四川盆地下古生界硅质页岩层系中硅质来源及其对有机质保存的影响初探

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内容提要:四川盆地下古生界两套硅质页岩层系(寒武系底部页岩和奥陶系—志留系五峰组—龙马溪组页岩)是其重要的页岩气产层,五峰组—龙马溪组硅质含量与 *TOC* 及含气量呈正相关性,暗示硅质矿物对该地区页岩气的生成具有重要意义。但硅质来源复杂,且存在后期硅质流体的注入。因此,本文在总结前人的研究成果基础上,对四川盆地寒武系底部硅质岩系与五峰组—龙马溪组硅质岩系的硅质特征进行了详细的岩石学分析,揭示了3种硅质来源特征:①同沉积无机硅质流体;②生物硅;③后期无机硅质流体。其中寒武系底部以同沉积无机硅质流体与生物硅来源为主,同沉积硅质流体导致有机质快速石化埋藏,有机质内部结构及形态保存较好;五峰组—龙马溪组硅质来源以后期无机硅质流体与生物硅来源为主,有机质在保存过程中受硅质流体的影响很小,有机质腐泥化作用充分,内部结构和形态多数保存较差。该研究初步揭示了两套页岩层系的硅质发育岩石学特征,及其对有机质保存的影响,期望能为四川盆地两套页岩气储层差异性提供新的思路与基础地质数据。

关键词:硅质页岩;硅质来源;有机质保存;下古生界;四川盆地

页岩气资源,是自生自储于富有机质页岩层系中的非常规天然气资源(Jarvie et al., 2007; Loucks and Ruppel, 2007)。四川盆地及其周缘是中国最重要的页岩气产区,具有页岩气地质储量为 62.56×10^{12} m^3,可采储量 9.94×10^{12} m^3(Zhang Yuying et al., 2019)。该地区下古生界发育两套重要的页岩气储层,下寒武统牛蹄塘组或筇竹寺组(孙玮等,2012;谢小敏等,2015; 周泽等,2016;Leng Jigao et al., 2016; Liu Honglin et al., 2016; Wang Ruyue et al., 2016; Zeng Weite et al., 2016;Zhang Yuying et al., 2018; 冯明友等,2018;吴安彬等,2020)与上奥陶统五峰组—下志留统龙马溪组(金之钧等,2016;冯动军等,2016;Li Ang et al., 2017; Shao Xinhe et al., 2017; 魏祥峰等,2017)。前期研究表明,龙马溪组页岩气优质储层通常位于该套地层底部富含硅质页岩层段,其自然伽马值较高(>150 API),岩石有机碳含量高(>2%),含气量高(>2 m^3/t;王玉满等,2012;刘树根等,2011,2013;郭彤楼和张汉荣,2014;赵佩等,2014;毕赫等,2014)。以典型的焦页1页岩气井为例,五峰组—龙马溪组下段页岩 *TOC* 含量为0.55%~5.89%,平均2.5%,其中底部硅质页岩 *TOC* 含量最高(2.56%~4.97%,平均3.89%),是该井页岩气的主要产气层段(金之钧等,2016)。前人的研究发现,涪陵地区五峰组—龙马溪组下部优质页岩硅质矿物含量与 *TOC* 含量具有较好的正相关性,硅质矿物含量越高,有机碳含量越高(金之钧等,2016; Yang Feng et al., 2016; Xu Zhuang et al., 2019);同时岩石孔隙度与硅质矿物之间也具有很好的正相关性(Yang Feng et al., 2016),暗示硅质组成对该地区页岩气生成具有潜在的重要意义。

