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实验地球化学的发展历史和研究展望

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摘要: 实验地球化学主要通过高温高压实验模拟, 对元素和同位素在地球内部条件下的行为、性质和效应进行研究, 从而对成岩成矿、岩浆演化、流体交代、壳—幔—核分异等地质现象和过程进行制约。实验地球化学的最初诞生, 主要是针对传统地球化学、岩石学和矿床学研究中遇到的难以解决问题进行正演辅助。实验地球化学的发展, 与高温高压实验设备和现代分析技术的成熟和完善密切相关。近半个世纪以来, 实验地球化学的不断成长壮大, 极大促进了传统地球化学乃至整个地球科学相关领域的发展。在未来的 10 到 20 年内, 实验地球化学有望在以下 3 个方面进一步加强和取得重要科研成果: (1) 深部地球和早期地球; (2) 挥发分和地球宜居性; (3) 行星形成演化实验模拟。

关键词: 实验地球化学; 高温高压; 发展历史; 研究展望。

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Advances and Perspectives of Experimental Geochemistry

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Abstract: Experimental geochemistry involves the simulation of the physical and chemical conditions of the Earth's interior. By this, the behavior, nature and effects of elements and isotopes are studied experimentally, so as to constrain processes such as petrogenesis and mineralization, magma evolution, fluid metasomatism, and differentiation. The field of experimental geochemistry emerged as a tool to offer forward modeling for challenging issues that are difficult to address by studies with traditional geochemistry and petrology. The rapid development of the field is attributed to the improvement of facilities for generating high-pressure and high-temperature conditions and the availability of modern analytical techniques. In the past about half century, the growing research in the field of experimental geochemistry has greatly promoted the development of traditional geochemistry and even the entire earth science related fields. In the next one or two decades, experimental geochemistry is expected to further strengthen important scientific achievements in the following aspects: (1) deep Earth and early Earth; (2)

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volatiles and habitability of the Earth; (3) experimental simulations and planetary science.

Key words: experimental geochemistry; high pressure and high temperature; research history; research perspectives.

0 引言

地球内部几乎超过 99% 以上的部分,都处于超过 1 GPa 和 500 °C 的高温高压条件下.这使得整个地球内部是一个巨型的天然高温高压反应炉,其组成物质长期处在复杂的化学和物理反应中.实验地球化学,顾名思义,主要是通过高温高压实验模拟,研究元素(包括主量、微量元素)和同位素在高温高压条件下(乃至相变过程中)的行为、性质和效应;利用化学、物理、物理化学等领域的新手段,探索地球内部的地质作用现象和过程,从而为正确理解、制约和解决与地球化学相关的重要科学问题提供基础依据.

事实上,实验地球化学与实验岩石学、实验矿床学、矿物物理学乃至其他实验地球科学都存在直接或间接的联系.尤其需要说明的是,学术界对实验地球化学的概念并没有统一的认识,甚至国内外一些早期的教科书中把实验地球化学等同于实验岩石学、实验矿物学、成岩成矿实验等.在某种程度上,这是因为高温高压实验研究的综合性非常强,其在地球科学中的应用充分体现了地球科学作为一门系统科学(即地球系统科学)的特点.实验地球化学的诞生和发展,与传统地球化学乃至整个地球科学的发展息息相关,也与实验仪器设备的发展和测试技术的完善高度相辅相成(杨晓志, 2015).实验地球化学促进了传统地球化学乃至整个地球科学的发展进程,对人们深刻认识深部地球、早期地球和类地星体的地球化学演化和模型起到了重要的积极作用.本文就实验地球化学的发展历史和研究现状进行简要回顾与展望.

