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Li 同位素在矿床学中的应用：现状与展望

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摘要: 矿床的形成受制于多种复杂的地质作用, 包括全球尺度的板块构造运动、岩浆活动、变质沉积改造等过程, 并普遍伴随热液活动、流体迁移、水-岩相互作用、元素分异及同位素分馏等一系列局部区域地质和地球化学过程。在过去的矿床学研究中, 地球化学方法主要围绕在主、微量元素和传统的稳定同位素等手段, 解决了很多矿床成因问题。但仍存在不少的多解和难解问题, 比如许多矿床在矿化类型、蚀变分带与金属矿物组合方面具有诸多相似之处, 常规地球化学指标难以区分。随着测试精度的提高和自然储库组成的完善, Li 同位素近些年来已成为新兴的稳定同位素体系。Li 同位素在自然界过程中高达 80‰ 的同位素分馏使其具有更好的辨识能力, 同时兼有直接和间接指示作用, 有潜力成为研究各种复杂成矿过程的良好示踪剂。本文总结了近年来有关矿床学中 Li 同位素的研究和应用进展, 以俯冲带成矿为主, 阐述了斑岩型-热液矿床、伟晶岩型矿床和沉积矿床等类型矿床的 Li 同位素地球化学特征, 并探究新的 Li 同位素方法在矿床中的应用前景。基于 Li 同位素体系在各类矿床的应用实例, 我们认为 Li 同位素体系将为矿床学研究提供更多的指示信息和依据。

关键词: Li 同位素; 同位素分馏; 俯冲带; 成矿流体; 斑岩铜矿; 地球化学。

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Exploration of Li Isotope in Application of Ore Deposits

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Abstract: The formation of ore deposits is subject to varieties of complex geological processes, including plate tectonics, magmatic activities, metamorphic sedimentary transformations and other processes on global scale, and is generally accompanied by a series of geological and geochemical processes such as hydrothermal activities, fluid migration, fluid-rock interaction, elemental differentiation and isotopic fractionation. Over the past decades, studies of geochemical methods have focused on major, trace elements and traditional stable isotopes, that have addressed the genesis of ore deposits. However, there are still some difficulties and multi solutions. For instance, portions of ore deposits in the mineralization, alteration zoning, and metal mineral assemblage have many similarities, so the conventional geochemical indicators are difficult to be distinguished. With the improvement of analysis and the integrity of natural reservoir composition, Li isotope has become a new stable isotope system in recent years. The Li isotopic fractionation of up to 80‰ in the natural process enhances the identification ability of Li isotope, as well as in direct or indirect indicator function. Thus, Li isotopic systematics has the potential to be a good tracer to study various complex metallogenic processes. Here, this paper summarizes the studies and applications of Li isotopes on ore deposit in recent

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years, mainly in subduction zone mineralization, the geochemical behavior and characteristics of Li isotopes in porphyry-hydrothermal deposits, pegmatite deposits and sedimentary deposits, then it explores an application prospect of new Li isotopic method for ore deposits. Based on the applications of Li isotopic system in various ore deposits, it proposes that Li isotopic system will provide more indicative information for ore deposit study in the future.

Key words: Li isotopes; isotopic fractionation, subduction zone; ore-forming fluid; porphyry copper deposit; geochemistry.

0 引言

锂(Li)在自然界中属于微量元素,在地壳中的平均含量约为 25×10^{-6} ,是稳定同位素家族中较轻的元素和最轻的碱金属元素,Li 同时也是主要稀有金属的主要来源(汪齐连等,2006;Dill,2015).自然界富集 Li 的主要矿物有锂辉石、锂云母、磷锂铝石、透锂长石等.在工业上,Li 广泛应用于新型储能(如锂电池)、节能、产能(如核聚变发电)、军用以及民用领域(如心脏起搏器)等行业,是近年来新兴战略能源矿产,也是重要的“关键金属”(Linnen *et al.*, 2012;彭爱平,2012;刘丽君等,2017b;毛景文等,2019).自然界中的 Li 有两种稳定同位素, ^6Li 和 ^7Li ,其丰度分别为 7.5% 和 92.5%.Li 同位素间高达 16% 的相对质量差异导致地质过程中产生显著的 Li 同位素平衡分馏,目前已经观察到自然界不同地质储库间的 Li 同位素分馏可达 80%(Tomascak *et al.*, 2016),因此 Li 同位素成为研究各种地质过程的良好示踪剂,被广泛应用在宇宙事件(Chaussidon and Robert, 1998)、示踪流体来源(Bottomley *et al.*, 1999)、洋壳蚀变、板块活动以及海底热液活动等地球化学过程等研究(Chan *et al.*, 1992, 1999; Moriguti and Nakamura, 1998;雷吉江等,2008;Yang *et al.*, 2015; Dellinger *et al.*, 2017; Pogge von Strandmann *et al.*, 2017, 2020; Choi *et al.*, 2019; Wei *et al.*, 2020).

国际上通用的 Li 同位素组成表达方式为: $\delta^7\text{Li}(\text{‰}) = [({}^7\text{Li}/{}^6\text{Li})_{\text{样品}} / ({}^7\text{Li}/{}^6\text{Li})_{\text{标样}} - 1] \times 1000$.Li 同位素标样为美国国家标准技术研究院提供的纯化 Li_2CO_3 即 L-SVEC,其 ${}^7\text{Li}/{}^6\text{Li} = 12.039$ (Chan *et al.*, 1992)、12.019 (Decitre *et al.*, 2002) 和 12.119 (Kobayashi *et al.*, 2004).相比热电离质谱(TIMS)高精度测试法,新一代多接收电感耦合等离子体质谱仪(MC-ICP-MS)使 Li 同位素测试技术有了飞跃性的突破,测试精度从 TIMS 时代的约 2‰ 提升为现在的 0.2‰~0.3‰,分析效率也得到很大的提高(Millot *et al.*, 2004).

在过去 20 多年里地质储库的 Li 同位素地球化学研究取得了许多重要进展,不少学者总结了地球系统中自然界各类地质样品的 Li 含量和同位素组成范围(Tang *et al.*, 2007;万红琼等,2015;Tomascak *et al.*, 2016;Penniston-Dorland *et al.*, 2017).例如,大洋沉积物相对玄武质洋壳具有较高的 Li 含量,已报道的含量最高可达 80×10^{-6} (Bouman *et al.*, 2004; Leeman *et al.*, 2004),全球各地区的大洋沉积物总体 $\delta^7\text{Li}$ 值介于 $-4.3\text{‰} \sim +14.5\text{‰}$ (Chan and Hein, 2007);蚀变洋壳的 Li 含量为 $0.6 \times 10^{-6} \sim 21.8 \times 10^{-6}$ (Chan *et al.*, 2002), $\delta^7\text{Li}$ 值介于 $-1.6\text{‰} \sim +11.8\text{‰}$ (Chan and Edmond, 1988; Chan *et al.*, 1996; Verney-Carron *et al.*, 2015),而大陆地壳的 Li 含量介于 $8 \times 10^{-6} \sim 35 \times 10^{-6}$, $\delta^7\text{Li}$ 值介于 $0\text{‰} \sim +4\text{‰}$ (Teng *et al.*, 2004, 2008);此外,地幔橄榄岩的 Li 含量通常 $< 2 \times 10^{-6}$,尽管 Penniston-Dorland *et al.* (2017) 最新的统计地幔橄榄岩全岩 $\delta^7\text{Li}$ 值介于 $-9.7\text{‰} \sim +9.6\text{‰}$,但是地幔储库值目前仍存在分歧(Tomascak *et al.*, 2016),我国的学者研究则认为地幔橄榄岩的 $\delta^7\text{Li}$ 值最低可达 -30‰ (Tang *et al.*, 2007, 2010, 2012, 2014; Zhang *et al.*, 2010; Su *et al.*, 2012; Xiao *et al.*, 2015).近年来矿物间 Li 同位素分馏的相关实验研究结果对 Li 同位素组成的成因提供了重要信息(刘纯瑶等,2019),特别表现在对各个主要地质储库和一些重要地质过程的认识方面.上述工作大大促进了人们对不同地质背景下全球 Li 同位素体系的认识.

