

吉泰盆地石炭系黄龙组黑色碳质泥岩物源、 构造背景与古风化作用：来自稳定元素 地球化学的证据

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摘要: 吉泰盆地在白垩纪—古近纪时期, 沉积了巨量的富锂钾卤水, 同时富含硼、铷、铯、溴、碘等新兴矿产资源, 综合利用价值高。本文采用 X 射线荧光光谱(XRF)和电感耦合等离子体质谱(ICP-MS)分析了吉泰盆地石炭系黄龙组黑色碳质泥岩的主、微量元素和稀土元素地球化学特征。通过对其物源、构造背景及古风化作用的详细分析, 探讨了其对上覆卤水矿层的影响。吉泰盆地 M2 钻孔底部细碎屑岩具有 LREE 富集, HREE 较平缓的特征, 且均表现明显的 Eu 负异常, 综合 La/Th-Hf 和 Zr-TiO₂ 图解, 认为其主要来自长英质/基性岩浆岩和古老沉积组分的混合源区, 为卤水形成带来了丰富的深源物质。微量元素和稀土元素的含量和比值(La/Th、Cr/Th、Th/Sc), 反映出被动大陆边缘的构造背景特征。通过计算化学风化指数(CIA、CIW、PIA 和 ICV)及 Al₂O₃-(CaO+Na₂O)-K₂O 图解, 表明其化学风化程度较高, 有利于元素的迁移和富集, 为卤水矿床的形成提供了丰富的成矿元素。

关键词: 吉泰盆地; 地球化学; 物质来源; 构造演化; 风化作用

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Provenance, Tectonic Setting and Palaeo-weathering of Black Carbonaceous Mudstones from Carboniferous Huanglong Formation in the Jitai Basin: Evidence from Stable Element Geochemistry

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Abstract: In the Cretaceous–Paleogene period, a huge amount of lithium-potassium rich brine was deposited in the Jitai Basin, which is rich in boron, rubidium, cesium, bromine, iodine and other emerging mineral resources, with high comprehensive utilization value. In this paper, the major, trace and rare earth element geochemical characteristics of Carboniferous Huanglong Formation black carbonaceous mudstone in the Jitai Basin were analyzed by X-ray fluorescence spectrometry (XRF) and inductively coupled plasma mass spectrometry (ICP-MS). Based on the detailed analysis of its provenance, tectonic setting and paleo-weathering, the influence of it on the overlying brine ore bed is discussed. The fine clastic rocks in the lower part of the M2 borehole in the Jitai Basin are characterized by LREE enrichment and gentle HREE, and they all show obvious negative

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Eu anomaly. Based on La/Th-HF and Zr-TiO₂ diagram, they are mainly from the mixed source area of felsic/basic magmatic rocks and ancient sedimentary components, which brings abundant deep source materials for the formation of brine. The contents and ratios of trace elements and rare earth elements (La/Th, Cr/Th, Th/Sc) reflect the tectonic background characteristics of passive continental margin. The chemical weathering index (CIA, CIW, PIA and ICV) and the diagram of Al₂O₃-(CaO+Na₂O)-K₂O show that the chemical weathering degree is high, which is conducive to the migration and enrichment of elements, and provides rich ore-forming elements for the formation of brine deposits.

Key words: Jitai Basin; geochemistry; provenance; tectonic evolution; weathering

碎屑岩地球化学特征作为地质信息的重要载体,真实详细地记录了物源、构造、环境和生态演化等重要信息(王春连等,2022)。在沉积的过程中,水体和沉积物中对氧化还原敏感的微量元素的分布、循环和分异(亏损和富集)不仅与它们自身的化学性质有关,而且还受沉积介质的物理化学条件和古气候条件的控制(Nameroff et al., 2004; Tribouillard et al., 2004, 2006; Zhang et al., 2016; 兰叶芳等,2022; 游超等,2022; 周博文等,2022)。因此,一些溶解于水且对气候变化敏感的主微量元素可以作为研究古气候演化的重要代用指标。