1 实验地球化学的诞生背景

20 世纪的 40 至 60 年代,是传统地球化学研究快速发展的时期(Holland and Turekian, 2014).微量元素和同位素的深入应用以及新的示踪体系的出现,使得传统地球化学研究手段日益多样化.大量地质现象的观测和地球化学数据的积累,极大地促进了地球化学和岩石学的学科发展.但同时也产生了一系列通过常规研究手段难以妥善解决的难

题,比如流体(包括熔体)的性质、流体与岩石的相互作用以及共存相间的元素分配和同位素分馏等等.这主要是由于天然地质样品往往经历了复杂多期次的地质作用,但人们能获取的样品往往主要只保留了其最终演化阶段的一些组成信息,对这些地质样品中间过程的反演是地球化学研究的一大难题.高温高压条件下的实验研究就显得很有必要也非常迫切,因为这几乎是正演或复原地质样品演化过程中元素和同位素行为的唯一直接途径.

几乎是在同一时期,常压气体混合炉、热液型压机、活塞圆筒压机、多面砧压机和金刚石对顶砧等高温高压设备被不断发明和逐渐完善(Darken and Gurry, 1945; Hall, 1958; Jamieson *et al.*, 1959; Osborn, 1959; Weir *et al.*, 1959; Boyd and England, 1960; Dickson *et al.*, 1963).实验技术手段的日益丰富,温压参数控制方面的不断改进,使得在 60 多年前就实现了从浅部地壳到浅部地幔条件的室内模拟.这一时期也是包括电子探针、离子探针和质谱等现代分析技术高速发展的阶段,微束、微区、微量测定技术的涌现,也为高温高压实验中小体积和小质量的准确表征提供了重要契机.正是在这种局面下,一些学者通过高温高压实验模拟,开展了一些与元素和同位素相关的地球化学方面研究工作.从某种程度上可以说,实验地球化学就是在这种环境中应运而生并得到迅速发展的(Holloway and Wood, 1988).

2 实验地球化学的早期发展

从大约 1960 到 1990 年的近 30 年间,可以看作是实验地球化学的初级发展阶段.在这个时期,实验地球化学研究主要关注近地表过程中元素和同位素的地球化学行为,相关工作围绕传统地球化学和岩石学研究中一些自身难以解决的难题,藉由实验模拟来开展相关的辅助性工作.这些实验研究的重点,主要是针对当时传统地球化学研究中的前沿或基础问题,包括岩石蚀变、部分熔融、流体作用、水岩反应等地质过程以及相关的元素和同位素行为等等.这些工作的着眼点主要是地壳和浅部地幔较浅深度的地球化学作用过程,实验温压条件相对

较低,研究对象主要是主微量元素和同位素在蚀变、熔融、交代、变质和岩浆作用等常见典型地质作用过程中的性质和行为以及矿物在流/熔体中的溶解能力,由此对相关的地质现象和地球化学数据提供制约与解释。

实验地球化学的很多经典工作都是在这个时期奠定的,包括矿物与流体的互溶性(Wasserburg, 1958; Burnham and Jahns, 1962; Anderson and Burnham, 1965)、热液-蚀变-熔融过程中的元素行为(Boettcher, 1970; Cullers *et al.*, 1973; Shimizu, 1974; Bischoff and Dickson, 1975; Drake and Weill, 1975)、氧同位素分馏(O'Neil and Taylor, 1969; Clayton *et al.*, 1972)、氢同位素分馏(Suzuki and Epstein, 1976)以及硫和碳同位素分馏(Mook *et al.*, 1974; Smith *et al.*, 1977; Ohmoto and Rye, 1979)等等。这些实验数据的测定,为微量元素地球化学和同位素地球化学提供了重要的理论基础,极大地促进了这些相关学科的快速发展。几乎是在同一时期,一些学者对共存相间元素的地球化学分配性质也进行了实验研究,导致了一些经典地质温压计的出现,并被广泛用于包括岩浆岩和变质岩等体系平衡条件、成岩过程和演化历史等的判断(Ellis and Green, 1979; Graham and Powell, 1984; Harley, 1984; Brey and Kohler, 1990),这对固体地球科学的发展也有积极的促进作用。