汇聚板块的俯冲带为全球矿产资源提供巨量的金属物质来源,是研究矿床和同位素地球化学的有利场所,很多非传统金属稳定同位素(Cu、Fe、Zn、Mg、Mo、K 等)在矿床上相继得到应用(Gagnevin *et al.*, 2012;王跃和朱祥坤,2012;Mathur *et al.*, 2013;王跃等,2014;Wu *et al.*, 2017;Zheng *et al.*, 2019;李东永等,2019;李伟强等,2020).相比 Cu、Fe、Zn、Mg 等同位素,Li 同位素较早被开发和应用.自早期以来 Li 同位素主要应用于示踪深部地质过程(Tomascak *et al.*, 1999, 2000; Seitz and Wood-

land, 2000; Chan and Frey, 2003; Benton *et al.*, 2004; Elliott *et al.*, 2004; Nishio *et al.*, 2004; Liu *et al.*, 2019; Tan *et al.*, 2020). 近些年来在低温表生地球化学过程有突出的进展, 例如示踪大陆风化与海洋环境、生物大灭绝重大地质历史事件等全球性科学问题 (Pistiner and Henderson, 2003; Kısakürek *et al.*, 2004; Dellinger *et al.*, 2017; 苟龙飞等, 2017; Pogge von Strandmann *et al.*, 2017, 2020; Hindshaw *et al.*, 2018; Sun *et al.*, 2018; Choi *et al.*, 2019; Caves Rungenstein *et al.*, 2019; Wei *et al.*, 2020). 然而, 在矿床学方面, Li 同位素的研究和应用相对比较少.

在矿床学研究过程中, 常需要解决最基本的问题包括: 成矿物质来源、成矿流体起源与演化和成矿作用时序等. 同位素地球化学方法发挥了很关键的作用. 长期以来, 国内外矿床地质学家普遍利用最传统的 Pb-S 同位素来示踪矿质来源 (廖文, 1984; 周维全和周全立, 1992; Ripley, 1999; Ripley *et al.*, 2002; Penniston-Dorland *et al.*, 2008; 梁婷等, 2008; 黎心远等, 2018), 利用 H 和 O 同位素来示踪流体的来源 (黄典豪等, 1984; 毛景文等, 2005; Klemm *et al.*, 2007; Hoefs, 2009; 马旺等, 2019); 利用赋矿围岩 (如沉积岩、岩浆岩) 和热液蚀变、伴生矿物的 Ar-Ar、Sm-Nd 和 Re-Os 同位素体系等来确定成矿年龄 (Foster *et al.*, 1996; 蒋少涌等, 2002; 魏俊浩等, 2003; 郭春丽等, 2007; 李随民等, 2014; 庞绪成等, 2019; Rotich *et al.*, 2020), 取得了很多好的找矿成果. 众所周知, 地球化学找矿指示剂中有直接

指标和间接指标之分, 上述的这些地球化学指标, 多数是矿床的直接指标. 然而, 在缺乏直接指标或同位素分馏特征不明显的情况下, 间接指标的指示意义是不可忽视的. 例如, Xiao *et al.* (2011) 在俯冲带的相关研究中, 发现 O 同位素和 Sr-Nd-Pb 同位素在密切接触的不同岩性之间未发生明显变化, 但 Li 同位素却表现出显著的分馏特征, 显示了 Li 同位素的突出优势. 过去 Li 同位素在应用于地球浅层流体运移和流体-岩石相互作用的追踪时, 尽管人们意识到 Li 同位素体系可能提供有关深部地壳流体的重要信息, 但至今这种稳定同位素系统仍未在流体有关的矿床中得到系统的开发应用 (Richard *et al.*, 2018), 原因可能是缺乏理论支撑和分析技术改进.

对矿床来说, 深部地壳流体的出溶和有用矿物的沉淀一脉相承. 在有热液流体参与的情况下, 元素和同位素都有可能相应地发生改变. Yang *et al.* (2015) 通过测试石英-流体包裹体的 Li 同位素和均一温度, 将 Li 同位素用于矿床成矿流体研究中, 获得了矿床中一批热液石英和流体包裹体的 Li 同位素组成数据, 初步建立了石英-流体 Li 同位素分馏经验公式: $\Delta\delta^7\text{Li}_{\text{Quartz-fluid}} = -8.9382 \times (1000/T) + 22.22$ (线性相关系数 $R^2 = 0.98$, 175~340 °C) (图 1), 并定量估算了成矿流体中岩浆水-海水混合比例. 最近的研究也发现 Li 同位素分馏与稀有金属花岗岩成矿作用的多样性之间存在一致性 (Li *et al.*, 2018), 暗示 Li 同位素、流体、热液矿床三者之间存在某种既间接又密切的关系.

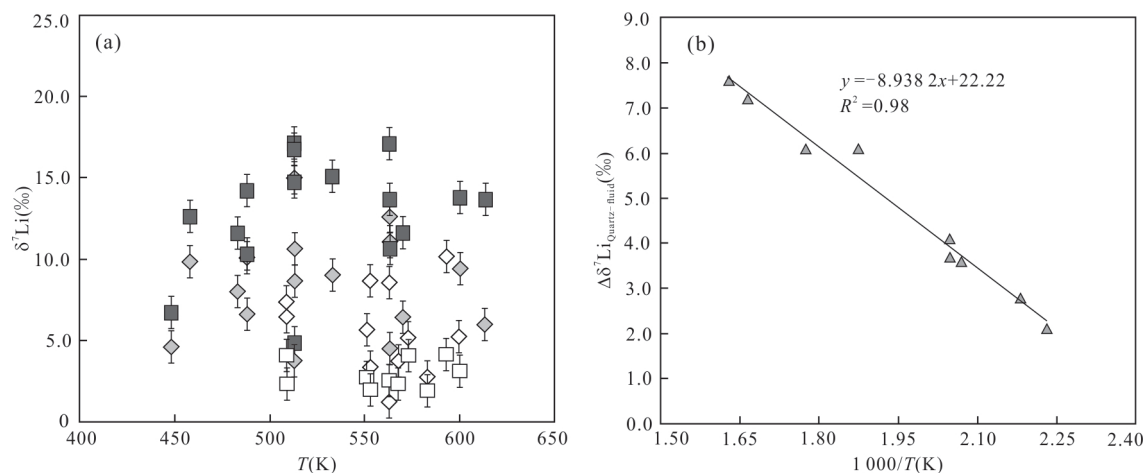


图 1 (a) 寄主石英中的 $\delta^7\text{Li}$ 变化和均一温度的流体包裹体; (b) 纯石英样品 (含原生流体包裹体) 中的 Li 同位素分馏系数 ($\Delta\delta^7\text{Li}_{\text{Quartz-fluid}}$) 与流体包裹体均一温度 ($1000/T$) 之间的关系 (据 Yang *et al.*, 2015)