中生代是华南乃至整个东亚大陆构造剧烈变化的时期,也是东亚构造发展的转折点(任纪舜,1990; 赵越等,2004; 董树文等,2007, 2010; Rodríguez-López and Wu, 2020)。白垩纪—古近纪时期,华南中部形成了江汉盆地、吉泰盆地等一系列断陷盆地。此时,这些盆地中的大多数湖泊演化成盐湖,并沉积了大量的盐岩和其他盐类矿物(姚秋昌和楼基胜,2008)。目前,在吉泰盆地周田组晚白垩世地层发现深层卤水,富含钾、锂、硼、铷、铯、溴、碘等高价值新兴战略矿产资源,其中 NaCl 的品位为工业品位的 2.6 倍, LiCl 的品位为工业品位的 3.8 倍,具有较大的工业价值(刘成林等,2009; 刘成林, 2013; 周敏娟等, 2017; 王春连等, 2020; 马厚明等, 2021)。

深部卤水沉积的形成受物质来源、沉积期古气候条件和盆地构造条件等因素的控制(刘成林等, 2010; 魏海成等, 2016; Sun et al., 2017; 王春连等, 2018, 2021)。目前吉泰盆地的基础地质研究还显薄弱,现有资料不足以揭示深层卤水的形成机理。为了更好地了解吉泰盆地深层卤水的成因,各成矿元素的来源及富集程度,本文报道了黄龙组黑色碳质泥岩的主、微量、稀有元素地球化学数据,探讨其对上层周田组卤水矿产的影响。

1 地质背景

吉泰盆地位于江西中部隆起与西南凹陷的过渡地带,在中生代燕山运动时该区陆壳在隆起的背景上表现为急剧而广泛的差异性断块运动,并在先期和新生断裂的影响与制约下,形成了吉泰断陷盆

地。盆地长约 120 km, 宽 10~30 km, 面积约 1850 km²(图 1, 余心起等, 2005)。盆地基底因地区而异, 北部以晚古生代地层为主, 南部以早古生代浅变质岩系为主。盆地及周边地层发育, 除奥陶、志留系缺失外均有出露。盆地基底为震旦系—寒武系的巨厚地槽型复理石建造, 泥盆系为陆相-滨海相碎屑岩沉积, 石炭系为陆相含煤和碳酸盐建造, 二叠系为浅海相碎屑岩, 下三叠统以碳酸盐岩为主, 上三叠统为含火山碎屑的陆相沉积物, 侏罗系发育劣质煤层和火山碎屑岩。白垩纪地层在盆地内部主要出露厚达几千米的白垩纪陆相红色地层(卢秋芽, 1991)。盆内蕴藏着较丰富的石膏、石盐等重要矿产资源, 富锂矿赋存于上白垩统周田组(周敏娟等, 2017; 王春连等, 2020)。盆地的生成和发展受到盆缘断裂和控盆断裂的控制, 盆地界限由赣江大断裂、吉水大断裂、遂川大断裂、永新—峡江断裂以及遂川—兴深断裂构成(王春连等, 2020)。M2 井位于盆地中下部, 主要由下部的深灰色含碳粉砂质泥岩和上部的紫红色粉砂岩组成, 根据盆地内的地层对比, 下部可能为石炭系黄龙组, 上部应为上白垩统周田组。

2 样品采集和分析方法

本文共采集来自吉泰盆地 M2 钻孔黄龙组的黑色碳质泥岩样品 29 件。黄龙组的岩心以灰色和深灰色为主, 岩性以碳质泥岩或粉砂岩为主, 构造不发育(图 2)。通过岩心编录和野外照片, 可以观察到大量黄铁矿和脉状石英的存在。为了使实验结果更具代表性, 样品按照一定的间隔和岩性变化进行挑选(图 1)。同时, 为了不受次生沉积物的影响, 所挑选的样品均不存在脉石英。

本次实验采用 X 射线荧光光谱法(XRF)测定了 SiO₂、TiO₂、Al₂O₃、Fe₂O₃、FeO、MnO、MgO、CaO、Na₂O、K₂O、P₂O₅ 等主要元素的氧化物。采用电感耦合等离子体质谱仪(ICP-MS)测定痕量稀土元素的含量。用碱熔法将样品熔成玻璃片, 并在连续 X 射线荧光光谱仪(AB104L, Axios -mAX)上进行测试。采用电感耦合等离子体质谱仪(ELEMENT XR)测定溶出的微量元素和稀土元素。所有实验均在北京核工业地质研究院分析实验室进行。