我国静高压实验技术也是在这一时期得到初步发展的。老一辈学者们筹建或研发了一些高温高压实验装置,并利用这些设备开展了一些与地球化学相关的研究工作。1959年中科院地质研究所在国内率先筹建高温高压实验室开展成岩成矿模拟研究,随后70年代中期中科院地球化学研究所设计研制了YJ-3000t型六面顶压机(谢鸿森和候渭, 1992)。之后的80年代,熊大和等(1981)使用金刚石对顶砧对地质流体开展了一些研究,Zeng *et al.* (1989)利用高压釜研究了矿物在热液流体中的溶解性等。这些工作也是围绕当时国内地学领域非常前沿的方向,此外,一些学者还研究了矿物和岩石的元素组成以及脱水作用对其物理性质(包括流变、导电性等)的影响,并用于地球物理野外探测数据的解释(顾芷娟等, 1990)。

可以说,从最初诞生之日起,实验地球化学最显著的特征就是聚焦传统地球化学研究和岩石学的前沿问题,从高温高压实验角度对其进行有效约

束。传统地球化学对天然地质样品的研究,由于其研究对象是经历了复杂地质作用和漫长演化历史的最终产物,对样品形成演化过程的制约其实并不直接;同时,天然地质样品起源深度和空间分布往往有限,人们在获取深部天然地质样品方面具有较大的难度和挑战性。实验地球化学的强大之处,是能准确模拟地球内部的温度、压强、逸度等条件,从而对天然地质样品可能经历的中间阶段逐步还原,并把每一阶段可能产生的地质烙印解析出来。这也是实验研究获取的微量元素分配系数和同位素分馏系数,被大量用于天然样品组成平衡判别的重要原因。此外,还可以在即使没有天然样品的条件下,通过模拟深部地球和早期地球极端高温高压氛围进行地球化学方面的研究。

3 实验地球化学的研究现状

自1990年来的近30年间,实验地球化学不断发展壮大,其研究范畴、研究对象和研究内容上,与前30年间有了显著不同。除了为传统地球化学研究提供辅助支撑外,实验地球化学围绕地球深部和行星科学的一些重大科学问题和学科发展需要,逐渐在越来越多的研究领域发挥着突出作用。如果说实验地球化学在30年前仅关注近地表附近深度、能实现的温压条件有限且只能在一些零星科研机构开展,那么在过去的30年间就是取得了突飞猛进的进展。总的来说,实验地球化学研究具有了更显著的多元化和多样化特征,特别是作为一门独立学科独立性也更为突出,世界范围内的高温高压研究机构也不断增多。

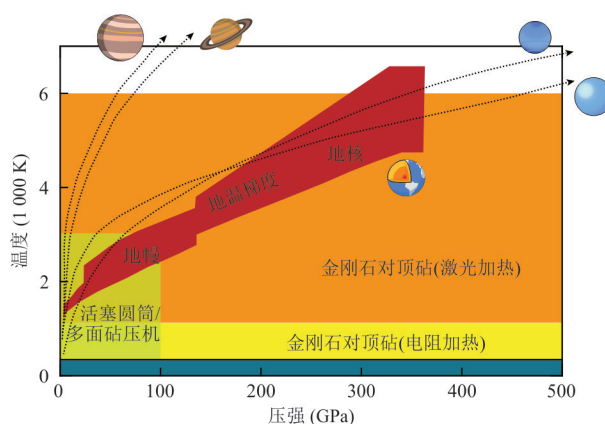


图1 高温高压设备温压条件与行星内部对比
Fig. 1 Experimental conditions and planetary interior

修改自 Bass(2004)