Fig. 1 Variation of $\delta^7\text{Li}$ in the host quartz and fluid inclusions with the measured homogeneous temperatures (a); relationship of Li isotopic fractionation factor ($\Delta\delta^7\text{Li}_{\text{Quartz-fluid}}$) with homogeneous temperatures ($1000/T$) in fluid inclusions hosted from pure-quartz samples (b) (containing primary fluid inclusions) (modified from Yang *et al.*, 2015)

很显然,利用 Li 同位素在流体中显著分馏的特点来研究热液矿床是可行的.此外,研究成矿过程的 Li 同位素地球化学特征,通过二元混合计算,还能较为准确估算成矿物质(流体)通量.在某些地质成矿过程中 Li 同位素的分馏水平与传统的 C、H、O、S 等同位素体系相比, Li 同位素的精度和灵敏度更高、辨识能力更好.

1 Li 同位素示踪矿床过程的理论基础

在自然界, Li 作为微量元素以 +1 价阳离子形式广泛存在于各种矿物、熔体或地质流体中, 不受氧逸度变化的影响.同时, Li 作为最轻的碱金属元素, 具有高度化学活动性, 在大多数矿物中 Li 含量较低, 而在黑云母、堇青石、碱性长石、石英的 Li 含量可以高达几十到几百 mg/kg, 更是在一些特殊矿物中如锂辉石、透锂长石、锂云母中 Li 以主量元素形式存在(Vigier *et al.*, 2009); 其次, Li 作为中等不相容元素, 部分熔融过程中会优先富集于硅酸盐熔体中, 导致壳幔分异过程中 Li 在地壳($13 \times 10^{-6} \sim 35 \times 10^{-6}$)比地幔($1.2 \times 10^{-6} \sim 1.5 \times 10^{-6}$)相对富集(Teng *et al.*, 2008); 从超基性岩($2 \times 10^{-6} \sim 14 \times 10^{-6}$)到酸性岩($30 \times 10^{-6} \sim 55 \times 10^{-6}$) Li 含量也逐渐升高, Mg/Li 比值在岩浆分异过程中有规律地下降(图 2a).一方面, 随着分离结晶作用的持续进行, 岩浆中 H₂O、Li、F、B、Cl 和 P 等流体及挥发组分不断增加, 从而出现锂云母、锂辉石、透锂长石、萤石、黄玉、电气石和磷灰石等富 Li 特征矿物(Li 以氯化物或氟化物络合物迁移), 高度演化的花岗质岩浆可以结晶出锂电气石, 因而这种花岗岩又被称为 Li-F 花岗岩(朱金初等, 2002; Zhang *et al.*, 2008).特别重要的是, 在岩浆演化后期产生的气成-热液交代作用, 可使母岩中的 Li 发生迁移和富集, 以酸性的花岗伟晶岩中 Li 含量激增最为显著, 进一步富集形成有工业价值的 Li 矿床.另外, 大多数有色金属 Cu、Pb、Zn、Ag、W、Sn 等矿床和关键金属(稀土、稀有、稀散)矿床, 也都与酸性岩浆岩的演化和后期风化密切相关(陆一敢等, 2015).值得注意的是, Li 在蚀交代过程中与主要成矿元素有着大致耦合的变化规律, 可以作为重要的找矿标志.

从流体出溶到热液阶段, Li 同位素的地球化学行为是值得探究的问题.由于 Li 较强的流体活动性, 在洋壳蚀变和板块俯冲等与流体有关的过程中, ⁷Li 会优先富集于流体相中, 并伴随着显著的 Li

同位素分馏(Brenan *et al.*, 1998).有学者分析了西藏 Luozha 和 Dala 地区花岗岩(淡色花岗岩和二云母花岗岩), 发现 Li 的含量和 $\delta^7\text{Li}$ 值与稀土总量 ΣREE 有良好的相关协变(图 2b、2c); 同时在岩浆晚期的高度演化阶段(通常 $\text{SiO}_2 > 70\%$), SiO_2 和 ⁷Li 亦显示良好的相关性(图 2d; Tian *et al.*, 2017b).也有研究表明在相对开放的系统, 岩浆晚期流体出溶会导致 Li 同位素产生显著的分馏(Chen *et al.*, 2018; Fan *et al.*, 2020).经典理论推测, 在平衡状态下, 低配位数的晶体结构拥有更高的化学键能, 优先富集重的同位素(Brown, 2009).实验研究发现, 温度是控制 Li 同位素分馏的一个关键因素(Tomaschak *et al.*, 1999).Wunder *et al.*(2006) 通过实验测定了单斜辉石与含 Cl 和 OH 离子的水溶液之间的 Li 同位素分馏(实验条件: 压强 2.0 GPa、温度 500~900 °C).结果发现, ⁷Li 总是优先进入流体相.该研究表明, 随着温度的降低 Li 同位素的分馏程度增大, 这一结论与相对低温(通常 $P < 1.0 \text{ GPa}$, $T < 400 \text{ °C}$)系统中较大的 Li 同位素分馏现象相一致(Teng *et al.*, 2006a).此外, Li 有较快的扩散速率, 在矿物或熔体中 ⁶Li 比 ⁷Li 扩散快(Dohmen *et al.*, 2010).基于以上 Li 在流体中的地球化学行为, 矿床学者逐渐认识到 Li 同位素的应用可以推广到示踪矿床成因的研究中(Aulbach and Rudnick, 2009; Liu, 2017).