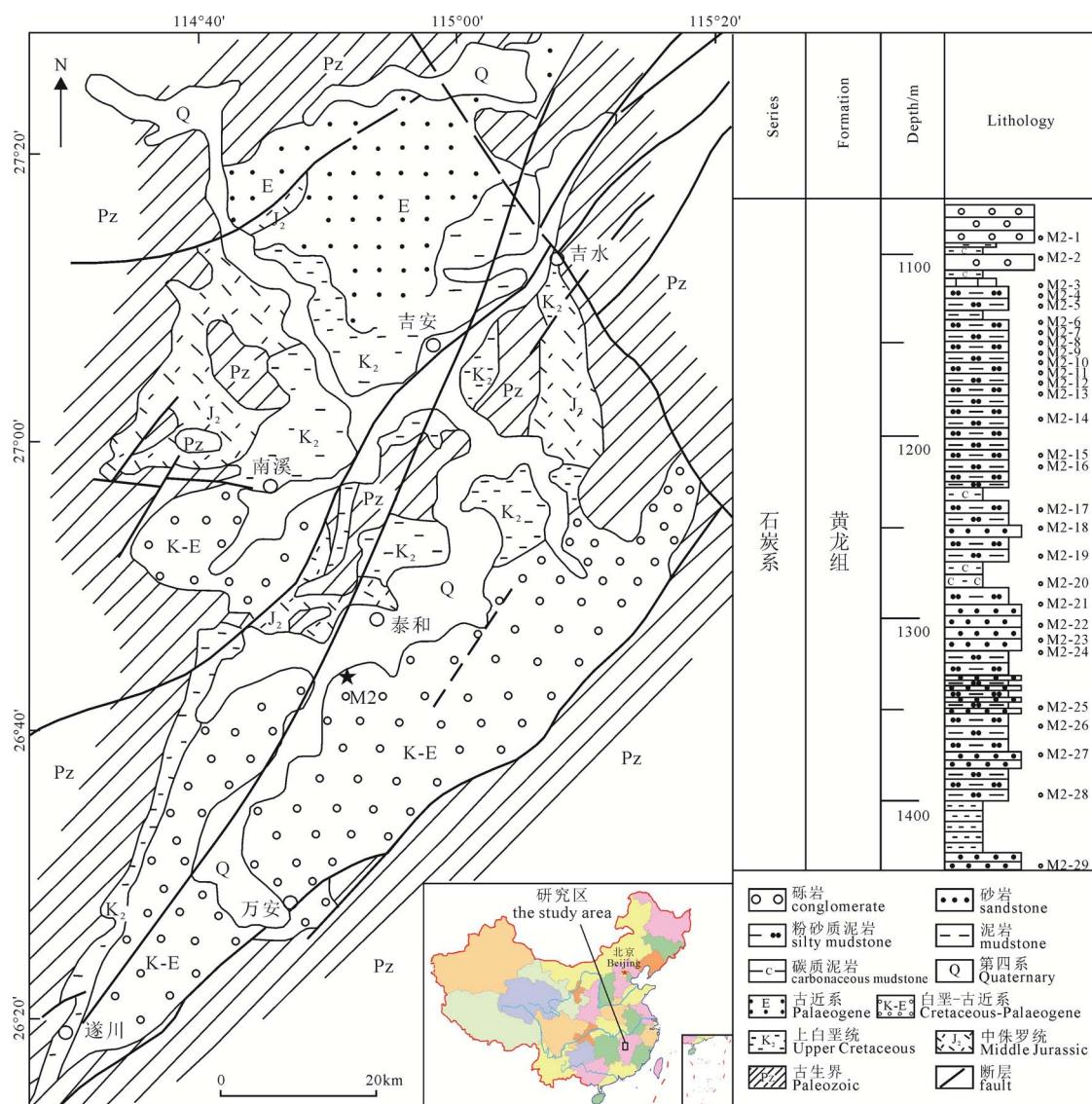


图 1 吉泰盆地地质构造简图(改自余心起等, 2005)