这主要得益于3个方面的发展。首先,传统地球化学数据的大量积累,为实验地球化学研究开展奠定了基础,推动了实验地球化学的研究进程。其次,高温高压实验技术总体较为成熟,活塞圆筒压机、多面砧压机和金刚石对顶砧这些关键设备的安装和使用进入了常态化阶段。截止目前,高温高压实验设备已经允许开展从地球的地表到地核乃至其他行星的各类模拟研究(图1)。再者,分析测定手段的多样化,特别是微区、微量、微束、原位分析技术的不断改进和完善,为实验地球化学的发展注入了新的活力。高温高压实验样品一般较小,特别是高压下实验产物尺寸往往在微米至毫米尺度(Manghani and Yagi, 1998; Miletich, 2005; Tyagi and Banerjee, 2017),对这些极小样品的精确表征非常重要。电子探针、离子探针等微区微束分析技术的改进和分辨率的提高(Shimizu and Hart, 1982; Reed, 2005; Goldstein, 2012)以及单接收器和多接收器质谱等微量痕量分析技术的改进(Vanhaecke *et al.*, 2009; Magyar *et al.*, 2016),为准确测定实验产物的化学成分奠定了重要基础。此外,红外、拉曼、X射线等谱学技术的发展,也为测定实验产物的组成和结构等提供了重要条件,这些谱学技术与高温高压设备的联用,更是实现了原位条件下样品(包括流体、熔体、矿物等)的直接观测。同步辐射光源具有穿透能力强、高强度和高度准直特性,同步辐射技术与高温高压实验结合,进一步强化了实验地球化学研究的手段。

基于此,实验地球化学实现了对熔体—流体性质的原位观测(Shen and Keppler, 1997; Bureau and Keppler, 1999; Le Losq *et al.*, 2017; Romanenko *et al.*, 2018)、深部地幔乃至核幔边界附近的氢—碳—氮—硅—铁等同位素分馏(Yang *et al.*, 2014a; Dalou *et al.*, 2015; Li *et al.*, 2016b; Romanenko *et al.*, 2018; Dalou *et al.*, 2019)乃至深部地幔和地核的元素性质以及物相组成(Hart and Dunn, 1993; Li and Agee, 1996; Corgne and Wood, 2002; Lin *et al.*, 2007; Polyakov, 2009; Zhang *et al.*, 2014)等的直接制约。此外,人们还对全球尺度上的一些大型地质事件和类地星体组成等方面进行了直接研究,包括软流圈的成因(Mirerdel *et al.*, 2007; Green *et al.*, 2010)、地核增生和核幔分异(Wood *et al.*, 2006)、地球上挥发分的起源(Li *et al.*, 2016a; Yang *et al.*, 2016)、核幔边界的

水—岩反应(Yagi, 2016; Hu *et al.*, 2017; Liu *et al.*, 2017)、早期地球的热演化(Watson and Harrison, 2005; Trail *et al.*, 2011)以及火星地核的成分(Fei and Bertka, 2005)。得益于分析技术的不断发展和突破,实验地球化学研究在精细度方面有了很大进步,特别是早期一些定性研究逐渐实现了量化。这使得一些早期相对粗糙的模型逐渐精细化,同时一些量化的模型也刷新了原先的认识,对于准确认识地球的组成、性质和演化提供了重要依据。

实验地球化学早期关注地球物质的化学组成和性质,为传统地球化学研究提供理论依据和服务。但近30年来,实验地球化学作为一门独立学科的趋势越来越明显且与地球科学其他学科的交叉性不断增强,特别注重从多学科角度综合研究地球内部作用机制和运行规律。从元素赋存、储量和分配等性质上,实验地球化学与矿床学、岩石学、地球化学、地球物理、矿物学和流变学等学科间的联系进一步加强,相互间界限也变得更加模糊。事实上30多年前,Holloway and Wood (1988)在实验地球化学经典专著《Simulating the Earth: Experimental Geochemistry》中,就已经把变质岩体系中固体—固体和固体—流体间的反应、流体的 P - V - T 轨迹及其物理化学性质、火成岩体系中熔体—矿物和熔体—流体间的相容性、元素赋存和元素分配以及包括黏滞度、元素扩散、晶体生长和弹性性质等在内的地球物质物理性质包含在实验地球化学范畴内。近30年来,实验地球化学更是通过深入研究矿物、岩石、流体、熔体中元素和同位素在高温高压下的行为和性质(乃至矿物在岩浆中的可溶性等: Li *et al.*, 2020),对地球和其他星体的成因、组成、演化、动力学等方面开展系统研究。特别是根据挥发分、微量元素和同位素等的实验数据,对小到纳米尺度和大到行星尺度的诸多地质事件进行理论约束(Rubie *et al.*, 2004; Dasgupta and Hirschmann, 2006; Mirerdel *et al.*, 2007; Green *et al.*, 2010; Trail *et al.*, 2011; Armstrong *et al.*, 2019)。另一方面,实验测定的矿物化学成分数据,也为深入认识相关的导电性和流变强度等矿物物理和地球物理学研究提供了边界依据;对共存相间同位素分馏的认识,也为深刻理解天然地质样品演化和岩石成因等地学问题提供了重要基础。

