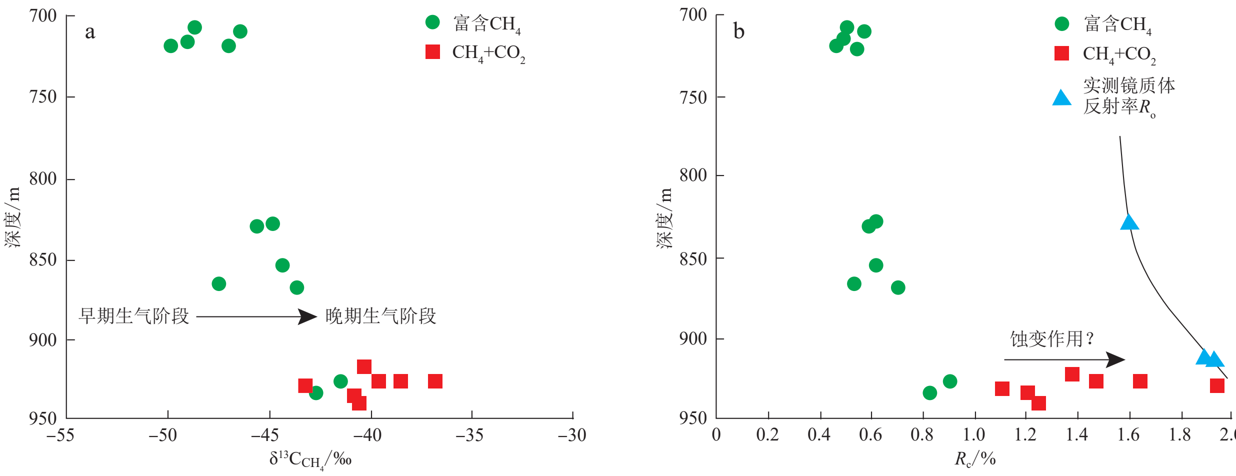

图8 页岩中方解石流体包裹体甲烷$\delta^{13}C$与成熟度(R_c)的垂向演化规律(数据来自文献[154])

Fig. 8 Variations in $\delta^{13}C$ values and R_c of fluid inclusions in calcites in shales (data from reference [154])

a. 方解石裂隙充填物中流体包裹体甲烷稳定碳同位素比值($\delta^{13}C_{CH_4}$)随深度的变化;b. 成熟度R_c(R_c为据包裹体$\delta^{13}C$计算出的镜质体反射率)随深度的变化

光,对应未成熟—生油窗—湿气窗—干气窗),对烃源岩评价和油气勘探具有重要意义[153]。

5 方解石对页岩力学性质和脆性的影响

页岩脆性指数直接关系到压裂的效果,是页岩油气储层评价的一个重要参数[155-157]。基于矿物组成的页岩脆性评价方法可遵循如下公式进行计算:

$$R_{BI}=\frac{W_{si}+W_{car}}{W_{si}+W_{car}+W_{clay}} \quad (3)$$

式中:R_{BI}为基于矿物组成的岩石脆性指数,无量纲;W_{si},W_{car}和W_{clay}分别为长英质矿物含量、碳酸盐质矿物含量和黏土矿物含量[158],%。

页岩的脆性大部分情况下主要受石英和黏土矿物的主导[159],但在碳酸盐富集层段,硅质矿物与碳酸盐组分共同构成裂隙发育的矿物基础。高脆性页岩层(以碳酸盐-硅质混合相为特征)在应力加载时更易发生破裂。尽管方解石可增大页岩脆性指数并参与酸性溶蚀反应,但其在孔隙演化中表现出双重效应:一方面通过增强脆性帮助页岩裂缝扩展,另一方面通过晶间充填作用抑制基质孔隙度[160-161]。同时方解石的断裂韧性较高,这意味着它具备较高的强度与一定的塑性。页岩中适量的方解石会增加其脆性,而过量的方解石则会使页岩产生高内聚力,阻碍裂缝的形成和扩展(图9)[162]。