Fig. 1 Geological structure diagram of the Jitai Basin (modified from YU et al., 2005)

样品制备前, 用微钻除去外表风化面, 并将其研磨至 200 目。所有的地球化学分析样品都被粉碎并研磨到小于 200 目。XRF 的测试方法如下: 首先, 在镍锅中混合 0.4 g 粉末样品和无水 $\text{Li}_2\text{B}_4\text{O}_7$, 并用合适的氧化剂和浓度为 120 mg/mL 的 NH_4Br 液体稀释混合物。然后, 根据测试仪器的程序指示, 将镍罐放置在名为 CLAISIE 的取样机上, 之后得到高温液化的熔体, 并将其放在模具中, 冷却后, 装入密封的塑料袋中, 储存在干燥器中, 待测。ICP-MS 的测试方法如下: 首先, 称重约 2.5 g 的粉末样品, 将其放入耐高压烧杯中, 并加入溶液 HF-HNO_3 和烧杯的比例是 1:1 把搅拌均匀。在 80°C 下加热 24 小时蒸发液体。第二, 在溶液蒸发到接近干燥后, 分别加入 0.5 mL HClO_4 , 1.5 mL HF 和 1.5 mL HNO_3 。第三, 把盛有溶液的烧杯放进去烤箱 180°C 加热至少 48 小时, 以确保样品完全溶解。最后, 稀释溶液至 50 mL 并加入 1% 的 HNO_3 , 用于测量。

蚀变化学指标($\text{CIA} = \text{Al}_2\text{O}_3 / (\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} + \text{K}_2\text{O}) \times 100$)、风化化学指标($\text{CTW} = \text{Al}_2\text{O}_3 / (\text{Al}_2\text{O}_3 + \text{CaO} + \text{Na}_2\text{O}) \times 100$)、斜长石蚀变指标($\text{PIA} = (\text{Al}_2\text{O}_3 - \text{K}_2\text{O}) / (\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} - \text{K}_2\text{O}) \times 100$)以及成分变异指标($\text{ICV} = (\text{Fe}_2\text{O}_3 + \text{K}_2\text{O} + \text{Na}_2\text{O} + \text{CaO} + \text{MgO} + \text{TiO}_2) / \text{Al}_2\text{O}_3$), 反映了长石向黏土矿物的转化, 可用于追溯源区的风化历史(Nesbitt and Young, 1982; Harnois, 1988; Fedo et al., 1995; Cox et al., 1995)。 CaO^* (硅酸盐相中 CaO 含量) = $\text{CaO} - 10/3 \times \text{P}_2\text{O}_5$ (McLennan et al., 1993)。

3 分析结果

3.1 主量元素地球化学

主量元素的含量见表 1。在所有的测试样品中, SiO_2 是主要的氧化物, 其含量在 43.87%~86.46%, 平均为 64.02%。 Al_2O_3 (7.05%~31.12%, 平均 18.44%) 和 Fe_2O_3 (1%~26.81%) 含量次之。相比之下, CaO (0.094%~2.15%)、 MgO (0.323%~2.91%)、

Na_2O (0.085%~0.483%)、 K_2O (0.68%~5.88%)、 TiO_2 (0.199%~1.25%) 的含量占比偏少。含量最少的主量元素是 MnO (0.008%~0.099%) 和 P_2O_5 (0.022%~0.376%)。黄龙组样品具有低 $\text{Al}_2\text{O}_3/\text{SiO}_2$, 高 $\text{Al}_2\text{O}_3/\text{TiO}_2$, 高 $\text{K}_2\text{O}/\text{Na}_2\text{O}$ 的特征。

3.2 微量元素地球化学

所有测试样品的微量元素含量见表 2。微量元素含量的范围跨度较大, 在上地壳平均标准化图谱中

(图 3), 大部分元素的含量低于上地壳(UCC)的平均值。其中含量最高的微量元素是 Ba (134×10^{-6} ~ 712×10^{-6} , 平均 478.38×10^{-6}), 其次是 Zr (155×10^{-6} ~ 492×10^{-6} , 平均 295.28×10^{-6}); 含量最低的微量元素为 Ta (0.426×10^{-6} ~ 2.24×10^{-6} , 平均 1.44×10^{-6})。在上地壳标准化图解中, 样品具有 V 元素相对富集和 Sr 元素亏损的特征。部分微量元素与稀土元素的比值见表 4。