吴蓝宇等(2016)对涪陵地区典型五峰组—龙马溪组页岩气井研究发现,这套硅质页岩发育地层有机碳含量与含气量之间具有很好的相关性(R^2 =

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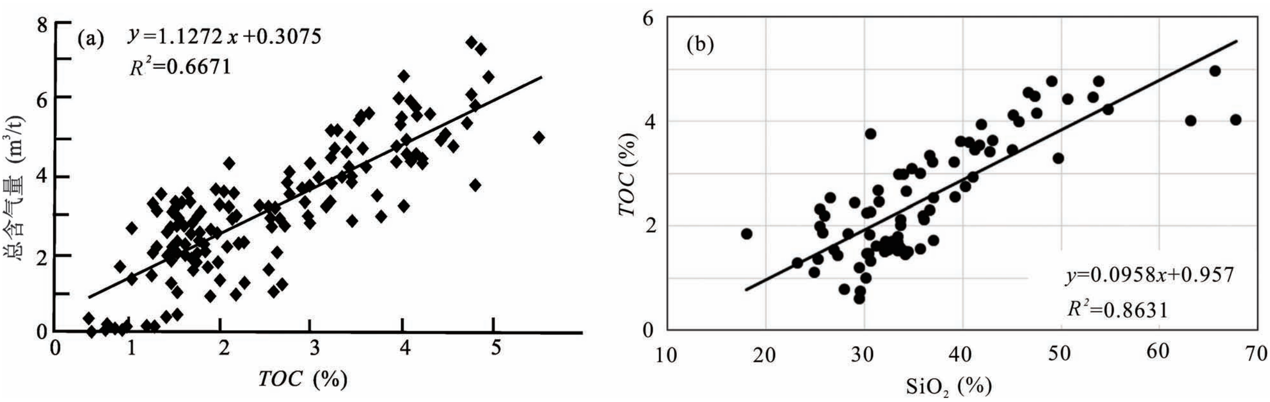

图 1 四川盆地涪陵气田五峰组—龙马溪组硅质页岩层段页岩含气量与 *TOC*(a, 据吴蓝宇等,2016& 修改)和 *TOC* 与 SiO_2 含量(b,据 Xu Zhuang et al., 2019 修改)相关散点图

Fig. 1 Relationships of gas content vs *TOC* (a, modified from Wu Lanyu et al., 2016) and *TOC* vs SiO_2(b, modified from Xu Zhuang et al., 2019) in the shale members of the Wufeng Formation—Longmaxi Formation in Fuling gas field, Sichuan Basin

0.6671,图 1a)。同时,该层段 *TOC* 含量与 SiO_2 含量之间具有很好的相关性(图 1b,Xu Zhuang et al., 2019)。而非优质页岩段硅质矿物含量与 *TOC* 含量相关性较差,揭示了优质页岩段与非优质页岩段硅质矿物成因的不同(金之钧等,2016)。因此,五峰组—龙马溪组硅质页岩储层中硅质含量与 *TOC* 及含气量呈正相关性,暗示硅质矿物对该地区页岩气的生成具有重要意义。

但由于硅质来源与成因较复杂,有学者对该地区下古生界硅质岩的成因也做了详细的研究,并计算了过剩硅(excess silicon)的含量(卢龙飞等,2016;刘江涛等,2017;Zhang Kun et al., 2018)。但实际上,过剩硅是指高于正常碎屑沉积环境下的 SiO_2 含量,并不能区分硅质来源的生物硅与热液硅特征,而热液硅也存在同沉积与后期注入的不同。因此,本文在前人研究的基础上,采集了贵州羊跳地区寒武系底部牛蹄塘组野外剖面样品,与四川盆地焦石坝与丁山地区五峰组—龙马溪组岩心样品,进行了显微镜下岩石学特征的精细观察。提出了三种可能的硅质来源,并初步探讨了两套硅质页岩层系硅质来源的不同及其对有机质保存的影响,期望能为四川盆地两套页岩气储层差异性提供新的思路与基础地质数据。

1 关于硅质岩的成因

硅质的成岩作用主要有化学作用、生物和生物化学作用及火山作用、热水作用(张亚冠等,2015)。前人对沉积岩中的硅质进行了岩石学、常量元素、微量元素、稀土元素、同位素分布特征的研究,揭示了硅质的成因认识。目前有 3 种主要的成因认识:

(1)热液成因:在常温下海水中的硅含量极低($<10\times10^{-6}$;Calvert, 1983),不可能达到饱和而直接沉淀,仅靠放射虫和海绵等硅质生物形成厚度很大的硅质沉积是很难的(涂光炽,1990)。而硅的溶解度随着水温的增高而迅速增加,海水中硅的含量在 150℃时达到 600×10^{-6}(Krauskof, 1956),在 200℃时海水中的含硅量是 50℃ 的 10 倍(Holland, 1967)。富硅的热水遇到冷海水后,硅的含量可能超过常温水中溶解度的 10~20 倍(Rona, 1978),呈过饱和状态。因此,在海底热泉附近可能有硅质的直接沉淀(Rona et al., 1980)。热泉附近因营养丰富,硅质生物及其他生物十分繁荣,Dick(2019)在现代东太平洋海底热液喷口黑烟柱附近观察到了大量的微生物和底栖动物。20 世纪 80 年代以来,国内外很多专家学者对硅质岩研究表明,海底或大陆许多硅质岩为热液成因硅质岩(Adachi et al., 1986; Yamamoto, 1987; 陈洪德与曾允孚,1989;伊海生等,1989;周永章,1990;周永章等,1994;夏邦栋等,1995;冯胜斌等,2007;李凤杰等,2010)。寒武系底部硅质岩发育时期,存在大规模的火山活动和海底喷流作用;这为硅质岩的热液来源提供了必要的条件,同时也导致了寒武纪早期海洋的物质循环与水体化学性质的特殊性(Steiner et al., 2001; Kamber and Weeb, 2001; Chen Daizhao et al., 2009)。Adachi 等(1986)和 Yamamoto(1987)认为热液中形成的硅质具有 SiO_2 含量高,Al_2O_3 和 TiO_2 含量低的