实验证明页岩抗拉强度与方解石脉方位角呈负相关关系。当加载方向与脉体垂直时抗拉强度最高为3.562 MPa,平行加载时则降至3.110 MPa。二者比值

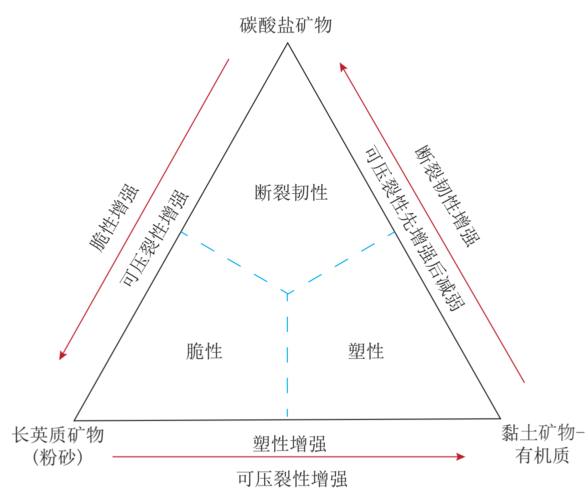

图9 不同岩石成分的力学性质及其对岩石可压裂性的影响(据文献[162]修改)

Fig. 9 Mechanical properties of various rock components and their impacts on rock fracability (modified after reference [162])

为1.15[163]。方解石脉的增韧作用在倾角0°~15°区间最为突出,超过15°后作用减弱(图10)。这一现象表明,方解石脉的空间分布是导致页岩抗拉性能各向异性的关键因素[164]。

6 方解石对页岩储层孔隙和裂缝的影响

方解石对页岩储层主要呈现3方面影响:①通过胶结作用固结矿物颗粒,致粒间孔隙丧失,储层渗透能力降低;②在成岩阶段经历溶解作用产生次生

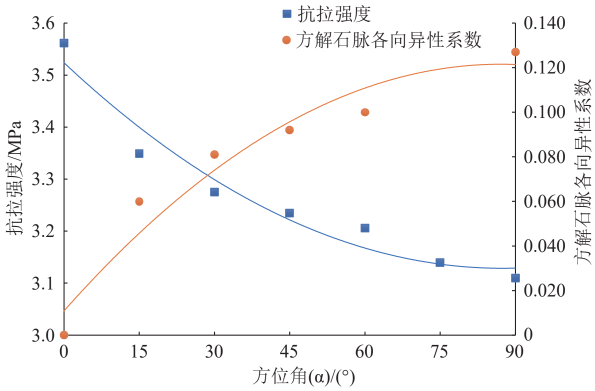

图 10　不同方位角下页岩抗拉强度和方解石脉各向异性系数变化趋势（数据来自文献[163,165]）

Fig. 10　Trends of shale tensile strength changes and the anisotropy coefficient variation of calcite veins at different azimuths of calcite veins (data from references [163, 165])

孔隙系统；③方解石的存在显著增强了页岩的裂缝发育潜力（表 5）[112,166]。

成岩演化分析表明，方解石矿物经历多阶段地质改造，包括转化、压实、溶解、胶结及重结晶过程。溶解过程可改善储层物性，而与之相反，机械压实、胶结固化及晶体重结晶过程则导致储集空间致密化[56,167-168]。方解石胶结物在页岩成岩过程中形成晶间孔隙系统与层状微孔隙结构，可适度改善储气物性，但溶蚀作用强度较低，仅在方解石矿物内部形成微米级溶蚀孔隙（图 11a），且孔隙喉道连通性较差，次生孔隙对物性改善效果有限[169]。值得注意的是晚成岩阶段铁方解石的交代作用，不仅造成黏土矿物微孔隙被充填，同时引发孔隙喉道堵塞，导致页岩储层孔隙度显著降低[160]。页岩中方解石脉发育到一定规模时会形成复合裂隙网络（以层理缝与溶蚀缝为主），有利于构建立体输导体系，显著改善烃类流体的渗流条件。方解石的差异沉淀与溶解作用会重塑孔隙结构，而其含量分布则直接控制裂缝发育密度[170]。因此，系统研究钙质矿物的赋存状态及成因模式，有助于揭示储集空间特征，为非常规油气甜点区预测提供关键依据[171]。

