

矿床地球化学国家重点实验室近十年 成矿示踪技术方法若干进展

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摘 要: 成矿示踪技术方法是矿床学研究的基础, 在深入揭示成矿物质来源、活化、迁移和富集成矿等方面发挥着至关重要的作用。近年来关键矿产成为研究的重点, 由于此类矿产通常具有“稀”“细”“伴”的特殊地质属性, 已有方法和理论已难以满足研究的需要。为解决上述问题, 矿床地球化学国家重点实验室近十年来开展了系列元素-同位素(包括年代学)高精度、高空间分辨率分析方法以及元素分配-同位素分馏实验模拟和理论计算研究, 逐步形成了能够强力支撑关键矿产研究的技术方法体系。这些技术方法被广泛应用于钨、锡、锑、金、铜、钼、锂、稀土和稀散等关键金属研究, 极大地促进了成矿理论创新和找矿突破。本文对实验室近十年来相关技术方法的发展与应用进行了梳理, 以期是关键矿产研究和找矿勘查提供方法学参考。

关 键 词: 关键矿产; 成矿示踪; 元素-同位素; 微区原位; 实验模拟; 理论计算

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Some advances of tracing techniques and methods for studying metallogeny at the State Key Laboratory of Ore Deposit Geochemistry over the past decade

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Abstract: The tracing techniques and methods for studying metallogeny are fundamental to the ore deposit research, playing a crucial role in deeply revealing the sources, mobilization, migration and enrichment of the ore-forming materials, and the formation of ore deposits. As the research of critical or strategic mineral resources, which commonly have unique geological characteristics including the “low-grade”, “fine-grained” and “accessory” features, becomes hotspot of the ore deposit research, the available methods and theories can not meet the research needs of critical or strategic mineral resources. To address these challenges, relevant researchers of the State Key Laboratory of Ore Deposit Geochemistry have developed a series of high-precision and high-spatial-resolution analytical methods for elemental and isotopic (including geochronology) analyses, as well as experimental simulations and theoretical calculations on elemental partitioning and isotopic fractionation, and have progressively established a technical and methodological framework that can be used to strongly support researches of critical mineral resources over the past decade. The techniques and methods have been widely applied to study critical mineral resources such as tungsten, tin, antimony, gold, copper,

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molybdenum, lithium, rare earth elements and dispersed elements, greatly promoting the innovations of metallogenic theories and breakthroughs of mineral exploration. This paper reviews the developments and applications of these techniques and methods over the past decade, aiming to provide methodological references for critical mineral research and exploration.

Key words: critical mineral; tracing techniques for study metallogeny; element-isotope; in-situ analysis; experimental simulation; theoretical calculation

0 引言

作为矿床学研究的基础,成矿示踪技术方法对深入理解成矿物质来源、活化、迁移和富集成矿极为关键。与大宗矿产相比,关键金属矿产通常具有“稀”“伴”“细”的特征,即元素地壳丰度低、与主元素共/伴生出现、载体矿物极细,同时元素地球化学行为多样,成矿在时间和空间上呈现出高度的不均一性(翟明国等,2019;李文昌等,2022)。这些地质属性决定了关键金属矿产在源-运-聚示踪、找矿勘查评价和高效提取利用等方面面临巨大的挑战(李文昌等,2022)。单纯依赖宏观地质特征、全岩地球化学数据以及传统的元素分配-同位素分馏理论难以精准回答关键金属元素“从何而来、如何迁移、何处富集”等关键科学问题,因此对成矿示踪技术方法提出了更高的要求。

现代分析技术的快速发展为精细解析成矿过程提供了契机,使得矿床学研究从对成矿特征的观察描述转变为对成矿事件的精准回溯和定量约束(钟宏等,2021)。特别是随着激光、离子、电子等微束分析技术的发展,从更微观尺度、更高精度揭示元素赋存状态以及元素-同位素组成成为了矿床学研究的重要趋势和前沿。此外,随着可视化高温高压实验设备以及超算/理论计算能力的提升,“仿真”模拟成矿过程,构建适配的元素分配-同位素分馏理论,在拓展矿床学研究的深度和广度上发挥着越来越重要的作用。近十年来,矿床地球化学国家重点实验室围绕关键金属矿产成矿示踪,开展了系列元素-同位素(包括年代学)高精度、高空间分辨率分析方法以及元素分配-同位素分馏实验模拟和理论计算等研究,逐步形成了相互支撑的技术方法体系。这些技术方法被广泛应用于钨、锡、铋、金、铜、钼、锂、稀土和稀散等关键金属矿产的研究,在成矿理论创新和找矿突破中发挥了重要作用。本文对实验室近十年来相关技术方法的发展与应用进行了梳理,以期是关键矿产成矿示踪与找矿勘查提供方法学参考。

1 元素-同位素微区分析方法和标样

元素-同位素微区原位分析在甄别复杂地质事件、

刻画“瞬时”成矿过程等方面具有独特优势,成为精准示踪成矿物质来源、精细刻画成矿过程不可或缺的研究手段。除了仪器本身性能外,微区原位分析的精度和准确度在很大程度上依赖于基体匹配的标样。由于天然样品复杂多变,基体匹配的元素-同位素标样长期缺乏,严重制约了微区原位分析技术的发展和應用。