在绝大多数地质作用过程中, 流体都担任着元素迁移的载体、化学反应的活化剂的角色.大量研究表明, 岩石、矿物、元素以及同位素在有流体的情况下会表现出迥异的物理和化学性质(张文淮, 1984; Heinrich *et al.*, 2003; Rusk *et al.*, 2004; 孙贺和肖益林, 2009; Audétat and Li, 2017).深部巨量熔/流体在岩石圈/软流圈系统循环是金属元素活化、迁移的必要条件, 也是大型-超大型矿床形成的诱因, 可以说没有成矿流体就没有矿床, 这一认识得到矿床学家的一致认可(Audétat *et al.*, 1998; Hedenquist *et al.*, 1998; Klemm *et al.*, 2007; 池国祥和赖健清, 2009; 卢焕章, 2009; 孙贺和肖益林, 2009; 倪培等, 2014, 2018; 莫宣学, 2020b).在热液流体对同一源区萃取金属的过程中, Li 的重同位素先被淋滤出来, 晚期淋滤的流体相对早期淋滤的流体富集轻 Li 同位素.在各种不同背景成矿作用中, 流体的存在使得 Li 同位素发生很大的异常变化, 一旦这些异常变化与其他成矿指标构成线性相关, Li 同

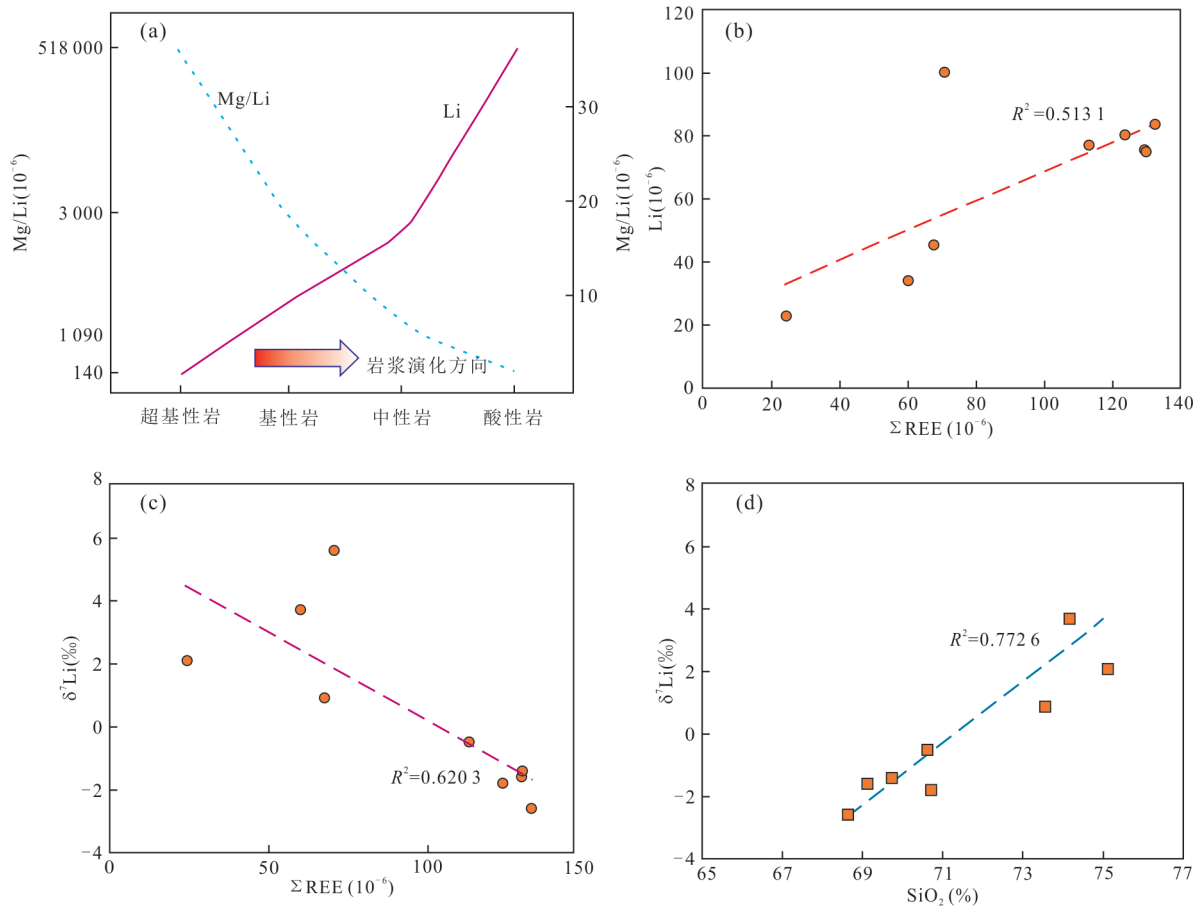


图 2 不同岩石类型中 Mg/Li 比值和 Li 含量变化(a); 西藏地区 Li & $\delta^7\text{Li}$ 和 ΣREE & SiO_2 协变示意(b~d)

Fig.2 Mg/Li ratios in different rock types (a); Li & $\delta^7\text{Li}$ vs. ΣREE & SiO_2 covariant diagram for granite in Tibet (b~d)
a. 据刘英俊(1987)修改; b~d. 据 Tian *et al.* (2017b)

位素就可以成为矿床找矿的标志。因而,以响应变化的流体为“纽带”,利用这种“间接指示”,我们可以尝试建立 Li 同位素和矿床之间的关系,形成了 Li 同位素在矿床上应用的理论支撑。

大多数金属矿床产于汇聚边界的俯冲或碰撞环境。世界三大成矿域如环太平洋成矿域、古亚洲成矿域、特提斯—喜马拉雅成矿域都位于俯冲—碰撞板块边界 (Cooke *et al.*, 2005; Hou *et al.*, 2009; Sillitoe, 2010; 侯增谦等, 2020; 莫宣学, 2020a)。在俯冲作用过程中,岩石由于脱水会在不同深度产生含有不同元素种类和不同量的流体或者熔体,这些富水流体、熔体或者超临界流体的产生和迁移会导致俯冲板片岩石本身发生元素分异和同位素分馏 (Spandler and Pirard, 2013; Wang *et al.*, 2020; 肖益林等, 2020), 而流体活动性元素 (如 B、Li、As、Sb、Pb、Cs、U、Sr 和 Ba 等) 变化尤其显著 (肖益林等, 2015)。同时,俯冲板片产生的流体或熔体会交代上覆的地幔楔,使上覆的地幔楔岩石发生地球化学特

征的改变,发生部分熔融,从而导致岛弧岩浆作用。而正是这些普遍存在的熔流体迁移和交代,以及岛弧岩浆作用等,使得俯冲带成为元素的迁移和再分配的大型工厂,也使得金属元素的迁移和富集成为可能,并最终形成俯冲带范围内广泛发育的金属矿床 (Candela and Piccoli, 2005; Richards, 2009; Sun *et al.*, 2015; Zhang and Audétat, 2017; Wang and Xiao, 2018)。俯冲边界所形成的弧盆体系可以发育不同种类的矿床,如斑岩型铜矿、浅成低温型金矿、VMS 型矿床等 (Sillitoe, 2010; 陈华勇和肖兵, 2014; 陈华勇和吴超, 2020; 姚书振等, 2020), 这些矿床通常与流体—热液活动作用关系密切,利用 Li 同位素示踪俯冲带的成矿将是矿床研究新的方向。

已有研究表明, Li 同位素在示踪板块俯冲及壳幔物质循环过程中发挥了重要作用,与俯冲带相关的矿床将是 Li 同位素理论和实践的主要场所 (Moriguti and Nakamura, 1998; Elliott *et al.*, 2006; Penniston-Dorland *et al.*, 2012; Tian *et al.*, 2019)。万