有机质成熟过程中的脱羧基作用会释放大量 CO_2，形成酸性流体环境，方解石等酸溶性矿物被溶蚀，形成溶孔，储层孔隙度也随之增加，部分孔隙中可见有机质充填（图 11b）[84]。在背散射电子图像下可见有少量的微裂缝（图 11c，d），且大多数被方解石、硅质和黄铁矿全充填。基于裂缝形成机制与几何形态，充填于裂隙中的方解石脉可划分为 3 类：① Ⅰ 型对应高角度构造缝的钙质充填体；② Ⅱ 型为网状构造缝的方解石胶结物；③ Ⅲ 型属于非构造成因的不规则脉状矿物。构造活动强烈的斜坡带与断背斜轴部以 Ⅰ 型和 Ⅱ 型脉体为主，而宽缓背斜及向斜核部则主要富集 Ⅲ 型脉体[172]。

方解石包裹体提供了油气运移与页岩裂缝周期性开启—闭合的对应信息[173]。对方解石捕获的原生包裹体研究发现，油气活动的初始运聚期常与方解石生长时间相近，表明在生烃增压或构造变形引发裂缝开启后，烃类运移与方解石胶结具有同期性，因而方解石脉体可持续记录油气的运移历史[172,174]。

7　总结与展望

1）页岩中方解石按粒度大小、产出形态和长/短轴比等特征划分为多种类型，主要包括泥晶方解石胶结物、分散颗粒状、结核状、纹层状和生物成因泥晶方解石，以及纤维状、刃状和等轴状亮晶方解石。其中纤维状亮晶方解石脉是流体超压存在的重要证据。页岩中方解石脉的形成受流体超压、区域构造

表 5　中国部分含油气盆地页岩储层中的方解石对比

Table 5　Comparison of calcites in shale reservoirs in some petroliferous basins of China

盆地	富方解石主要地层	页岩中方解石对压裂效益的影响	典型研究案例（文献）
松辽盆地	青山口组、嫩江组	积极：长岭凹陷青山口组一段页岩压裂后，裂缝扩展效率提高	[178-180]
		消极：林甸地区青山口组一段页岩局部层段，过量方解石胶结导致原生孔隙被充填，压裂液难以有效注入，裂缝扩展受阻	
渤海湾盆地	沙河街组、孔店组、嫩江组	积极：沙河街组四段上亚段方解石纹层组合、沾化凹陷沙河街组钙质页岩脆性指数显著提升，有利于水力压裂形成复杂裂缝网	[181-182]
		消极：沾化凹陷沙河街组局部层段原生孔隙被充填，渗透率降至 0.5×10^{-3} μm²以下，压裂液难以有效注入	
四川盆地	茅口组、五峰组-龙马溪组	积极：五峰组和龙马溪组底部的钙质页岩层段显著提升岩石的脆性指数，促进水力压裂过程中形成复杂缝网	[183-184]
		消极：在黏土矿物主导的页岩层段（如川东北地区部分层段），方解石胶结物过量堵塞原生孔隙	