为解决元素-同位素高精度、高空间分辨率分析中存在的难题,实验室大力推进微区原位分析平台的建设,近十年来先后购置了多套激光剥蚀-电感耦合等离子体质谱(LA-ICP-MS)分析系统,包括193 nm准分子激光剥蚀系统、飞秒激光剥蚀系统、三重四极杆质谱仪、高分辨扇形磁场质谱仪以及多接收质谱仪等,同时引进了西南地区首台纳米离子探针(Nano-SIMS)。研究团队重点针对矿石矿物和贯通性矿物等开展研究。在元素分析方面,利用LA-ICP-MS建立了多种氧化物(如磁铁矿、铬铁矿)、硫化物(如闪锌矿、黄铁矿)、硅酸盐(如石英)和碳酸盐(如方解石)矿物的微量元素微区原位分析方法(Huang et al., 2015, 2016, 2017, 2023; 孟郁苗等, 2016; 蓝廷广等, 2017; Song et al., 2020; Liang et al., 2022; He et al., 2025; Chen D et al., 2026)。上述方法中,针对矿石矿物(如磁铁矿)标样缺乏且元素不均匀的问题,提出了“无内标-多外标”校正策略,利用不同的外标校正不同的元素,通过建立多外标校正曲线,实现对不同元素的高精度测定(孟郁苗等, 2016)。该方法为基体匹配标样缺乏的矿物微量元素LA-ICP-MS分析提供了灵活解决方案。在同位素分析方面,利用LA-MC-ICP-MS建立了闪锌矿、辉铋矿等硫同位素以及磁铁矿铁同位素微区原位分析方法(Dai et al., 2024a, 2024b; Huang et al., 2024);利用NanoSIMS建立了黄铁矿和黄铜矿超高空间分辨率($1 \times 1 \mu\text{m}^2$)硫同位素分析方法(Chen et al., 2022, 2023),磷灰石氧同位素和挥发分同时分析方法(Chen Y W et al., 2024),以及电气石硼同位素分析方法(Dong et al., 2025)。针对石榴子石氧同位素微区分析时普遍存在的基体效应,利用二次离子探针(SIMS)建立了含Ti石榴子石氧同位素分馏精细校正策略,显著提升了微区氧同位素分析准确性(Fu et al., 2025)。

在标样方面,实验室通过天然样品和人工合成相结合的方法,开发了多个硫化物、氧化物、碳酸盐等矿物元素-同位素微区原位分析标样(表1)。例如,通过对国内外典型矿床筛选,开发了适用于原位硫同位素分析的天然辉锑矿和黄铜矿标样(Chen et al.,2023; Dai et al.,2024a),适用于微量元素分析的铬铁矿标样(Chen D et al.,2026)。针对天然标样成分不均匀或元素-同位素含量低等局限性,以人工合成的方法,开发了量大、均一性好、元素-同位素种类多的标样。例如,利用水热合成结合低温高压致密化技术,成功制备了适用于LA-MC-ICP-MS硫同位素分析的闪锌矿标样(Dai et al.,2024b),适用于微量元素分析的方解石标样(He et al.,2025);采用高温-高压烧结技术,制备了化学成分和铁同位素均一的人工磁铁矿、赤铁矿、钛铁矿、钒钛磁铁矿标样(Huang et al.,2023,2024,2026);首次采用“水热前驱体合成+超高压烧结”技术,制备了满足SIMS严苛要求的黄铁矿硫同位素标样(Chen Y W et al.,2026)。值得一提的是,“水热前驱体+超高压烧结”通过将成分控制与致密化过程分离,避开了传统方法难以兼顾均一性与结构完整性的长期困境。这一策略的成功,不仅为硫同位素微区分析提供了急需的高质量标准物质,更具备广阔的应用潜力,未来可应用于其他硫化物(如黄铜矿、磁黄铁矿)、氧化物(如锡石、黑/白钨矿)和硅酸盐等矿物,有望缓解微区分析领域标准物质短缺的困境。这些工作系统性地构建了覆盖多种矿物体系的微区原位分析标样,在提升微区原位分析精度和准确性方面发挥了重要作用。