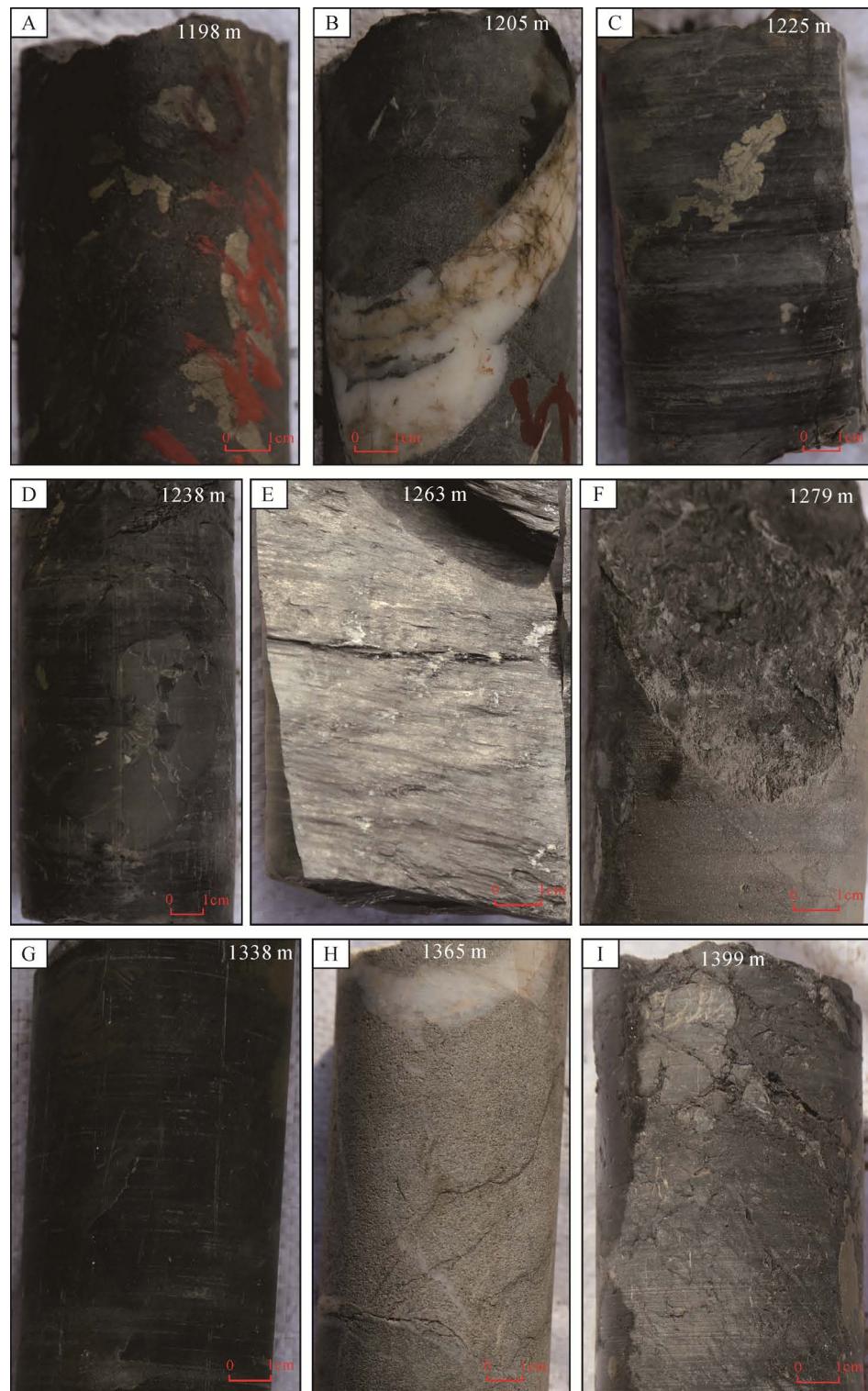


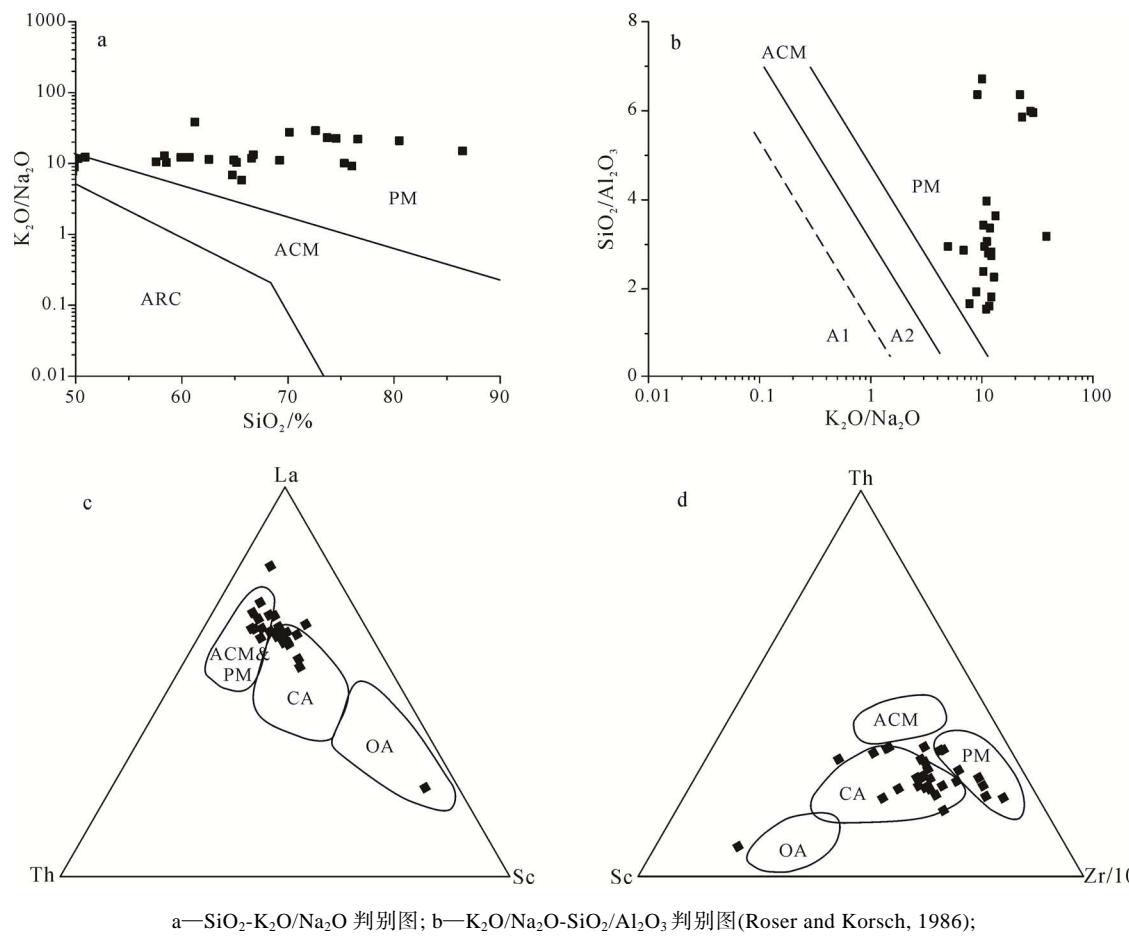
图 2 M2 井黄龙组部分岩心照片
Fig. 2 Some photographs of Huanglong Formation in Well M2