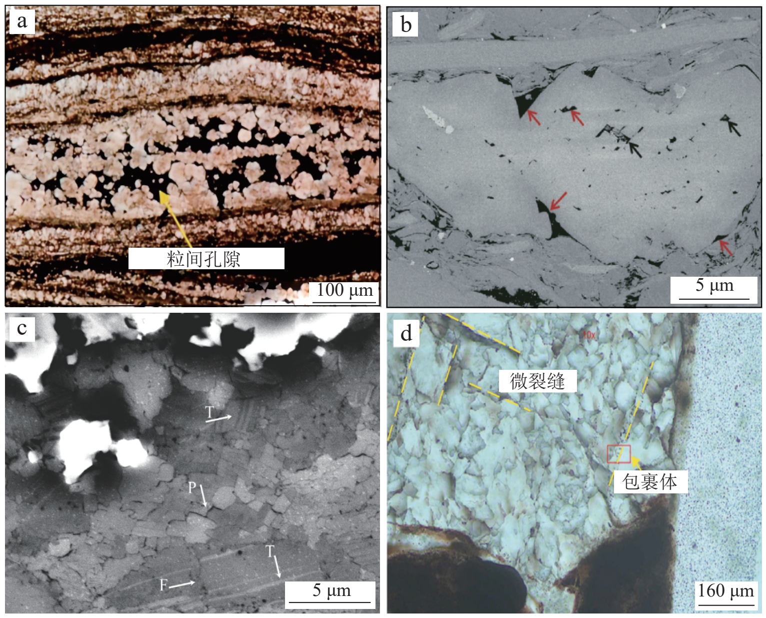

图11 页岩中方解石的孔隙与裂缝特征显微照片

Fig. 11 Pores and fractures in calcites in shales

a. 孔隙(黄色箭头)分布于亮晶方解石晶体之间,孔隙的长轴长度超过100 μm,且孔隙空间有效连通,渤海湾盆地东营凹陷,F1井,埋深3 440. 80 m,单偏光[175],普通薄片;b. 方解石中的溶蚀孔隙,被有机物充填(红色箭头)和未被有机物充填(黑色箭头),四川盆地龙马溪组,永页1井,背散射电子图像[176];c. 页岩中横切断层方解石的背散射电子图像,脉迹交叉处的超细方解石颗粒含有裂缝(F)、双晶(T)、交错晶界以及晶界处的孔隙(P),美国Copper Creek断层[177];d. 微裂缝和包裹体在亮晶方解石脉中发育,渤海湾盆地东营凹陷,N55-X1井,埋深3 590. 17 m,单偏光[67],普通薄片

应力及结晶力共同控制。陆相与海相页岩中方解石形成机制相似,但物源主体不同。在成岩热演化过程中,方解石在200 ℃前以重结晶和交代作用为主,200 ~250 ℃存在溶蚀窗,250 ℃以上因离子干扰引发逆溶蚀。

2)页岩中方解石可指示沉积环境。页岩中发育保存完好的泥晶方解石,常指示低能静水的沉积条件,结合氧化-还原指标可进一步判别缺氧环境。页岩与介壳的不同组合可关联不同的古环境指标,为生烃潜力评估提供参考。方解石包裹体则蕴含古沉积与成岩环境的重要信息。

3)充填于页岩孔隙与裂缝中的方解石会导致页岩致密化,降低页岩的储层物性,但早、中成岩阶段的方解石若遭到烃类流体的溶蚀作用改造,可产生次生溶蚀孔隙,对储层物性有一定改善作用。成岩晚期方解石对页岩的影响主要与超压作用相关,受超压影响方解石破裂产生孔隙与裂缝,极大地改善页岩的储层物性。由此可见方解石对页岩储层物性的影响是把双刃剑,即可抑制也可改善储层物性,具体是积极作用占主导还是消极作用占主导,则需要对不同地区的具体情况展开具体分析。

4)页岩中方解石流体来源与演化机制的多维度解析、烃类活动与方解石沉淀的耦合关系、方解石对页岩储层物性与油气赋存的"临界效应"、基于机器学习方法(如随机森林算法)建立方解石含量-脆性指数-产能预测模型等都是未来页岩中方解石研究的重要方向。同时未来还需要进一步对比研究各不同富方解石页岩油气区的方解石成因、分布规律及其对储集空间的控制作用,指导页岩油甜点评价及预测。

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