上述技术方法已被广泛应用于各类矿床成矿物质来源和成矿过程研究中。例如,基于铁氧化物和硫化物微量元素LA-ICP-MS分析,限定了铁铜矿的物质来源、成因类型和高品位矿石的形成机制(Huang et al.,2016,2018;陈应华等,2018;Sun et al.,2018;Zhao et al.,2020;邓聚庭等,2023),揭示了铜镍硫化物矿床和钒钛磁铁矿床的物质来源和富集机制(Kang et al.,2022;Liang et al.,2022,2023,2024,2025)。利用硫化物元素-同位素微区原位分析,揭示了我国西南地区代表性MVT铅锌矿床的成矿物质来源及稀散金属的赋存状态和分布规律(陶琰等,2019;Zhou et al.,2022; Mu et al.,2023;Wei et al.,2025;Qin et al.,2026)。将硫化物元素-同位素微区原位分析应用于金矿研究,系统揭示了胶东金矿从深部到浅部、从早期到晚期、从蚀变岩型到石英脉型含金黄铁矿微量元素-硫同位素变化过程,发现初始成矿物质具有低 $\delta^{34}\text{S}$ 值,而随着成矿流体上升,与前寒武纪基底和中生代花岗质围岩发生各种水-岩相互作用,导致 $\delta^{34}\text{S}$ 值升高和宽泛变化,提出成矿物质主要来自交代地幔、部分来自前寒武纪基底的成矿模型,为解决胶东巨量金的地壳-地幔来源之争提供了重要依据;同时基于黄铁矿微量元素点分析和面扫描,提出流体沸腾形成可见金的机制(Hu et al.,2022;Lan et al.,2023;Xiao et al.,2024; Wang et al.,2025)。上述研究有力地推动了从岩浆到低温热液等各类矿床成矿理论和成矿规律的认识,充分体现了微区原位元素-同位素分析技术在现代矿床学研究中的核心支撑作用。

表 1 本实验室开发的元素-同位素标样

Table 1 The element-isotope referencing samples developed by our laboratory

矿物类型	样品名	元素标样	同位素标样	属性	参考文献
黄铜矿	IGSD	—	S同位素	天然	Chen et al.,2023
辉锑矿	BJ-Snt	—	S同位素	天然	Dai et al.,2024a
闪锌矿	Sph-LD	微量元素	S同位素	合成	Dai et al.,2024b
黄铁矿	HPnc-Py	—	S同位素	合成	Chen Y W et al.,2026
方解石	CAL-D	微量元素	—	合成	He et al.,2025
铬铁矿	UG1-W	微量元素	—	天然	Chen D et al.,2026
磁铁矿	MtTR-1	微量元素	—	合成	Huang et al.,2023
	MtFe-1	—	Fe同位素	合成	Huang et al.,2024
	MtFe-2	—	Fe同位素	合成	
赤铁矿	HemFe-1	—	Fe同位素	合成	Huang et al.,2026
钛铁矿	IlmFe-1	—	Fe同位素	合成	
钒钛磁铁矿	VTiMtFe-1	—	Fe同位素	合成	
	VTiMtFe-2	—	Fe同位素	合成	

注:“—”表示无。

2 高精度成矿年代学

成矿时间约束是矿床学研究的基础问题,直接关系到对成矿背景的厘定、矿床成因和成矿规律的认识以及成矿-找矿模型的构建等。随着矿床学研究向精细化发展,高精度定年变得越来越重要。LA-ICP-MS分析兼具高精度、高空间分辨率以及快速、低成本的特点,是理想的定年手段。相对其他同位素定年体系,U-Pb同位素体系广泛存在于多种矿物和矿床中。因此,LA-ICP-MS U-Pb定年已成为矿床学最普遍、最有效的定年方法之一。然而,受限于铀含量低、普通铅含量高以及基体效应等因素,LA-ICP-MS U-Pb定年主要集中于锆石、磷灰石和独居石等少数高铀矿物,大量直接记录成矿过程的矿石矿物和脉石矿物难以获得可靠年龄。

针对矿石矿物和脉石矿物定年难题,实验室近年来通过开展激光剥蚀行为、元素-同位素分馏机制、基体效应校正策略和标样研发等研究,建立了多个矿石矿物和脉石矿物的LA-ICP-MS微区原位U-Pb定年方法。例如,通过联合使用NIST系列玻璃或其他较为成熟的U-Pb定年矿物标样,分别建立了黑钨矿、白钨矿、锡石、氟碳铈矿、钛铁矿、钛磁铁矿和磁铁矿等矿石矿物LA-ICP-MS U-Pb定年方法(Tang et al.,2020,2022a,2022b,2024a,2024b),以及方解石、石榴子石、符山石和萤石等脉石矿物U-Pb定年方法(Chen et al.,2021;Tang et al.,2021,2025;Xing et al.,2022;Guo et al.,2024)。其中,萤石LA-ICP-MS U-Pb定年为国际上首个建立的基体匹配萤石飞秒激光U-Pb定年方法(Tang et al.,2025),白钨矿同样首次实现基体匹配定

年(Tang et al.,2022a),而钛磁铁矿是新发现可进行U-Pb定年的矿物(Tang et al.,2024b),同时创造性地利用磷钇矿为氟碳铈矿U-Pb定年外标,促进了氟碳铈矿U-Pb定年方法的推广应用(Tang et al.,2022b)。上述方法中提出的针对不同矿物的组合校正策略,有效降低了基体效应和普通铅校正带来的不确定性,证实合理的分析条件和校正策略下,非基体匹配外标校正同样可以获得可靠的U-Pb定年结果,这为标样缺乏的矿物U-Pb定年提供了重要参考。此外,通过详细厘选,开发了黑钨矿、白钨矿、锡石、磷钇矿、钛铁矿、钛磁铁矿、磷灰石、萤石和方解石等定年新标样(表2)。相关标样储量丰富、铀含量高、均一性好,不仅丰富了U-Pb定年的标样体系,同时也为国内外相关单位提供了标准物质,极大地促进了LA-ICP-MS U-Pb定年技术的应用和发展。