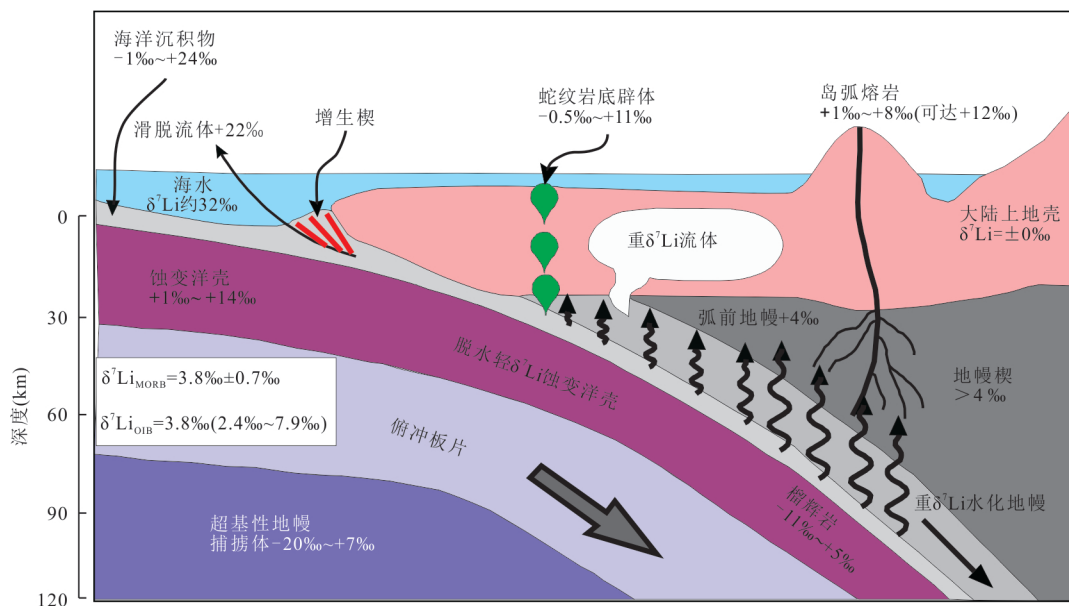


图3 俯冲带的Li同位素体系

Fig.3 Schematic illustration of Li isotope systematics in subduction-zone

据 Tang *et al.* (2007, 2010); 汤艳杰等(2009)修改

红琼等(2015)在前人研究的基础上,进一步详细总结了俯冲带Li同位素体系地球化学行为特征,并完善了Li同位素储库(图3)。以上这些理论基础和先导工作,将为示踪矿床提供一个重要的潜在地球化学指标和依据。

2 Li同位素在俯冲带斑岩型矿床研究中的应用

斑岩铜多金属矿床是与俯冲带相关的最显著和最典型的金属矿床(Sillitoe, 1972; Cooke *et al.*, 2005; 侯增谦等, 2007, 2012, 2020; Zheng *et al.*, 2012)。尽管有上百年的研究历史,斑岩型矿床的研究仍然是一项庞大的系统工程,迄今仍然存在不少问题,传统的岩石化学、微量元素划分、识别原岩的源区和金属矿质的来源问题尚存争议(陈华勇和吴超, 2020)。由于铜是硫化物的基本组成元素,所以理论上Cu同位素示踪硫化物熔体如何产生、迁移、定位和富集是首选。但前人将Cu同位素应用于西藏碰撞环境下形成的冈底斯斑岩铜矿床,探讨成矿物质来源、流体和铜初始富集的关系时,发现Cu同位素在高温条件下可能发生分馏(Wu *et al.*, 2017; Zheng *et al.*, 2019),而且后期叠加干扰的因素多,因此作为矿床直接指标的Cu同位素并不能完全解决问题。

与Cu同位素相比,Li同位素与斑岩矿床并非直接相关。Tian *et al.* (2017a)测得西藏南部的新生下地壳Li含量介于 $7.1 \times 10^{-6} \sim 37.2 \times 10^{-6}$,均值为 15.4×10^{-6} ; $\delta^7\text{Li}$ 值介于 $+0.8\text{‰} \sim +6.6\text{‰}$;结合主微量元素和Sr-Nd-Pb及Hf等同位素,认为在地幔熔融和玄武岩分馏结晶、岩浆分异和地壳深熔过程中,Li同位素几乎不发生明显分馏。围岩蚀变和低级变质作用对样品元素分布和Li同位素组成的影响似乎也不显著(Teng *et al.*, 2008),因此可以示踪源区(Tian *et al.*, 2017a, 2018;图4)。由于受到印度上地壳俯冲产生的富 ^7Li 流体的交代, $\delta^7\text{Li}$ 高于大陆地壳的正常值(约 $+1.4\text{‰}$),而新生下地壳是由大陆岩石圈地幔交代拉萨下地壳形成的,并叠加早期特提斯洋俯冲物质,可能是控制藏南部斑岩型铜多金属成矿的关键因素(芮宗瑶等, 2004)。由此可看出,Li同位素与其他体系手段结合,将是示踪源区的重要手段。

已有较多的研究表明,深熔和早期的岩浆分异对Li同位素没有显著分馏(Tomascak *et al.*, 1999; Schuessler *et al.*, 2009; Teng *et al.*, 2009),但关于这个问题部分学者认为目前并没有一致的认识,还需要更多的数据和个例解剖才能深入了解这些过程的Li同位素行为,且将来需要更多的研究。由于含矿斑岩体一般是由中酸性花岗闪长质岩浆进一步演化形成,要经历一系列高温部分熔融、结晶分异

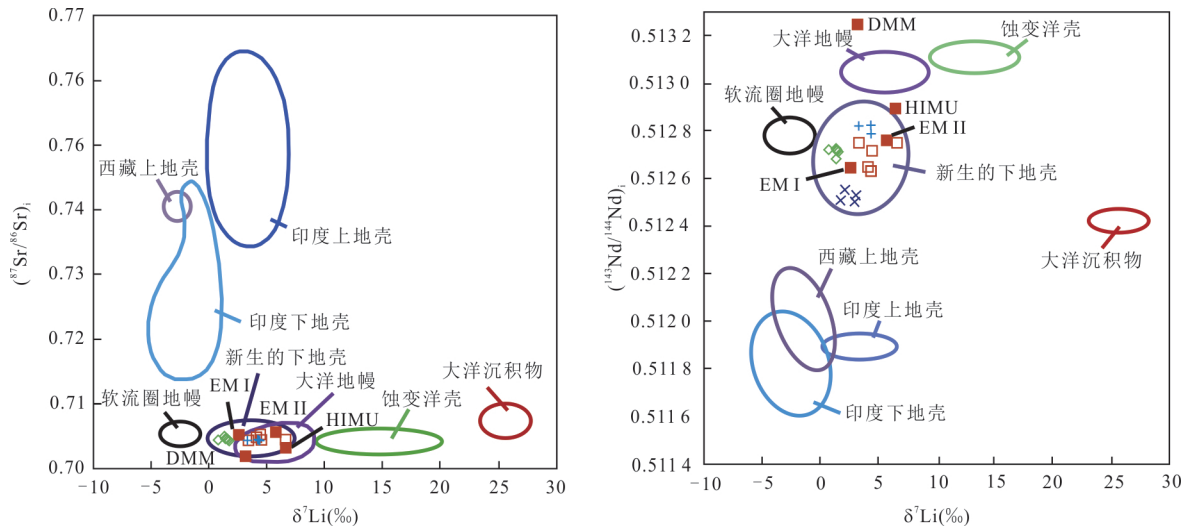


图 4 (a) $(^{87}\text{Sr}/^{86}\text{Sr})_i$ vs. $\delta^7\text{Li}$ 和 (b) $(^{143}\text{Nd}/^{144}\text{Nd})_i$ vs. $\delta^7\text{Li}$ 相关投影