La-Th-Sc 和 Th-Sc-Zr/10 图解, 黄龙组的样品投点主要处于被动大陆边缘区域和大陆岛弧区域。综合微量元素、稀土元素的比值判别图, 吉泰盆地石炭系黄龙组碎屑岩的沉积构造背景以被动大陆边缘构造为主, 同时具有大陆岛弧构造的特征。舒良树等(2008)通过研究华南东段早古生代的沉积环境、火山岩、镁铁岩及早古生代造山带, 表明华南东段为被动大陆边缘浅海-半深海沉积环境。泥盆纪至三叠纪时期, 扬子板块为大西洋式被动大陆边缘; 从印支运动开始后, 逐渐由被动大陆边缘向主动大陆边缘演化(王鸿桢, 1986; 任纪舜, 1990)。在晚古生代时期, 华南陆块岩石学和沉积相特征表明, 由早期的陆内断陷演变成被动大陆边缘的陆表海环境(李双应和金福全, 1994; 吴大燮等, 1994)。舒良树等(2012)利用锆石 U-Pb 测年、沉积环境和地层对比等手段对华南地区构造演化的基本特征做了详细梳理, 认为在早古生代时期的板内构造事件使扬子块体和华夏块体再次聚合, 泥盆纪开始逐渐形成初步统一的中国南方岩相古地理格局。早石炭世—中三叠世时期, 整个华南基本处在一个稳定的

滨海-浅海环境没有出现大规模的火山活动及深海大洋的物质沉积。因此, 吉泰盆地石炭系黄龙组碳酸泥岩具有被动大陆边缘的构造背景特征。

4.3 源区古风化

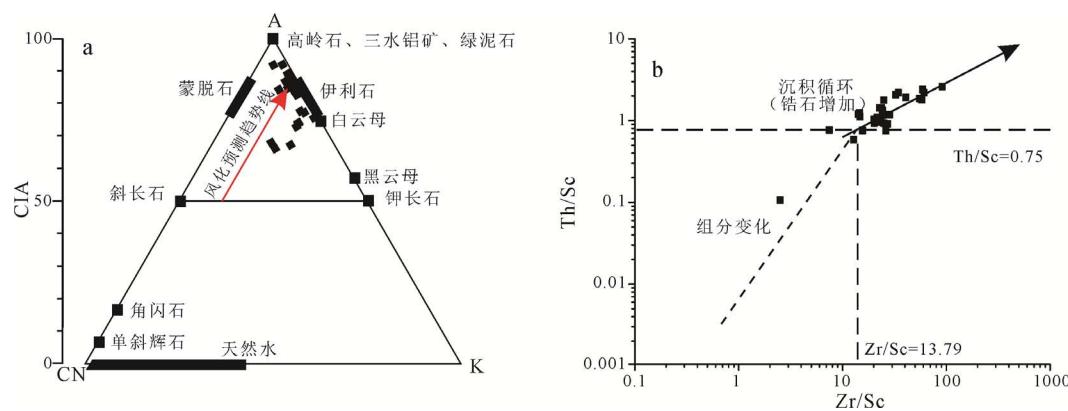
化学风化作用在潮湿气候下起主导作用, 强烈地控制着硅质碎屑岩沉积物的主微量元素组成(Nesbitt and Young, 1982; Harnois, 1988; Middelburg et al., 1988; McLennan et al., 1993; Fedo et al., 1995)。而在干旱条件下, 原岩普遍存在物理风化作用, 既原岩被机械分解呈较小的颗粒, 但是矿物学和化学成分并没有发生重大变化。因此, 化学风化指数能够为源区古风化提供很好的测量手段。利用 A-CN-K(由 Al_2O_3 、 CaO 、 Na_2O 和 K_2O 的分子比例分布和蚀变化学指标(CIA)构成)三元图可以确定化学风化和成岩过程中钾交代的元素迁移率(Nesbitt and Young, 1982, 1984)。此外, Harnois (1988)、Fedo et al.(1995)、Cox et al.(1995)利用 CIW、PIA、ICV 等化学风化指标评价源区的古风化作用, 同时避免了涉及钾富集的问题。



ARC—岛弧; ACM—活动大陆边缘; PM—被动大陆边缘; CA—大陆岛弧; OA—大洋岛弧; A1—岛弧背景; A2—演化岛弧背景。

a—diagram of $\text{SiO}_2\text{-}\text{K}_2\text{O}/\text{Na}_2\text{O}$; b—diagram of $\text{K}_2\text{O}/\text{Na}_2\text{O}\text{-}\text{SiO}_2/\text{Al}_2\text{O}_3$ (after Roser and Korsch, 1986); c—diagram of La-Th-Sc; d—diagram of Th-Sc-Zr/10 (after Bhatia and Crook, 1986); ARC—island arc; ACM—active continental margin; PM—passive margin; CA—continental island arc; OA—oceanic island arc; A1—arc setting; A2—evolved arc setting.