以上技术方法在厘清矿床成因和成矿规律、促进成矿理论创新等方面发挥了重要作用。比如,将锡石U-Pb定年方法应用于东南亚巨型锡矿带,首次建立了东南亚锡成矿年代学格架,确定290~270 Ma锡成矿作用与古特提斯俯冲有关、230~210 Ma与大陆碰撞作用有关、90~50 Ma与新特提斯俯冲有关(Yang et al.,2020;Mao et al.,2020,2022)。通过白钨矿U-Pb定年,厘定了湘西沃溪巨型钨铋金矿主要形成于141 Ma,而非前人认为的加里东期,且可能存在后期流体改造事件(Tang et al.,2022a;祝亚男等,2023),同时也确定了湘中渣滓溪钨矿主要形成于晚侏罗世160~150 Ma(Wang T X et al.,2024),而非前人报道的晚三叠世。通过铁氧化物(钛铁矿、钛磁铁矿和赤铁矿

表 2 本实验室开发的矿物微区U-Pb定年标样

Table 2 The *in-situ* U-Pb dating referencing mineral samples developed by our laboratory

矿物类型	样品名	U含量/(μg/g)	Pb含量/(μg/g)	年龄/Ma	参考文献
黑钨矿	KA-19	9.3~45.3	0.8~5.8	429.2±6.8	Tang et al.,2020
	KA-18	19.6~72.9	1.4~5.6	425.9±4.3	
	NM	11.8~136	0.4~2.7	142.3±1.3	
锡石	BGH/KA	0.8~23.6	0.1~1.5	425.6±5.7	
白钨矿	WX27	0.3~24.6	0.1~0.8	141.2±2.8	Tang et al.,2022a
磷钇矿	Zagi	991~1161	7~9	36.7±0.4	Tang et al.,2022b
钛铁矿	HG79	0.02~0.31	0.01~4.03	259.3±1.3	Tang et al.,2024a
	XL32333	0.15~0.93	0.02~0.05	196.0±2.7	
钛磁铁矿	HG79c	0.01~0.47	0.02~0.84	259.2±2.8	Tang et al.,2024b
磷灰石	QH	61~749	3.12~24.8	159.5±0.8	Xia et al.,2025
萤石	HN	3.30~8.79	0.12~0.34	158.7±1.4	Tang et al.,2025
方解石	DC22	约4.0	0.3	153.5±0.8	

等)LA-ICP-MS U-Pb定年,限定康滇铁铜成矿带发生多期铁成矿事件,包括早期发生在约1080 Ma,晚期发生在(886±40)Ma和(853.2±9.9)Ma(Tang et al., 2024a, 2024b)。利用萤石LA-ICP-MS U-Pb定年方法,将晴隆锑矿成矿时代限定至60 Ma(Tang et al., 2025),而非前人的约140 Ma。在华南地区发现一批时代相近的钨、锑、萤石成矿事件,如广西丹池铁锌锡矿床中的锑矿化(62.8 Ma; 肖昌浩等, 2024)、湖南锡矿山(约60 Ma; Xu et al., 2022)、浙江临安萤石矿(约60 Ma)、广西某钨锑矿(约60 Ma),表明华南在该时间段存在一期区域性的成矿事件。上述研究对构建华南高温钨锡和低温锑金成矿关联、建立统一的陆内成矿理论提供了关键的年代学证据。

3 成矿流体原位分析和示踪技术

作为古流体的保存者,流体包裹体记录了成矿流体的细微变化,是精细研究成矿流体来源和演化过程的绝佳对象。长期以来,流体包裹体研究主要通过冷热台显微测温获得温度、盐度和压力等参数,或者通过激光拉曼光谱、傅里叶变换、红外光谱等鉴定组分信息,难以定量获得单个流体包裹体的元素(如微量元素)信息。虽然离子色谱仪和四极杆质谱仪等可对群体流体包裹体成分(气体及阴、阳离子等)进行定量分析,但获得的是混合结果,不能满足精细成矿研究的需要。随着LA-ICP-MS分析技术的发展,单个流体包裹体成分LA-ICP-MS分析应运而生。该方法能够实现单个流体包裹体中多达几十种元素的同时原位高精度检测,在精准揭示成矿流体性质、来源和演化等方面有着传统方法无可比拟的优势。国际上从20世纪90年代就开展了单个流体包裹体成分LA-ICP-MS分析研究,但仅少数发达国家建立了相关分析方法,国内截止到2015年尚未建立。此外,随着激光拉曼光谱定量分析技术的发展,利用激光拉曼光谱仪与冷热台和热液金刚石压腔等可视化高温高压设备联用,可实现单个流体包裹体成分和相态随温压变化的实时观测,对“仿真”模拟成矿过程、精准揭示成矿机理起着越来越重要的作用。

2015年以来,实验室购置了LA-ICP-MS分析系统,在国内率先开展了矿床学领域单个流体包裹体成分LA-ICP-MS分析研究(蓝廷广等, 2017)。通过优化流体包裹体人工合成方法,合成了适用于LA-ICP-MS分析的多元素流体包裹体标样,同时研制了适用于流体包裹体分析的激光剥蚀样品池,克服了LA-ICP-MS分析过程中流体包裹体信号强度低、持续时间短、误差大等难题,建立了稳定可靠的单个流体包裹体成分