特征。Bostrom(1983)认为与热液沉积有关沉积物中,富 Fe、Mn,贫 Cu、Co 和 Ni;而 Al 的富集则与陆源物质输入有关。因此,Al/(Al+Fe+Mn)值可作为衡量沉积物中热液沉积物含量多少的标志(Bostrom et al., 1969; Adachi et al., 1986; Yamamoto, 1987);(Cu+Co+Ni)—Fe—Mn 三角图也用于区分热液和非热液沉积(Rona, 1988)。

(2)生物成因:硅质主要来源于硅质生物,如海绵骨针、放射虫、硅鞭藻、硅藻等(Boggs, 2006; Robert and Stephen, 2007; Day-Stirrat et al., 2010; Milliken et al., 2012)。五峰组—龙马溪组产气层段硅质页岩中石英含量高,一般大于 45%,认为这类硅质主要来源于生物硅(杨玉卿和冯增昭,1997; Schieber et al., 2000; Loucks and Ruppel, 2007; 杨水源和姚静, 2008; 卢龙飞等, 2016)。Yang Xiangrong 等(2018) 分析了硅质岩中的石英含量与生物钡含量呈正相关,认为生物硅质是该地区重要的硅质的来源。王玉满等(2016)对川南龙马溪组页岩岩相进行了划分,认为硅质页岩(华蓥—长宁一带)和钙质硅质混合页岩(威远—泸州—綦江以南(王玉满等,2015)是深水沉积环境的特有岩相,发育水深在 100 m 以下,富含有机质,古生物化石,如放射虫、海绵骨针、尖笔石等(刘伟等,2010;樊隽轩等,2012,2013;张春明等,2012;王淑芳等,2014)。在早寒武世硅质岩中也发现了大量的放射虫和海绵骨针等硅质生物(张位华等, 2003; 谢小敏等, 2015)。因此,硅质生物是这两套页岩储层硅质的一个重要来源。

(3)交代成因:交代成因硅质是指在岩石沉积之后,硅质流体交代原生矿物形成的硅质矿物。一般来说,交代成因硅质岩的原岩多为碳酸盐岩,有成岩期交代(杨吉与李英,2002)和地表次生交代等多种类型(李文恒,1980)。朱嗣昭(1999)在南京及巢湖地区下二叠统孤峰组中发现了这类硅质岩(称为“浮石状隧石”),是一种交代碳酸盐岩成因的硅质岩,显著特征是岩石中存在大量菱形和不规则状孔隙,孔隙周边和孔隙间主要由微晶石英组成。施贵军等(1999)在皖南浙西交界地区黄龙组也发现交代成因硅质岩。郭福生等(2003)对浙江江山晚石炭世藕塘底组上段发现“钙骨假象燧石岩”,特征是大量钙质生物碎屑(包括腕足类、海百合茎,珊瑚等)被完全硅化,常见由多个微晶石英交代的自形细晶白云石假象以及碳酸盐交代残余;元素 Al/(Al+Fe+Mn)值显示,该地区硅质岩全部属于正常海水沉积区,微量元素与稀土元素特征等显示该硅质源于与热水沉积无密切关系,属于成岩交代成因硅质岩。Marin-Carbonne 等(2011)对南非 Onverwacht Group 的燧石进行综合的原位氧同位素与硅同位素,及硅质脉体中流体包裹体特征,认为该燧石中硅质为交代成因。Rakociński 和 Borcuch(2016)在波兰晚泥盆弗拉斯阶碳酸盐岩中也发现了硅化的头足类动物化石,认为快速的硅化有利于化石带的保存。