图 6 M2 井样品构造背景判别图
Fig. 6 Discriminant drawing of structural background, Well M2



a—A-CN-K 图解(Fedo et al., 1995); b—Zr/Sc-Th/Sc 图解(McLennan et al., 1993)。

a—diagram of A-CN-K (after Fedo et al., 1995); b—diagram of Zr/Sc-Th/Sc (after McLennan et al., 1993).

图 7 M2 井样品风化、分选和再循环判别图

Fig. 7 Comparative diagrams showing influence of weathering, sorting and recycling for M2

未风化的岩石和UCC的平均CIA值在50左右或以下(Taylor and McLennan, 1985),页岩CIA的值在70~75之间,黏土的CIA值接近100。黄龙组样品的CIA值在68.77~99.52之间,平均83.77,表明了较大的化学风化程度差异。其中较低的化学风化主要出现在钻孔的下部(表4),岩性较粗以粉砂岩为主,碳质含量较少,而上部的泥岩和粉砂质泥岩则表现出重度化学风化程度。在风化过程中,CaO和Na₂O相对于K₂O优先被去除,因此斜长石比钾长石更容易被风化。黄龙组样品经历化学风化后,CaO和Na₂O含量降低,投点大多处于A-K边界附近。而在沉积后的钾交代蚀变过程中,K相对于Al表现出被去除,从而使样品投点靠近A(Al₂O₃)顶点附近。钾长石和其他不稳定矿物的逐渐枯竭导致黑云母、白云母和伊利石矿物的增加。最终随着风化作用的加深,岩石组分将以高岭石、三水铝石和绿泥石富集为主。风化过程也可以由长英质岩石向蒙脱石转变,再转向白云母、黑云母及伊利石,最后转变为高岭石、三水铝石和绿泥石(图7a)。通过计算,黄龙组样品的CIW值为80.18~98.70,平均95.24;PIA的值为79.46~99.36,平均95.91(表4)。这些值都表明黄龙组沉积物有着强烈的化学风化作用(Harnois, 1988; Fedo et al., 1995),有利于元素的迁移和富集,为上覆卤水矿的形成提供了多样的成矿元素。对于ICV值而言,ICV值小于1,表明沉积物可能经受强风化作用的首次沉积物或经历了再沉积的产物。Th/Sc和Zr/Sc的比值暗示了沉积物的循环作用,导致锆石等重矿物逐渐富集,而锆石是Zr和Th的主要宿主(McLennan et al., 1993)。对比Zr/Sc-Th/Sc判别图(图7b),表明沉积物经历了再循环过程。地球化学特征表明黄龙组沉积物经历了较强的风化作用和再循环作用,而且上部风化作用更为强烈,气候也更为潮湿。

5 结论

(1)通过分析吉泰盆地M2井碳质泥岩样品La/Th-Hf和Zr-TiO₂的比值,轻稀土/重稀土比值和Eu的异常特征,认为吉泰盆地黄龙组的物源主要为长英质/基性岩和古老沉积物的混合区域,为富锂钾卤水矿提供了物质来源;

(2)通过主微量元素和稀土元素(TiO₂、La、Sc、Hf、Th、Zr)的含量和比值,反映出吉泰盆地黄龙组的碳质泥岩具有被动大陆边缘的构造背景特征;

(3)Th/Sc和Zr/Sc的比值表明吉泰盆地黄龙组上部泥岩受到沉积再循环的影响。根据化学风化指数(CIA、CIW、PIA和ICV)及Al₂O₃-(CaO+Na₂O)-K₂O图解,黄龙组碳质泥岩是在沉积源区经历了较为强烈的风化作用和再循环形成的沉积物。强烈的化学风化作用有利于元素的迁移和富集,为卤水矿床的形成提供了丰富的成矿元素。

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