LA-ICP-MS分析方法。分析结果表明,流体包裹体元素分析相对误差绝大多数在±10%以内,相对标准偏差(RSD)小于5%,与国际同行实验室相当或更优。随后将飞秒激光引入单个流体包裹体成分分析,将分析成功率提升至90%以上(蒋子琦等, 2024),解决了传统193 nm准分子激光在分析石英中流体包裹体成分时成功率极低的问题。同时利用高分辨激光拉曼光谱仪与冷热台和热液金刚石压腔等可视化实验设备联用,结合熔融石英毛细硅管合成流体包裹体法,开展了成矿流体/元素高温高压下的原位实时观测研究。

上述方法的建立为关键矿产成矿理论创新提供了重要支撑。蓝廷广等(2017)和Lan等(2018)利用单个流体包裹体成分LA-ICP-MS分析方法对华北克拉通陆内环境的王家庄铜钼矿床开展了研究,发现该矿富气相包裹体更富铜,而含子矿物包裹体富钼,暗示铜和钼可能具有不同的搬运机制,即铜更倾向于进入气相,而钼则倾向于进入高盐度液相。结合流体沸腾现象的存在,这可能是导致该矿上铜下钼分带的重要因素。Mu等(2021)将单个流体包裹体成分LA-ICP-MS分析方法应用于云南金顶世界级铅锌矿研究,发现该矿闪锌矿和方解石中流体包裹体元素组成与盆地卤水相似,证实成矿流体为与围岩发生元素交换的残留海水(形成富钙、锌和铅流体),流体在上升过程中与低盐度、富H₂S的还原流体混合,最终导致成矿元素沉淀。Zhao等(2021)对福建紫金山罗卜岭斑岩铜钼矿开展了系统的单个流体包裹体成分LA-ICP-MS分析,发现该矿各阶段流体均源自罗卜岭花岗岩闪长斑岩,斑岩在低压条件下直接出溶高盐度流体相和低密度气相,钼、铜、铅、锌、银在高盐度流体相中迁移,部分铜可能以气相形式迁移,包裹体元素含量的变化指示钼主要在约500 ℃沉淀,而铜在350~450 ℃沉淀。Wang H等(2022)首次对胶东金矿开展了单个流体包裹体成分LA-ICP-MS分析,发现金矿的流体包裹体组成与中、高级变质流体相似。结合研究区域演化历史和热力学模拟,提出胶东巨量金矿成矿流体可能来自底侵中下地壳的基性岩脱水的成矿新模型。Peng等(2023)将单个流体包裹体成分LA-ICP-MS分析方法应用于华南天堂山铷-钨-锡多金属矿共生分异机制研究,发现该矿成矿流体均初始源自岩浆流体,中晚期有大气降水加入,花岗岩岩浆直接出溶富含钨、锡、铷的岩浆流体,钨-锡和铷为早期流体向外演化关系,水-岩反应是早期钨和铷沉淀成矿的主要机制,而流体沸腾+大气水混入是晚期锡沉淀的主要机制。Jiang等(2024)将单个流体包裹体成分LA-ICP-MS分析与实验模拟结合,开展了CO₂对钼成矿影响的实验模拟研究,发现钼主

要分配进入高盐度流体相, CO_2 的存在容易导致流体在深部发生不混溶形成高盐度流体, 促使钼进入高盐度流体, 这对深入理解我国独特发育的大型斑岩钼矿(富 CO_2)的成矿机制和深部找矿勘查具有重要指示意义。Mao等(2024)将单个流体包裹体成分LA-ICP-MS分析与理论计算结合, 建立了沸腾包裹体组合中富气相流体包裹体元素含量更准确的评估方法。

在流体包裹体激光拉曼原位实时观测方面, Chen C等(2024)利用熔融石英毛细硅管合成 Li_2SO_4 、 Na_2SO_4 、 K_2SO_4 和 Cs_2SO_4 硫酸盐流体包裹体, 使用激光拉曼光谱建立了25~250 °C之间硫酸根含量的定量分析方法, 为测定天然矿床(如稀土矿床)中成矿流体的硫酸盐浓度奠定了基础。Zhou等(2025)同样使用熔融石英毛细硅管合成流体包裹体的方法, 利用激光拉曼光谱开展了250~350 °C之间铝对硫酸盐热化学还原反应(TSR)影响的研究, 发现 AlCl_3 的加入有利于TSR的发生, 指示在低至250 °C的富铝地质环境中TSR有可能在短时间内发生并积累足够的还原硫, 促进硫化物矿床的形成。Wang等(2026)利用上述方法, 开展了25~300 °C之间 Cu^+ 和 Cu^{2+} 在富Cl流体中的溶解度和络合物种类研究, 发现 Cu^+ -Cl体系在高温下(>200 °C)铜主要以 $[\text{CuCl}_2]^-$ 存在, 而 Cu^{2+} -Cl体系在100~300 °C温度区间则以 $[\text{CuCl}]^+$ 和 $[\text{CuCl}_2]^0$ 为主要存在形式, Cu^{2+} 还原为 Cu^+ 可促进Cu的沉淀。