实际地质样品中,硅质的成因往往是综合的,如生物与热水综合成因(田云涛等,2007)等。据 Clayton (1986) 与 Douthitt (1982)等的研究,不同来源硅质具有不同的 δ^{30}Si 值,从低温地下水中自生沉淀的石英 δ^{30}Si 值(>1.4‰)>成岩过程中次生加大石英的 δ^{30}Si 值(0.8‰~1.4‰)>热水来源石英的 δ^{30}Si 值(−1.5‰~0.8‰)。宋天锐和丁悌平(1989)分析认为,生物成因硅质岩的 δ^{30}Si 值为−1.1‰~1.7‰, 交代成因硅质岩的 δ^{30}Si 值为 2.4‰~3.4‰, 火山喷发—化学沉积硅质岩的 δ^{30}Si 值为−0.4‰~−0.5‰。此外,常量元素组成、微量元素组成、稀土、放射性同位素(Rb-Sr、K-Ar、Pb 等)、稳定同位素(H、C、O、$^{87}Sr/^{86}Sr$、Sm-Nd、Si 等)、稀有气体元素(Ar)和有机地球化学等方面对不同成因硅质进行了精细的研究工作(宋天锐与丁悌平,1989; Murray, 1990, 1992; Murray et al., 1991; 盛吉虎与陈中新,1998),获得了不同成因硅质来源的地球化学信息。Holdaway 与 Clayton(1982)年提出过量硅(excess silicon)的概念,是指高于正常碎屑沉积环境下的 SiO_2 含量,其计算公式为 $Si_{excess} = Si_{sample} - (Si/Al)_{background} \times Al_{ample}$, 其中 $(Si/Al)_{background}$ 采用平均页岩比值 3.11,该方法广泛应用于硅质矿物研究中(刘江涛等,2017;Zhang Kun et al., 2018),但对于不同成因非碎屑来源的硅质的相对含量定量计算分析还比较缺乏。

2　寒武系底部硅质岩石学特征及其对有机质保存的影响

寒武系底部与五峰组—龙马溪组硅质页岩储层相比,寒武系底部硅质页岩具有机碳含量与有机质热演化程度更高(Tan Jingqiang et al., 2014; Wang Chao et al., 2018;Zhao Jianhua et al., 2018;Zhang Yuying et al., 2018, 2019),页岩储层厚度更大,脆性矿物(以硅质为主)更高(平均含量接近 80%; Zhang Yuying et al., 2019)等特点。然而,该层位页岩气的勘探开发并未取得很大的突破。虽然有学者