Fig. 4 Plotting diagrams of $(^{87}\text{Sr}/^{86}\text{Sr})_i$ vs. $\delta^7\text{Li}$ (a) and $(^{143}\text{Nd}/^{144}\text{Nd})_i$ vs. $\delta^7\text{Li}$ (b)

其中安山岩来自典中组,玄武岩来自叶巴组,辉长岩和闪长岩来自冈底斯岩基.地幔端元数据来自 Krienitz *et al.* (2012); Sr-Nd- $\delta^7\text{Li}$ 同位素组成的底图据 Tian *et al.* (2018)

等过程,部分学者研究认为部分熔融会导致 Li 同位素分馏 (Su *et al.*, 2012; Sun *et al.*, 2016), 因为部分熔融过程中可能残留的石榴子石中 ^6Li 优先占据八配位体位置, 因此残留的石榴子石具有低的 $\delta^7\text{Li}$ 值, 从而形成具有高 $\delta^7\text{Li}$ 值的花岗质熔体. Chen *et al.* (2018) 则发现在岩浆侵位过程中, 熔融与外部流体强烈相互作用导致花岗岩中稀土四分组效应, 同时也造成 $\delta^7\text{Li}$ 值升高. 无论哪种情况, $\delta^7\text{Li}$ 值变化的异常无疑有利于矿床勘探. 此外, Li、Rb、Cs、Cl、P 和 F 等流体组分的地球化学特征往往表征岩浆岩分异演化的程度, 高演化岩浆通常是大型矿床成矿的关键. 同时 Li 作为矿化剂, 在斑岩型系列矿床成矿过程, 利用相关指标协同 Li 同位素理论上对示踪成矿物质来源和成矿过程是可行的. 受前人工作启发, Lu *et al.*, (2021) 最新尝试将 Li 同位素应用于探讨西藏冈底斯斑岩铜矿床的成矿构造背景.

3 Li 同位素在含锂矿床研究中的应用: 以(花岗)伟晶岩型为例

随着新兴产业的快速发展, 锂矿资源的需求越来越大. 尽管世界上最大的锂矿是盐湖或卤水型锂矿, 但一方面卤水提锂的技术还需要不断完善, 同时 Li 同位素在卤水型锂矿床上的应用较晚. He *et al.* (2020a) 对藏北柴达木盆地的卤水型锂矿床的样品(卤水湖、晶间卤水、河水、雨水、温泉水、泉水

和原岩) 进行 Li 同位素分析, 发现其 $\delta^7\text{Li}$ 值在 $+4.3\text{‰} \sim +40.7\text{‰}$ 之间变化, 而且不同成因类型的样品变化范围不同; 研究者认为柴达木盆地卤水湖泊中 Li 的可能来源是早期富盐沉积物重溶锂的多次混合, 表明 Li 同位素可以作为一种有效的示踪剂来揭示卤水型 Li 的来源和重构 Li 在卤水中的富集过程.

相对卤水型锂矿而言, “硬岩型” 锂矿资源虽然比例小, 但由于成矿背景复杂多样, 矿床品位高且开采技术成熟, 因此对于 Li 资源开发的意义重大 (许志琴等, 2018). 国外针对伟晶岩矿床开展一些 Li 同位素分馏机理(平衡分馏、扩散动力分馏、混合或瑞利分馏等) 研究, 为解决岩浆源区性质方面提供证据, 但对于形成伟晶岩的流体是否与花岗原岩一致存在较大争议 (Teng *et al.*, 2006b; Barnes *et al.*, 2012; Deveaud *et al.*, 2015). (花岗) 伟晶岩体系中发生显著的 Li 同位素分馏在于 Li 与各个共存相(矿物、熔体、含水流体) 间的交换机制 (Wenger and Armbruster, 1991). 前已述及, 高温深熔下的锂同位素分馏是可以忽略不计, 岩浆分异过程中的超临界流体出溶以及封闭岩浆热液系统中的水岩反应均不会产生有意义的 Li 同位素分馏 (Tomascak *et al.*, 1999; Teng *et al.*, 2006b; Li *et al.*, 2018). 而花岗岩是在低温 ($650 \sim 850\text{ }^\circ\text{C}$) 条件下, 地壳物质经贫水熔融形成的, 期间伴随的分离结晶、气相出溶和缓慢冷却等过程都可能产生较大的锂同位素分馏

(Teng *et al.*, 2006a). Gordienko *et al.* (2007) 研究发现伟晶岩中锂辉石的 $\delta^7\text{Li}$ 值变化范围很小 ($+0.2\text{‰}\sim+2.4\text{‰}$); Maloney *et al.* (2008) 研究也表明伟晶岩各分带中的电气石样品同样缺乏明显的同位素分馏, 因此认为 Li 同位素并不能很好地解释花岗伟晶岩内部的结晶分异过程. 然而, Magna *et al.* (2013) 观测发现来自不同伟晶岩体的同一种矿物产生了高达 20‰ 的 Li 同位素分馏, 可能受到了复杂的流体-熔体-固体之间的分馏作用或其他因素(如古地理条件)的影响, 这种情况尤其发生在高度演化体系的晚阶段, 因此他们认为并不能简单建立伟晶岩与其可能源区之间的关联.

国内近几年在硬岩型的锂辉石矿床方面开展不少工作. 刘丽君等(2017a)在对亚洲最大的固体锂辉石矿床四川甲基卡伟晶岩型锂多金属矿的研究中发现, 锂辉石和二云母花岗岩的 Li 同位素组成在误差范围内具有非常好的一致性(表 1), 据此认为锂辉石来源于二云母花岗岩, 这一结论与前人在包裹体、同位素地球化学和年代学方面的证据相一致. 侯江龙等(2018)最近进一步对四川甲基卡锂矿床花岗岩体开展 Li 同位素地球化学研究, 探讨矿区稀有金属的来源与演化过程, 研究结果显示岩体 Li 含量介于 $192\times 10^{-6}\sim 470\times 10^{-6}$, $\delta^7\text{Li}$ 值介于 $-1.6\text{‰}\sim+0.9\text{‰}$, 均值为 -0.2‰ , 与平均上地壳值基本一致, 具有高 Li 含量低 $\delta^7\text{Li}$ 的特征. 研究还发现 $\delta^7\text{Li}$ 与 Li、Rb、Ga、 SiO_2 及 $\epsilon_{\text{Nd}}(t)$ 不存在明显的相关性, 因此岩体 Li 同位素组成不受岩浆结晶分异作用和围岩蚀变作用的影响, 可反映其形成时的源区特征(图 5), 证明了伟晶岩的成矿流体来源于二云母花岗岩.