4 成矿实验模拟和示踪方法

成矿实验模拟是揭示元素地球化学行为、反演成矿过程的重要手段。将高温高压实验和原位分析技术相结合, 可查明熔体、矿物、流体在不同地质过程中的相态和热力学参数, 揭示元素在不同相态中的存在形式、溶解度以及分配行为等。结合分子动力学和热力学计算, 可定量模拟成矿过程, 为构建高精度成矿-找矿模型、指导勘探找矿提供重要支撑。实验室是国内最早开展成矿实验模拟的机构之一, 经过数代人的努力, 目前已建立拥有多种大压机、活塞圆筒、冷封式高压釜、金刚石压腔以及激光拉曼光谱原位观测系统在内的先进实验模拟平台, 可开展-110~2500 °C和1~25 GPa的宽域温压实验, 实现从核幔边界至地表过程的元素地球化学行为模拟。

近年来, 实验室针对多种关键金属元素(如钨、钼、金、锑、铋、铊等)开展了迁移-沉淀-富集实验模拟研究, 在矿床成因方面取得重要认识。例如, 利用水热高压釜及毛细石英管与激光拉曼光谱相结合的原位观测技术, 开展了系列钨实验模拟研究, 主要结果包括: (1)在250~500 °C、0%~5% NaCl_{eq} 的热液中, 钨主要

以 HWO_4 形式迁移, 而非Na-W或W-Cl络合物, 否定了以往钨主要以Na-W络合物形式在热液中迁移的传统认识(Wang et al., 2019); (2)钨在含F的 $\text{NaCl-H}_2\text{O}$ 热液体系($\text{pH}<5$, $m_F<0.3$ mol/kg)中, 主要以 $\text{H}_3\text{WO}_4\text{F}_2$ 形式迁移, 发现富F热液中钨的溶解最高可达不含F热液的100倍(Wang X S et al., 2021); (3)在酸性-中性的含F、P热液($m_F<0.05$ mol/kg, $m_P<0.02$ mol/kg)中, 由于萤石(CaF_2)和磷灰石 $[\text{Ca}_5(\text{PO}_4)_3\text{F}]$ 的生成, 抢夺了形成白钨矿(CaWO_4)所需的 Ca^{2+} , 抑制白钨矿沉淀/促使白钨矿溶解, 导致热液中钨含量升高而形成更加富钨的热液, 进而定量刻画了富F、P热液促使低品位钨矿活化活化再富集成矿的精细过程(Wang et al., 2023)。

针对钼, 利用快速淬火冷封式高压釜和原位捕获高温高压流体技术, 发现: (1) NaHMoO_4^0 是钼在含盐热液体系中的主要溶解迁移形式, 钼的溶解度随着盐度和pH的增加而增大, 钼在流体/熔体间的分配系数与熔体铝饱和指数呈负相关(Shang et al., 2020; Jiang et al., 2021); (2)在富 CO_2 体系中, 由于 CO_2 的存在, $\text{NaCl-H}_2\text{O}$ 体系气液两相区范围显著扩大, 导致热液以高度富钼的高盐度流体从熔体中出溶, 相比于不含 CO_2 体系, 钼的萃取效率提高了约5倍, CO_2 的存在促使了钼的超常富集。

针对金, 开展了铍对金迁移-富集作用以及纳米金形成的实验模拟。Wang X S等(2022)实验发现, 在中酸性富铍(0.005~0.49 mol/kg NH_3^0)热液中, 金可与 NH_3^0 形成稳定络合物 $\text{Au}(\text{NH}_3)\text{OH}^0$; 但在典型的浅成低温低硫型热液金矿床的成矿热液中, 金的溶解仍然受到 HS^- 含量和pH的控制, 而非形成 Au-NH_3 络合物控制, 富铍蚀变是通过控制热液pH而影响金的富集, 为通过铍蚀变寻找金矿化提供了理论依据。Nie等(2023)通过开展含砷黄铁矿吸附纳米金实验, 发现砷的类质同象替代增加了黄铁矿表面反应活性, 促进了金的吸附富集, 并且在偏酸性、含有适量有机质以及较高盐度的热液中, 有利于含砷黄铁矿对纳米金的吸附和固定, 为揭示卡林型金矿等矿床中纳米金的富集提供了重要证据。

针对锑, Fu等(2020b)采用硅酸盐玻璃颗粒剂间捕获流体技术, 在850~1050 °C和100~200 MPa条件下系统研究了锑在含氯流体与流纹岩熔体间的分配系数, 发现锑在流体和熔体间的分配系数均值为 0.81 ± 0.18 , 且受流体盐度、氧逸度和温度影响弱, 流体从熔体中萃取锑的效率极低, 表明在岩浆演化过程中分异出的流体不足以为形成大型-超大型锑矿床提供足够的锑, 大型锑矿床的锑可能主要来自围岩再活化。