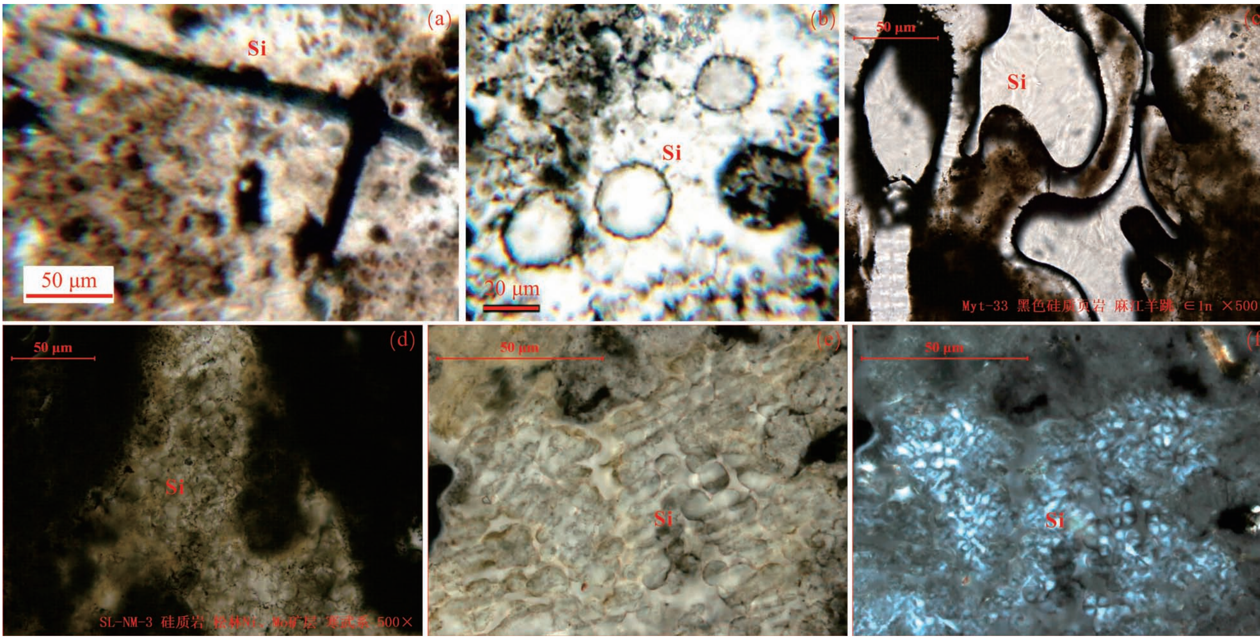

图 2 四川盆地边缘早寒武世牛蹄塘组底部硅质页岩显微照片：(a)—(c) 硅质页岩中保存较好的生物化石，(a)为海绵骨针，(b)为疑源类，(c)为须腕动物；(d)—(f) 具十字消光玉髓，(e)和(f)是(d)中玉髓的局部放大；(a)—(e)为透射光下偏光显微照片；(f) 为透射光正交偏光显微照片

Fig. 2 Micrographs of siliceous shales from Lower Cambrain Niutitang Formation in Sichuan Basin: (a)—(c) Well-preserved fossils in siliceous shales, (a) sponge spicule, (b) acritarch, (c) pogonophora; (d)—(f) chalcedony with cross extinction; (a)—(e) are micrographs of polarized transmitted light; (f) is micrograph of orthogonal transmitted light

将其归咎于寒武系页岩经历了更复杂的构造活动改造，裂缝发育，导致后期保存条件较差(Zhang Yuying et al., 2019)。但实际上，两套硅质页岩储层的硅质组成不同，也可能是其原因之一。

在四川盆地边缘贵州寒武系底部牛蹄塘组硅质页岩层系中，硅质岩呈层状、透镜状产出(谢小敏等，2015)，显微镜下硅质透光下呈圆形，正交光下具十字消光(图 2)，与美国黄石公园硅质热泉沉淀特征类似(Guidry et al., 2003)。谢小敏等(2015)在贵州寒武系地层中也见到了玉髓化现象和典型的热水生物，即须腕动物蠕虫(Monika et al., 2005; Kenneth, 2005; 梁狄刚等，2009)，从而认为该套硅质岩从形成与海底热液活动有关。寒武纪早期是大陆内部强烈拉张作用引起强烈的热液作用时期，前人通过微量元素分析、稀土元素、同位素地球化学、岩石学与矿物学分析，已做了大量精细的研究确定了这套地层沉积时期受到海底热液的影响(李胜荣等，1995; Steiner et al., 2001; Jiang Shaoyong et al., 2003; 韩善楚等，2003; 杨剑等，2004; 杨兴莲等，2007; 杨瑞东等，2007; 谢小敏等，2015; Liu Zhanhong et al., 2015)。

在这样一套硅质页岩层系中，发现了很多生物形态保存较好的原生有机质，如海绵骨针、疑源类、须腕动物及其他藻类等(杨瑞东等，2005; 杨兴莲等，2010; 谢小敏等，2015)。有机质保存完好的部分都是在富含硅质的区域(图 2a～c)，如疑源类表面微小的纹饰都能清晰可见(图 2b)；且疑源类细胞呈圆形，未出现沉积有机质受细菌降解、成岩压实引起的细胞变形。一般来说，油气生成过程中，有机质在复杂的沉积—埋藏过程中会经历细菌腐泥化作用，后续成岩—生烃演化阶段，生烃作用也会改造有机质形态结构，最终使得有机质的形态保存较差，尤其是生成大量油气之后，形态保存较好的有机质就更少。因此，在传统显微镜和在高精度的扫描电镜下，能鉴定出来的形态特征保存较好的生物，是否就代表了烃源岩中主力的生烃有机质特征，存在较大的疑议(Hutton, 1994)。初步推测这些形态保存完好的生物有机质可能与同沉积的热液事件有关，热液的加入使得生物快速死亡—埋藏—保存在硅质岩层中。也就是说，同沉积热液硅质可能瞬间石化了有机质，从而很好的保护了有机质的外形，这类现象在马达加斯加的浅海海岸海洋玛瑙中比较常见(图