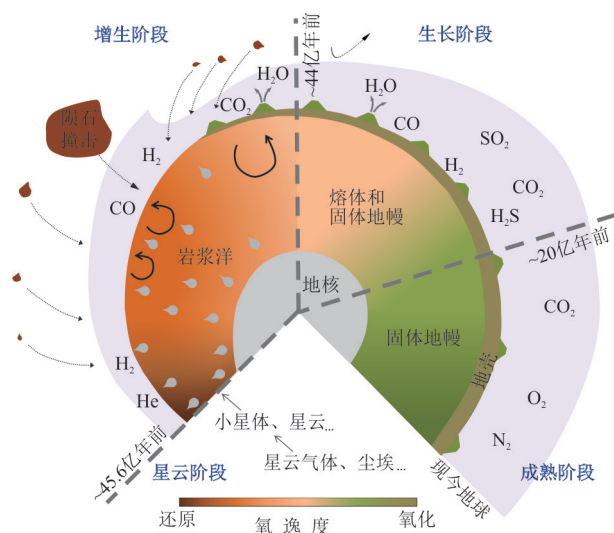


图 2 行星起源、挥发分和地球演化

Fig. 2 Planetary origin, volatiles and Earth's evolution

修改自 Gaillard and Scaillet(2004)

4 实验地球化学的前景展望

经过半个多世纪的积累,实验地球化学已经从最初比较单一的一种辅助研究手段,发展成为目前相对独立且极其强大的一门学科,其研究视野更是覆盖了从约 46 亿年前的地球诞生之初到当前、从地表到地下约 6 370 km 深度的地心以及从地球到月球和其他类地星体.地球自诞生之前的星云与行星初级阶段以至诞生后近 46 亿年的漫长地质演化过程中,一直伴随着剧烈的高温高压作用和物质反应(图 2).接下来的 10~20 年内,实验地球化学有望在以下方面发挥更为突出的作用:

4.1 深部地球和早期地球

深部地球是国际地学界常年持续关注的热点方向,向深部地球进军也是我国进入 21 世纪后的重要战略目标.人们能直接取样的地球内部深度一般局限于 200 km 内,只在极个别情况下会零星发现一些更深起源的天然地质样品.实验地球化学由于自身的特点,在深部地球研究上具有突出的优势.与早期研究集中在地壳和浅部地幔等较浅深度不同,近 30 年来实验地球化学逐渐把目光投向了深部地幔和地核.另外一个非常重要的研究方向是早期地球,围绕地球的早期诞生、分异过程和演化模式,主要发生在 40 亿年前甚至 45 亿年前.地质记录上地球化学组成可靠的早期地质样品极其缺乏,因而这个方向上实验地球化学也可以发挥出显著优势.