针对铋, Tang等(2023)实验发现在650~850 °C和200 MPa水饱和条件下, 烧绿石溶解度随温度升高而

显著增加,随熔体A/CNK值降低而增加。在过碱性熔体中,烧绿石溶解度低于铌铁矿,而偏铝质熔体中则相反,进而定量推导出溶解度积与温度和A/CNK的经验方程,揭示了温度降低和钙质含量增加促使烧绿石在过碱性岩浆中优先结晶的机制。

针对铋,Wang X S等(2024)实验揭示铋的迁移形式受到流体密度的控制。在较低密度流体中,铋主要以不含水的气相分子 $\text{BiCl}_3(\text{g})$ 形式迁移,随着流体密度的增加则主要以水合分子 $\text{BiCl}_3\text{H}_2\text{O}_n(\text{g})$ 的形式迁移。结合数值模拟,提出在岩浆-热液系统中气相/低密度超临界流体可大量迁移铋,进而促进铋在浅成环境中富集。

5 同位素示踪新理论新方法

同位素示踪是精准揭示成矿物质来源和成矿过程必不可少的研究手段。面对关键金属矿产的特殊性,传统的同位素示踪理论和方法可能失效,亟需创新同位素示踪理论和方法。近年来,伴随计算机技术与量子化学(及物理)计算方法的迅速发展,同位素示踪理论实现了深刻变革,理论与计算地球化学的学科支撑作用日益凸显。在技术方面,实现了多种同位素特别是金属稳定同位素的高精度测试,为关键矿产成矿理论的创新提供了重要支撑。

实验室是国内较早开展同位素示踪理论和方法研究的单位之一。在同位素分馏理论方面,近年来通过构建涵盖高温热扩散、核场位移效应、超低温分馏及多阶段过程放大效应的全新理论框架,开拓了团簇同位素、位置特异性同位素等新兴研究方向,革新了矿物-溶液体系高精度同位素分馏计算方法。比如,传统同位素分馏理论在高低温极端条件、重元素体系及复杂地质过程应用中遭遇显著瓶颈,实验室构建了一系列新型同位素分馏理论模型以解决上述问题。Li和Liu(2015)针对高温条件下(如硅酸盐熔体)温度梯度驱动的同位素分馏现象,提出了基于局部热力学平衡的统一理论模型,为理解成矿演化过程中的同位素异常提供了核心理论基础。Yang和Liu(2016)针对铈、钐、铕等重元素中存在的“奇-偶”非质量依赖分馏现象,系统阐释了核场位移效应的基本原理及其地质意义,证实核场位移效应驱动的分馏幅度可显著超越传统质量依赖效应,为解析成矿体系中同位素异常(如 ^{182}W)的成因开辟了新路径。Zhang和Liu(2018)构建了适用于超低温条件的平衡同位素分馏计算新方法,首次系统引入核自旋权重、玻恩-奥本海默近似修正及非平面分子反转分裂效应等关键量子修正项,进一步完善了同位素分馏理论体系,为全温度区间同位素示踪提供了更高精度的理论工具。此外,Zhang和Liu

(2020)还挑战了“地幔样品中放射性成因同位素异常仅源于母体衰变”的传统认知,创造性提出核场位移效应等产生的微弱质量非依赖分馏,可通过地幔多阶段熔融-结晶的封闭系统演化过程被显著放大,为利用高精度同位素示踪深部物质循环提供了全新研究维度。在预测矿物-流体和矿物-矿物间同位素分馏行为方面,实验室构建了一系列精准计算方法。Gao C等(2018)发展了变体积簇模型(Variable-Volume Cluster Model, VVCM)量子化学方法,成功计算了含镁碳酸盐、无定形碳酸钙、水镁石与含水物种之间的镁同位素平衡分馏值,证实局部构型采样对精确预测矿物-溶液分馏的关键作用,为阐明表生和岩浆过程中镁同位素地球化学行为提供了核心参数。Yin等(2023)提出一套新型计算方法用于精确预测水-岩相互作用的同位素分馏,该方法通过整合耦合簇CCSD(T)高精度计算、高阶能量项修正及溶剂化效应,为深入理解水-岩相互作用中的同位素分馏行为提供了可靠理论支撑。Gao C等(2023)基于第一性原理分子动力学训练的深度势能模型,结合路径积分分子动力学(PIMD)模拟,精确计算了水镁石与水之间的H和O同位素平衡分馏参数,明确了H同位素分馏对压力变化的极端敏感性,凸显了高压效应在成矿流体示踪中的重要意义。依托同位素分馏理论与计算方法的创新突破,实验室还开展了一系列新同位素体系的探索性研究。Liu和Liu(2016)发展了超越简谐水平的高精度同位素分馏计算方法,为新兴团簇同位素温度计的建立提供了高精度平衡分馏值参考标尺。Liu等(2021)针对烃类形成、迁移和转化过程示踪的实际需求,开展了位置特异性碳、氢同位素平衡分馏因子的精确计算。Zhang等(2020)从分子机制层面提出磷酸消化碳酸盐反应的平行路径新模型。上述研究为将团簇同位素与位置特异性同位素发展为新型地球化学温度计与示踪工具奠定了坚实的理论基础。