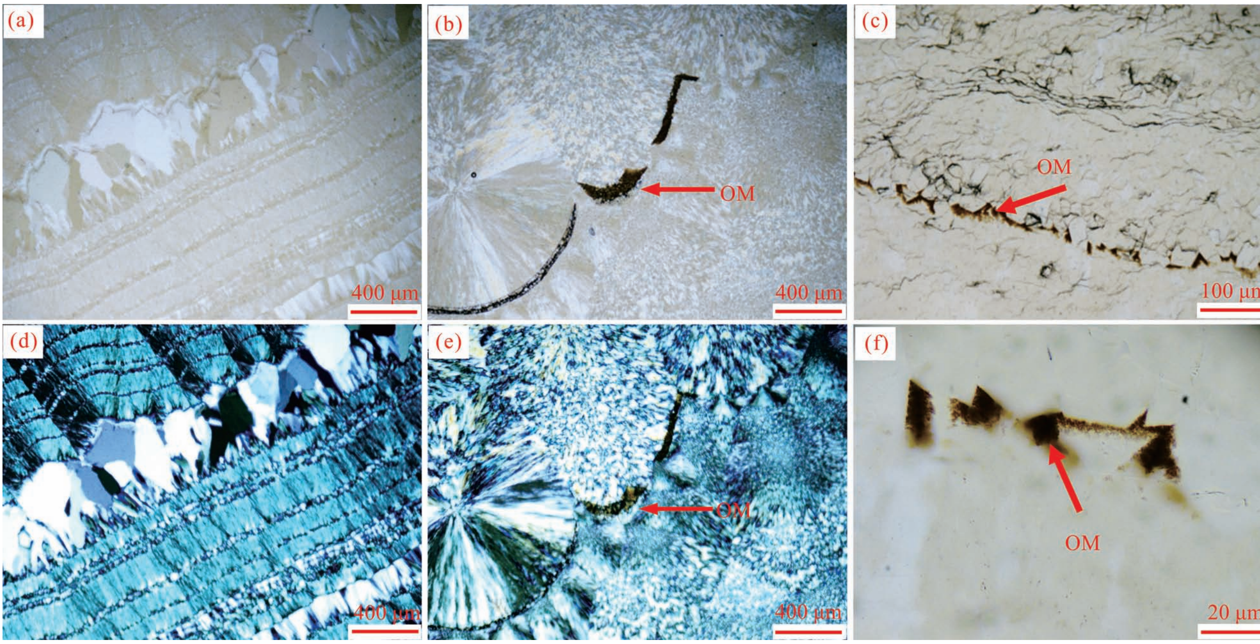

图 3 马达加斯加的浅海海岸海洋玛瑙显微照片,显示玉髓沉淀过程中包裹了有机质(OM)

Fig. 3 Micrographs of marine agate from the shallow coast of Madagascar, showing the inclusion of organic matter (OM) during the deposition of chalcedony

3)。这些保存较好的有机质一般发现在硅质含量较高的含泥或泥质硅质岩中,在这类岩石中,能指示成烃过程的固体沥青的含量并不高。而在硅质页岩或页岩中,有机质的保存相对较差,明显经历了沉积成岩及细菌的改造作用。这类快速被保存的有机质,由于并未经历缓慢的细菌腐泥化作用,后期埋藏过程中又受到硬度较大的硅质矿物的保护。

3　五峰组—龙马溪组硅质岩石学特征及其对有机质保存的影响

四川盆地五峰组—龙马溪组底部硅质页岩中硅质特征则完全不同,虽然有学者在该套硅质页岩中发现了海绵骨针和放射虫(卢龙飞等,2016;刘江涛等,2017),认为其主要是生物来源的。实际上,与寒武系底部的硅质页岩相比,五峰组—龙马溪组底部硅质页岩硅质生物的发育程度要少很多,且过量硅含量为 0~20%,而寒武系的为 20%~50%(Zhang Kun et al., 2018)。Yang Xiangrong 等(2018)对华南龙马溪组底部硅质页岩样品分析,发现 *TOC* 与硅质呈正相关关系时,可能与生物硅来源有关;但 *TOC* 大于 3.2 %时,*TOC* 与硅质含量呈现负相关关系(R^2 = 0.88,图 4)。是否可能在 *TOC* 高于 3.2% 的岩层中,硅质来源存在差异?