相关重要的研究工作包括,深部地球和早期核幔

分异过程中的元素分配和同位素分馏、俯冲带的元素迁移(包括成矿金属)和物质循环、地球上重要元素(包括氢、碳、金、硫等)的起源等.一些实验工作对这些方面进行了初步探索,包括微量元素的分配(Hart and Dunn, 1993; Li and Agee, 1996; Corgne and Wood, 2002; Corgne *et al.*, 2005)、Si/Fe/N 等稳定同位素的分馏(Georg *et al.*, 2007; Polyakov, 2009; Shahar *et al.*, 2009; Li *et al.*, 2016b; Dalou *et al.*, 2019),俯冲带条件下的元素迁移和物质循环(Keppler, 1996; Manning, 2004; Kessel *et al.*, 2005)以及一些元素的早期起源和地球演化过程等(Wood *et al.*, 2006; Li *et al.*, 2016a; Yang *et al.*, 2016; Armstrong *et al.*, 2019; Keppler and Golabek, 2019).但相关工作很不系统化、不同研究数据不一致性很大或者一些重要体系未曾涉及,这也影响了对重大地质作用过程典型地质过程的认识(比如克拉通破坏中元素迁移和金属成矿:汪在聪等, 2021).另外,下地幔底部的水-岩反应和物质结构(包括大型低剪切波速区,即 LLSVP)是学术界近来研究热点,物质成分变化和相关的化学反应可能是认识核幔边界的重要途径(Irifune *et al.*, 2010; Zhang *et al.*, 2014; Mao and Mao, 2020).

4.2 挥发分和地球宜居性

氢、碳、氮、硫、卤素、惰性气体等挥发分,在地球生命起源和演化以及地表宜居性和环境变化方面扮演了重要的角色.这些挥发分很可能主要赋存在地球内部,并对深部地球的物理、化学和动力学性质有显著影响.挥发分是地球内部最活跃的物质,它们在地球内部不同储库间以及地球内部与地表的交换、循环和反应贯穿了地球内外,显著改变了地球的动力学过程、化学演化模式、物理学性质、岩石成因、岩浆活动、板块构造作用、成矿元素迁移与富集等诸多方面,并直接影响了大气圈、水圈和生物圈的形成与演化(Huppert and Woods, 2002; Saal *et al.*, 2002; Keppler, 2013; Holland and Turekian, 2014; Wallace *et al.*, 2015).通过高温高压实验,对挥发分在地球物质中的赋存、储量、性质、效应和起源开展研究,是当前实验地球化学乃至整个固体地球科学的前沿领域.

已有工作表明,挥发分在地球内部的储量可能远超地表总量,大气圈和水圈是地球内外不同储库间动态交换的产物.简单说,氢主要储存在地幔硅酸盐矿物中,其赋存显著受温度、压强、氧逸度、共存相等因素的影响(Kohlstedt *et al.*, 1996; Yang,

2012, 2015, 2016; Yang *et al.*, 2014b, 2016; Liu and Yang, 2020; Jiang *et al.*, 2022; Zhang *et al.*, 2022); 氮主要储存在地幔硅酸盐矿物中, 其赋存受氧逸度影响很大 (Li and Keppler, 2014; Li *et al.*, 2014; Yoshioka *et al.*, 2018); 碳和硫在地幔硅酸盐矿物中的储存能力几乎可以忽略, 其在地幔中的赋存形式主要是碳酸盐、金刚石、石墨和硫化物等独立相 (Keppler *et al.*, 2003; Shcheka *et al.*, 2006); 卤素和惰性气体在地幔硅酸盐矿物中的储量也极为可观 (Beyer *et al.*, 2012; Shcheka and Keppler, 2012; Roberge *et al.*, 2015). 俯冲洋壳是将地表挥发分再循环到地球内部的重要载体, 但最近高温高压实验、岩石学与地球化学分析以及地球物理电磁探测的联合研究发现, 俯冲洋壳向深部地球中再循环水的能力可能很有限 (Liu *et al.*, 2021). 但已有工作亟待进一步加强, 特别是挥发分在地球内部的准确储量、被俯冲板块再循环的能力以及所产生的物理化学效应等方面.