最近 Fan *et al.* (2020) 发现西昆仑白龙山含锂伟晶岩矿床中随着花岗岩浆演化到晚期阶段, 熔体分离会导致贫 Li 伟晶岩 ($\delta^7\text{Li} = +2.3\text{‰}\sim+4.9\text{‰}$) 与富 Li 伟晶岩 ($\delta^7\text{Li} = -1.9\text{‰}\sim+0.4\text{‰}$) 具有较大差异的 Li 同位素组成. 此外, Chen *et al.* (2020) 根据新疆阿尔泰造山带中贫 Li 伟晶岩 ($\delta^7\text{Li} = +4.1\text{‰}\sim+14.5\text{‰}$) 和富 Li 伟晶岩 ($\delta^7\text{Li} = -1.0\text{‰}\sim+10\text{‰}$) 的同位素特征, 提出了含锂矿化伟晶岩并非传统认为的花岗质岩浆极端分异的产物, 而是角闪岩相条件下含白云片岩脱水低程度部分熔融的一种新成因. 综上所述, 尽管 Li 同位素应用于(花

表 1 四川甲基卡伟晶岩型锂多金属矿床锂辉石和黑云母质量分数和同位素组成

Table 1 Lithium concentrations and isotope compositions of spodumenes and biotites from the Jiajika lithium polymetallic deposit, Sichuan Province

样号	样品描述	Li(10^{-6})	$\delta^7\text{Li}(\text{‰})$
308	伟晶岩中锂辉石	34.3	-0.4
134-4	伟晶岩中锂辉石	33.6	-0.6
JY-4	二云母花岗岩中黑云母	7.4	+1.6

注: 数据引自刘丽君等(2017a).

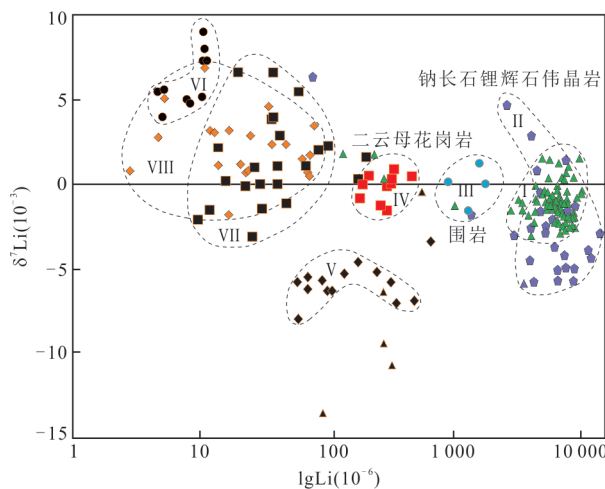


图 5 花岗岩和伟晶岩 $\delta^7\text{Li}$ 和 $\lg\text{Li}$ 的相关图

Fig.5 Relationship of $\delta^7\text{Li}$ vs. $\lg\text{Li}$ between granite and pegmatite

I. 甲基卡钠长石锂辉石伟晶岩; II. 加拿大小纳汉尼伟晶岩群 (Barnes *et al.*, 2012); III. 甲基卡伟晶岩脉围岩 (刘丽君等, 2017a); IV. 甲基卡二云母花岗岩; V. 厄尔士山花岗岩 (Romer *et al.*, 2014); VI. 荆山淡色花岗岩 (Sun *et al.*, 2016); VII. 布拉克山哈尼峰花岗岩 (Teng *et al.*, 2006a); VIII. 中国东北 A 型花岗岩 (Teng *et al.*, 2009); 据侯江龙等(2018)

岗岩)伟晶岩来源问题还不够成熟, 但诸如不同类型多阶段脉体的 Li 同位素与 Li 同位素组合可用来区分锂矿床母岩成因类型, 同时对稀有金属 (Li、Rb、Cs、Be、Nb、Ta 等) 找矿具有一定的指示意义, 尤其在含锂矿床中, Li 同位素可作为直接指标进行矿床成因示踪, 显现出其未来在该类型矿床研究中更为突出的应用前景(如李贤芳等, 2019).

4 Li 同位素在常见热液型矿床研究中的应用

热液矿床是一个广义的矿床类型, 这种类型矿

床更加强元素迁移、富集、沉淀卸载和流体的参与过程. 热液矿床的成矿环境可以是俯冲带的岛弧,也可以是弧后盆地、造山带或其他板块内部等;热液来源可为岩浆、沉积和变质成因,相比俯冲带的斑岩型矿床(广义上属于岩浆热液矿床),一般说来热液矿床更为复杂和类型多样.

早期有学者对海洋热液系统中的沉积物进行研究(Decitre *et al.*, 2004; Chan *et al.*, 2006; Chan and Hein, 2007),认为年轻的洋壳由于有较厚的沉积层覆盖,流体的组分可能因与沉积柱内沉积物的反应、金属硫化物的沉淀以及潜在的大型块状硫化物矿床的形成而改变,这种热液沉积物类似于现代日本黑矿,或是喷流沉积矿床(SEDEX)的前身.研究者对卡斯凯迪亚海槽、东加海沟、瓜伊马斯海盆、中河谷区等热液活动区的热液活动区对比中发现其Li含量介于 $4.8 \times 10^{-6} \sim 76.0 \times 10^{-6}$, $\delta^7\text{Li}$ 值介于 $-4.31\% \sim +9.36\%$,慢速堆积的多金属沉积物常表现出较高的 $\delta^7\text{Li}$ 值($+6\% \sim +10\%$),表明有海水的参与;再同时根据Li在流体和沉积物中的分异系数就可计算出孔隙水中的理论Li含量,结合喷口流体的 $\delta^7\text{Li}$ 还可以计算出理论上与沉积物发生平衡的流体量.计算值与实际值的差别越大,说明流体喷出越多,垂直上升的流体流速以及流体—沉积物的比率就可能越大.中河谷热液活动区的流体是沿着破碎带或断裂上升,因此很容易形成大型的热液硫化物矿床(Goodfellow and Franklin, 1993),而伊斯坎布海槽热液活动区的流体为分散上升,因而较难形成大的硫化物矿床(Gieskes *et al.*, 2002).由此可见,沉积物中Li的含量及同位素的信息能为判断热液成矿的规模提供重要证据.