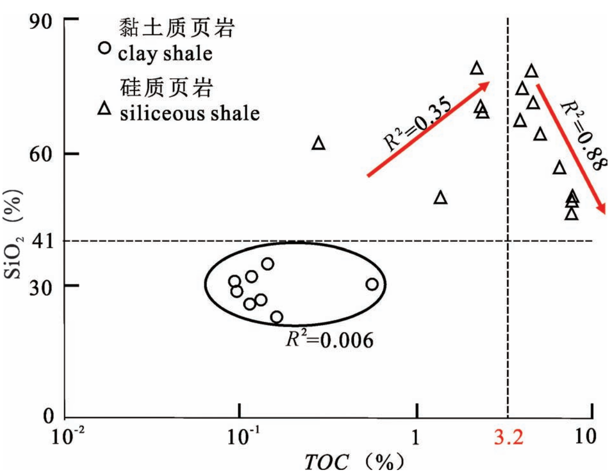

图 4 四川盆地志留系龙马溪组硅质页岩与黏土质页岩有机碳含量(*TOC*)与 SiO_2 含量相关关系图(据 Yang Rongxiang et al., 2018 修改)

Fig. 4 The plots of *TOC* vs SiO_2 contents of siliceous and argillaceous shale in Sichuan Basin (modified from Yang Rongxiang et al., 2018)

对丁山地区该套硅质页岩底部的薄片显微特征分析(图 5a—d),发现较多硅质脉体,有的硅质脉体切断了方岩储层底解石脉,或出现方解石脉被硅质脉体交代残余(图 5a、b),硅质颗粒边缘还残留少量钙质残余(图 5c),笔石皮层被硅化(图 5d)等现

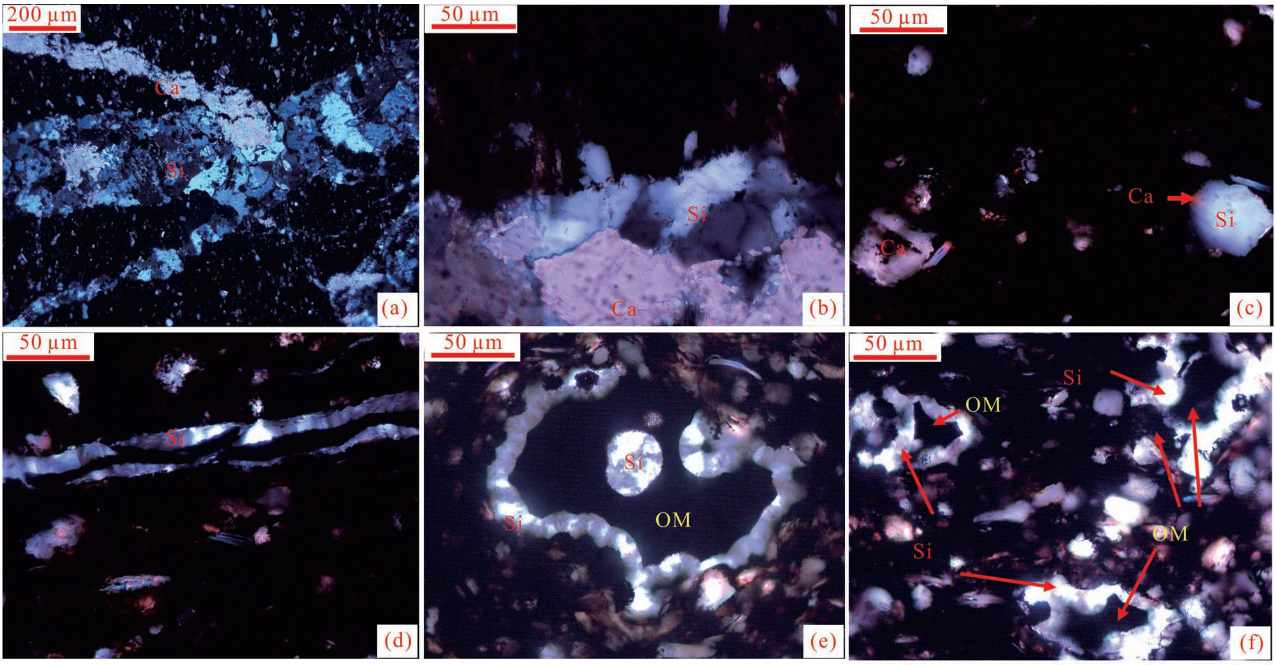

图 5 四川盆地丁山地区(a)—(d)和焦石坝地区(e)—(f)五峰组—龙马溪组底部硅质页岩中硅质正交光下显微照片:(a)、(b) 方解石脉被硅质条带交代残余;(c) 硅质颗粒边缘发现钙质物质残留;(d) 被硅化的笔石皮层;(e)、(f) 焦石坝地区五峰组—龙马溪组底部硅质页岩中,发育玉髓包裹有机质(OM)现象