4.3 行星形成演化实验模拟

自上世纪 60 年代末期人们开始对太阳系其他星体进行探测以来, 行星科学吸引了学者们的浓厚兴趣; 特别是进入 21 世纪后, 随着包括我国在内多个国家对于月球和火星探测计划的实施, 行星科学进入了一个高速发展时期 (Taylor, 1982; Shirley and Fairbridge, 1997; Board and Council, 2012). 行星物质的地球化学组成以及相关星体的演化, 是行星科学研究中极为重要的一个研究方向. 由于行星样品获取难度极大且花费极高, 实验地球化学为探索其他星体结构、组成和演化以及行星科学的发展提供了独特作用 (Syono, 1992; Smith, 1997; Fei and Bertka, 2005; Mao and Hemley, 2007). 实验地球化学数据能应用于其他类地星体, 其主要原因是矿物、岩石、流体 (包括熔体) 等物质的很多行为和性质 (比如元素分配和同位素分馏等) 属于它们本身固有, 并不依赖于所用实验材料的来源和产地. 因此, 无需其他星体样品, 就可以通过实验模拟开展研究.

根据研究性质, 这些工作主要包括: (1) 类地行星的热结构和化学组成, 其中较为突出的是对火星内部物质组成、地温梯度和化学成分的实验制约 (Fei *et al.*, 2000; Fei and Bertka, 2005); (2) 微量元素的分配及对核幔分异过程的制约, 其中较为突出的是对月球、金星和火星等星体深部作用过程的研

究 (Gaetani and Grove, 1997; Walter *et al.*, 2000; Draper *et al.*, 2003; Agee and Draper, 2004; McCanta *et al.*, 2004; Dygert *et al.*, 2020); (3) 挥发分 (包括挥发性亲铁元素) 的深部赋存和在月球及类地行星上的起源, 包括通过 O 的分配对地核诞生和组成以及 H、C、S、卤素等挥发分在月球、金星和火星等星体上赋存和起源的约束 (Rubie *et al.*, 2004; Tsuno and Dasgupta, 2015; Li *et al.*, 2016a; Yang, 2016; Yang *et al.*, 2016; Keppler and Gola-bek, 2019; Richter, 2019; Liu and Yang, 2020). 已有的实验工作相对比较零星化, 随着月球、火星和小行星带等地外星体样品的采集和地球化学数据的积累, 相关的实验地球化学研究必然进一步加强.

5 结论

长期以来, 实验地球化学为地球和行星科学的发展, 提供了重要的基础数据和理论框架. 可以说, 实验地球化学弥补了传统地球化学研究中的不足, 在某种程度上促进了地球科学 (和行星科学) 的发展进程. 实验地球化学在地球科学研究中所起的作用越来越显著, 但也伴随着一些不容忽视的问题. 主要包括: (1) 地球和其他类地星体并不是理想体系, 而已有的很多实验研究过于简单化和理想化, 实验室数据到天然地质体系的过渡和应用方面需要谨慎对待; (2) 实验地球化学提出的很多模型, 都只是针对地球和其他星体的一种可能性或假设, 事实上不少已有模型都被后来研究证实不准确或者是错误的; (3) 新的理论体系和新的框架模型迫切需要建立, 实验地球化学研究需要在综合考虑整个地球科学的系统性上进一步加强, 这也是由地球科学本身的系统性特征所决定的; (4) 相对于很多未被研究的体系, 目前已经开展的研究工作涉及面存在严重不足, 对很多元素和同位素的行为、性质和效应的认识仍有诸多未知之处.

相较于国际上的研究现状, 我国实验地球化学虽然近些年来发展较快, 但依然存在显著差距, 与国外发达国家相比有很大的提升空间, 在国际上相关领域仍然较为落后. 我国实验地球化学研究, 无论是在仪器平台搭建还是人才培养方面, 都亟需进一步加强, 由此才能为全面推进国内地学发展起到更重要的作用. 高温高压实验技术和现代分析测试技术的进一步改进和完善, 以及新的原创性理论的提出和标志性成果的出现, 也有望对实验地球化学

的发展注入新的活力,这也是国内相关研究领域有望取得新突破的地方。

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