周汀等(2013)对金沙江—红河富碱侵入岩带不含矿的剑川北岩体和含矿的万洞山岩体进行了Li同位素地球化学研究,结果显示,剑川北岩体和万洞山岩体的 $\delta^7\text{Li}$ 组成分别为 $+0.3\% \sim +6.2\%$ 和 $-6.5\% \sim +0.1\%$,含矿与不含矿岩体的 $\delta^7\text{Li}$ 值存在明显差异,指示含矿岩体的岩浆源区可能遭受过较大程度的板片脱水所释放的具有较低 $\delta^7\text{Li}$ 值的流体交代作用,因此通过Li同位素可以很好地区分含矿与不含矿岩体.Yang *et al.*(2015)针对成矿流体来源问题,选择我国最典型的VMS矿床——四川呷村大型铅锌铜多金属矿床开展了热液石英和流体包裹体的Li-O同位素地球化学研究,获得热液石英及其包裹体的Li同位素组成,估算并限定了岩浆

水/海水对成矿流体的相对贡献,揭示了流体演化与VMS成矿的关系,提供了岩浆水大量参与成矿的Li同位素新证据,这是Li同位素在精细地球化学方面示踪岩浆流体的新突破.Li *et al.*(2018)对中国南方雅山地区和西华山地区的含铋、钽、钨的锂富稀土花岗岩矿床进行研究,发现Li含量变化较大的雅山花岗岩($88.7 \times 10^{-6} \sim 7430.0 \times 10^{-6}$)的 $\delta^7\text{Li}$ 值变化范围较窄($-1.5\% \sim +1.5\%$);而Li含量变化较小的西华山花岗岩($37.8 \times 10^{-6} \sim 209.0 \times 10^{-6}$)具有较大的 $\delta^7\text{Li}$ 变化特征($-0.2\% \sim +4.4\%$),推测这可能与花岗岩岩浆分异和水—岩反应及岩浆期后的热液过程中的Li同位素分馏及稀有金属花岗岩石成矿作用有关.此外,在高度演化的岩浆系统中(如钨矿化),流体—岩石相互作用对Li同位素分馏的影响要比岩浆分异作用大得多.最近Xiang *et al.*(2020)发现演化的花岗岩在岩浆—热液转换过程中电气石的Li同位素发生较大分馏,该研究为矿物尺度Li同位素应用于热液矿床提供新的约束.

在热液矿床中,Li多数情况未必一定是成矿元素或伴生元素,但Li同位素可作为间接指标示踪矿床成因,体现了该同位素体系新的优势.当然,热液矿床形成过程尤为复杂,往往单一的研究手段和方法很难清晰解剖,需要其他有效方法的联合示踪.

5 Li同位素在沉积矿床研究中的应用

外生矿床由于成矿物质来源广泛,成矿过程也相对复杂,因此目前为止相关的Li同位素研究较少.Chan *et al.*(2007)对全球海洋锰铁矿床的Li同位素研究发现,在Fe-Mn矿床中水成型结壳的Li含量为 $0.6 \times 10^{-6} \sim 6.0 \times 10^{-6}$,热液型结壳的Li含量为 $148 \times 10^{-6} \sim 1188 \times 10^{-6}$,而水成—热液和水成—成岩型结壳的Li的含量为 $2.4 \times 10^{-6} \sim 64.0 \times 10^{-6}$.通过分析锰铁矿床表层吸附物和矿物晶格内吸附物的 $\delta^7\text{Li}$ 值,研究得出结论:(1)相对于水成结壳,Li主要赋存于热液矿床中,通过Li含量可以提供相应矿床的来源信息;(2)锰铁矿床中Li主要是以水合离子形式 $\text{Li}(\text{H}_2\text{O})^{++}$ 赋存于 MnO_2 表面,结壳中Mn和Fe的绝对和相对含量可以控制矿床中Li的含量;(3)Fe-Mn结壳原始的同位素特征经过沉积后与海水发生交换而完全改变,因此结壳中的 $\delta^7\text{Li}$ 值不能作为判断海水Li同位素组成长期变化的证据;(4)Mn-Fe氧化物—氢氧化物晶格内层在吸附Li形成络合物的过程中优先吸附 ^6Li .研究者认为,这种

以机械沉积改造过程为主的沉积矿床体系中, Li 同位素地球化学行为主要是受吸附作用主导, Li 同位素的分馏机制也千差万别(Wimpenny *et al.*, 2015; Zhang *et al.*, 2021)。但是总体而言, Li 同位素在沉积矿床的应用还需要深入和具体的研究。

6 Li 同位素在其他类型矿床研究中的应用

除了主要应用于金属矿床之外, Li 同位素在能源矿藏方面的应用也引起一些学者们的注意。吴泽鹏等(2018)利用 Li 和 B 同位素对页岩油气运移进行示踪研究, 认为 Li 和 B 与有机物相关, 因此可以追踪与烃类相关流体的生成和迁移。由于干酪根中存在 Li 和 B 元素, Li 和 B 存在于干酪根中最显著的证据是它们在油田盐水中具有高浓度, 其中 Li 达到 10~100 mg/L, B 达到 700 mg/L, 比海水和地下水高好几个数量级。在干酪根热成熟生成油气过程中 Li 和 B 元素被释放进入流体, 并被成岩伊利石所吸收。因此, 具有这些同位素特征的伊利石的年龄记录了油气运移的时间。最近几年在中国、俄罗斯、印度等煤矿中发现了高含量的伴生锂元素(Sun *et al.*, 2012, 2016; He *et al.*, 2020b), 随后对煤层的 Li 同位素研究认为 Li 的同位素分馏与煤成分中的黏土矿物有较大关系(He *et al.*, 2020)。此外, Harkness *et al.*(2015)分析了来自美国若干个盆地的煤和煤渣样品, 认为 Li 联合 B 同位素可以更有效示踪煤、石油和天然气燃烧带来的污染排放。由此可见, Li 同位素不仅在金属矿床上应用, 在能源矿藏和能源利用过程中产生的环境效应也值得探索。

7 总结和展望

综上所述, 随着高精度高灵敏度测试技术的发展和各类地质储库的建立, Li 同位素地球化学研究成为一个崭新而快速发展的领域, 参与探讨矿床学中的物质来源、流体出溶、流体演化、热液蚀变等关键成矿过程。诚然, 由于 Li 同位素组成变化与样品之间存在某种相关, 控制 Li 同位素变化规律的因素还有待深入研究。但基于变化规律, 通过对比已知的 Li 同位素组成来寻找与成矿有关的岩体, 进而探寻新的矿体具有很大的潜力。Li 同位素在矿床中的应用是同位素地球化学与矿床地质学科交叉融合的体现, 值得深入探究。相比直接指标对矿床的“源

区控矿”示踪, Li 同位素更侧重于“条件控矿”, 这是一条新的思路。基于前人的工作, 我们对 Li 同位素的地球化学行为和分馏机制有了基本认识。然而, 对于一些成矿过程中 Li 同位素的特征及相关地质意义尚缺乏统一的认识和解释, 单一的技术手段也不可能彻底解决复杂的矿床问题。

展望今后 Li 同位素在矿床的应用, 笔者认为包括以下几个方面: (1) 系统地开展不同类型矿床的 Li 同位素地球化学研究, 完善 Li 同位素的应用理论与积累各类地质储库数据, 建立具有普适性的 Li 同位素体系和技术方法; (2) 以流体为连接 Li 同位素和矿床的“纽带”, 观测各种条件下流体对 Li 同位素组成的影响, 关注“条件”引起的同位素分馏, 尤其重视引起 Li 同位素分馏的关键因素; (3) 代表最直接成矿初始物质的熔流体包裹体的 Li 同位素是个新的研究方向; (4) 探究 Li 同位素与其他同位素体系相结合的集成效果, 将有望突破以往研究瓶颈, 实现对矿床研究更高效的约束。可以预见, Li 同位素将成为矿床学研究中一种重要的地球化学手段。

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