Fig. 5 Microphotos under orthogonal light of siliceous shales of the lower Wufeng Formation—the Longmaxi Formation in Dingshan area (a)—(d) and Jiaoshiba area (e)、(f): (a)、(b) calcite veins are metasomatic with silica bands; (c) calcium residues were found at the edge of the silica particles; (e)、(f) the organic matter (OM) were found to be included by silica in Jiaoshiba area

象,表明硅质存在交代成因,切割关系说明硅质脉体充注的时间晚于方解石脉形成时间。在焦石坝地区,五峰组—龙马溪组底部见到较多的硅质包裹有机质的特征(图 5e、f),这类硅质正交下呈十字消光(玉髓现象),有机质的形态大小不一,棱角清晰,有机岩石学特征显示可能为固体沥青。也就是说,硅质可能是在油气生成的高峰时期注入,包裹了部分油(后期演化为固体沥青),这样形成了形态各异的硅质+有机质组合。这套硅质页岩中,除了笔石有机质保存较好外,藻类有机质主要呈无定形体保存,显示有机质在沉积埋藏过程中经历了充分的腐泥化过程,有助于后期油气的生成,晚期注入的硅质在对原生有机质的生烃潜力上影响不大。初步的电子探针分析,与焦石坝地区硅质较丁山地区硅质脉体富含 Fe 元素,显示不同的硅质流体充注特征。

4 结论

对寒武系底部硅质岩与五峰龙马溪硅质岩的特征分析揭示了 3 种硅质来源特征:①同沉积无机硅质流体;②生物硅;③后期无机硅质流体。虽然两套储层均存在硅质成分,但是硅质特征,来源及形成时间不同。其中寒武系底部以同沉积无机硅质流体与生物硅来源为主,同沉积硅质流体导致有机质快速石化埋藏,有机质(如疑源类等)内部结构及形态保存较好;五峰龙马溪组硅质来源以后期无机硅质流体与生物硅来源为主,有机质在保存过程中受硅质流体的影响很小,有机质腐泥化作用充分,内部结构和形态多数保存较差,以无定形体为主。因此,后期研究有必要将硅质岩的成分及来源进行精细分析,充分考虑硅质的岩石学特征及形成环境,区分同沉积时期的硅质与后期硅质矿物特征及含量,结合有机质含量、页岩储层特征进行相关性分析,探讨硅质成因对原生有机质和次生固体沥青有机质的影响,进而探讨其对页岩气生成的影响。

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Siliceous source and its influence on organic matter preservation of the two Lower Paleozoic siliceous shale reserviors in Sichuan Basin

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Abstract: Two sets of siliceous shale formations in Lower Paleozoic, the bottom of Cambrian and the Ordovician—Silurian Wufeng—Longmaxi Shales, are the important shale reservoirs in Sichuan Basin. The *TOC* and shale gas content of the Wufeng—Longmaxi shales are positively related to the siliceous quantity, suggesting that the siliceous materials could be very important for the shale gas formation. However, the origin of siliceous materials is complicated, and there could be some siliceous fluid filled after the rock sedimentation. This study, based on the summary of previous research results, has carefully analyzed the petrographical characteristics of siliceous minerals. Three types of siliceous sources have been detected: ① syngenetic hydrothermal inorganic siliceous fluid, ② bio-silica, and ③ late-filled inorganic siliceous fluid. Syngenetic hydrothermal inorganic siliceous fluid and bio-silica are the dominated siliceous sources in Lower Cambrian shales. Syngenetic hydrothermal inorganic siliceous fluid made the organic matter rapidly silicified, and the organic matter could be exquisitely preserved. The siliceous source in the Wufeng—Longmaxi Formation was mainly from late-filled inorganic siliceous fluid and bio-silica, and organic matter preservation has little influence by siliceous fluid. Thus, the organic matter has gone through slowly saprofication process, resulting in the morphology of organic matters are poor preserved. The study has initially revealed the petrographical characteristics of siliceous materials and the influences on the organism preservations for comparision the Lower Cambrian and the Wufeng—Longmaxi shale reservoirs, with hoping to provide new geological data and ideas for shale gas exploration in Sichuan Basin.

Keywords: siliceous shale; origin of siliceous materials; organic matter preservation; Lower Paleozoic; Sichuan Basin

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