实验室同样是国内较早从事金属稳定同位素研究的机构之一,系统开展了多种金属稳定同位素分析方法、分馏理论以及地质应用研究。比如,较早开展了钨、钼、钨同位素研究(温汉捷等,2008;张羽旭等,2009,2010),近十年开展了镓、锡、铟等同位素研究(Zhang et al.,2021a,2024; Song et al.,2024; Zhu et al.,2024),并对早期方法进行了完善(Liu et al.,2016, Zhang et al.,2018,2021b; 闻静等,2020)。针对铜同位素,近年来建立了超低含量样品(比如碳酸盐岩)双稀释剂法(Liu et al.,2016; Zhang et al.,2021b),以及高铁含量等特殊样品(比如含大量黄铁矿的地质样品)化学分离方法(闻静等,2020),形成了适用于绝大多

数地质样品的钼同位素分析方法体系。针对钼,近年来也建立了超低含量样品双稀释剂法(Zhang et al., 2018, 2023),同时建立了一柱多元素分离法(Zhu et al., 2020),即经一次分离纯化流程即可实现铅、锌、钼的纯化,极大地节省了分析成本并提升了效率。相关方法在揭示铅锌矿床物质来源、演化过程和成因机制等方面取得重要进展(Zhu et al., 2016, 2017, 2020, 2021; Song et al., 2023)。针对镓,建立了铝土矿和闪锌矿中镓化学分离方法以及同位素测试方法(Wen et al., 2021; Zhang et al., 2021a, 2024),解决了相关样品中镧、铈、钕、铈、铈等干扰严重问题,为揭示铝土矿、碳酸盐黏土型锂矿以及铅锌矿成因提供了重要支撑(Zhang et al., 2025)。实验室最近也建立了银、铜、锡等同位素分析方法(Gong et al., 2024; Song et al., 2024; Zhu et al., 2024),为揭示富银和铜的硫化物矿床以及钨锡等矿床成因机制提供了新的手段。这一系列同位素分析方法的建立以及相应的同位素分馏理论的完善,极大地拓展了金属同位素在成矿示踪方面的应用,弥补了传统方法的不足。

汞同位素是自然界少数具有显著质量分馏与非质量分馏的金属同位素体系。汞同位素非质量分馏主要产生于地表光化学过程,而成岩成矿地质过程难以产生汞同位素非质量分馏(Bergquist and Blum, 2007)。因此,汞同位素非质量分馏信号($\Delta^{199}\text{Hg}$)在成岩成矿过程中具有保守性,能为物质来源和地质过程提供准确、可靠的约束(Yin et al., 2024)。基于汞同位素的重要示踪潜力,实验室较早开展了成岩成矿汞同位素示踪研究,在岩矿汞同位素分析方法、成岩成矿示踪原理及成矿示踪应用方面取得了系列进展,形成了较为系统的汞同位素成矿示踪理论方法体系。比如,针对岩矿样品汞含量波动大、干扰元素多的汞同位素分析难题,Gao L J等(2024)建立了含锰催化剂反应管提纯汞的方法,实现了岩矿样品的汞同位素准确测定。通过对岩浆和热液体系的持续研究,确立了交代地幔、大陆地壳、蚀变洋壳等重要地质储库的汞同位素组成特征(Deng et al., 2021; Wang X Y et al., 2021; Yin et al., 2022; Tian et al., 2023),为汞同位素示踪应用提供了重要基准数据。通过对不同类型热液矿床的汞同位素组成的进一步研究,发现俯冲带及陆内等不同构造背景和地质条件下形成的热液矿床汞同位素组成存在显著差异,从而更精准地揭示了不同的成矿金属来源和物质循环模式(Yin et al., 2016; Xu et al., 2018; Fu et al., 2020a; Deng et al., 2021; Gao W et al., 2023; Liu et al., 2026),为完善区域成矿理论与建立成矿模型提供了重要依据。

6 总结与展望

围绕关键矿产研究对高质量分析数据的迫切需求,实验室近十年来开展了有针对性的成矿示踪技术方法研究,在高精度成矿年代学以及元素-同位素微区原位分析和实验模拟等方面取得系列进展,同时创新了元素分配-同位素分馏理论,支撑关键矿产成矿理论取得突破,服务找矿取得重要进展。技术方法的进步显著提升了成矿信息获取的精度和准确度,使得成矿年代学架构得以精准构建,成矿流体与物质来源实现多维度示踪,成矿过程得到精细刻画,推动矿床学研究由依赖宏观特征和间接推断逐步转向在多维度、多参数约束下的定量解析。

展望未来,面对关键矿产成矿系统极端的复杂性与非均一性,成矿示踪技术方法需在微区原位分析技术、非传统元素-同位素示踪理论、高温高压“仿真”实验模拟以及高精度地球物理探测等方面持续发展,同时推动数智融合,利用大数据和人工智能挖掘复杂数据中的成矿规律,将前沿分析技术与矿床问题紧密结合,推动高质量数据向资源预测、勘查和评价转化,在深化成矿理论认识和服务国家资源安全方面发挥更重要的作用。

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利益冲突声明: 作者保证本文无利益冲突。

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