

磁铁矿-磷灰石型铁矿的实验模拟研究进展与展望^{*}

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摘 要 磁铁矿-磷灰石(IOA)型铁矿, 或称基鲁纳型(Kiruna)铁矿, 或称陆相火山岩型铁矿, 在时空上常常与碱性-钙碱性的(次)火山岩有着紧密联系。该类型矿床在世界范围内广泛产出, 发育特征的磁铁矿-磷灰石-阳起石矿物组合, 但其成因还存在广泛争议。文章介绍并归纳了成岩成矿实验在 IOA 型矿床相关的岩浆原生过程方面取得的最新进展, 包括液态不混溶作用、岩浆磁铁矿-气泡悬浮模式和阳起石岩浆成因的实验验证, 探讨了磁铁矿以及磷灰石通过液态不混溶作用和气泡悬浮完成超常富集成铁矿浆的可能性。在此基础上, 指出了相关实验目前尚存在的问题及未来的研究方向。

关键词 地球化学; 磁铁矿; 磷灰石; 阳起石; 基鲁纳型铁矿; 陆相火山岩型铁矿; 实验; 岩浆过程

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Progress and prospective of experimental research on iron oxide apatite ore deposits

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Abstract

Iron oxide-apatite ore deposits (IOA), also referred to as Kiruna-type deposits or volcanic type iron deposits, occur in a number of places all over the world and are characterized by magnetite, apatite and actinolite mineral assemblages, and are often spatially and temporally related to alkaline, calc-alkaline (sub-)volcanic rocks. The origin of IOA ore deposits is still controversial, with two main opinions being magmatic or hydrothermal origins. However, it is difficult to get further constraints on the origins of IOA ore deposits through traditional research methods which are mainly based on description. This paper aims to present a detailed review of the recent achievements and problems in high-temperature and high-pressure experimental study of the magma processes of IOA ore deposits, which include experimental studies of liquid immiscibility, magnetite-bubble flotation, and igneous origin of actinolite in IOA ore deposits, and discusses the possibility that “ore-magma” was either formed by liquid immiscibility or by magnetite-bubble flotation. Based on these summaries, this paper points out the existing problems related to the formation of IOA deposits and the further research directions.

Key words: geochemistry, magnetite, apatite, actinolite, Kiruna-type deposits, volcanic type iron deposits, experiment, magma processes

精细刻画成矿过程和机制是当前成矿学的发展趋势之一。由于自然系统的复杂性, 导致地质现象

和地球化学数据常常具有多解性, 所以以描述为主要内容的传统矿床学研究在这个方面则显得有些

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“力不从心”。因此,以成岩成矿过程及物理化学条件为主要模拟对象的高温高压实验,能够通过控制变量来寻找矿床成因关键控制因素(Lukkari et al., 2007; Lledo et al., 2008; Webster et al., 2009; Song et al., 2016; Derrey et al., 2017; Hou et al., 2017; 2018)。这不但是解决与成矿作用相关的一系列关键科学问题的重要途径,也是矿床学进一步发展的内在要求。

磁铁矿-磷灰石(Iron Oxide-Apatite, 简称 IOA)型铁矿,国际上也称之为 Kiruna 型铁矿(基鲁纳型),在中国也称为陆相火山岩型铁矿或者“玢岩铁矿”。此类型铁矿在时空上常与中酸性火山岩-次火山岩密切相关,是高品质铁矿石的主要来源之一。矿石中除了磁铁矿和磷灰石以外,还含有阳起石,形成特征的磁铁矿-磷灰石-阳起石矿物共生组合。IOA 型铁矿在全球广泛产出,例如中国东部宁芜和庐枞 2 盆地、新疆阿尔泰地区、伊朗 Bafq、瑞典 Kiruna 矿集区、秘鲁 Marcona 和智利 El Laco 等地(Naslund et al., 2003; Hou et al., 2011; Jonnson et al., 2013)。该类型铁矿一直是国内外学术界关注的热点,但其成因至今还存在很大争议。针对其成因,前人提出的模式主要包括岩浆和热液 2 种,而岩浆成因除了传统的铁矿浆不混溶模式以外(Naslund et al., 2003),还有磁铁矿气泡悬浮富集模式(Knippling et al., 2015a),热液成因模式则有热液交代和流体出溶沉淀成矿 2 种(Hildebrand, 1986; 中国科学院地球化学研究所, 1987; 赵永鑫, 1993; Gleason et al., 2000; Sillitoe et al., 2002; Dare et al., 2014)。值得指出的是,岩浆模式中的 2 种类型并不排斥岩浆期后热液过程,并且是将二者统筹考虑的。由于对于岩浆系统来说,热液过程总是相对晚于岩浆过程的,而且部分 IOA 型矿床并不发育与成矿有关的热液蚀变作用(如阿巴宫),所以先搞清楚岩浆期发生了什么,即厘清 IOA 型矿床的原生过程(primordial process)是建立成矿模式的前提。具体来说,查明岩浆期究竟是否有铁质的(超常)富集,是否有铁矿浆的出现,挥发分在岩浆作用中发挥了怎样的作用是解决该类型矿床成因的关键。可是由于岩浆期后热液作用的叠加改造,导致自然样品很难完整保留岩浆过程的相关信息。正因如此,本文即从成岩成矿实验的角度,聚焦 IOA 型矿床形成的原生过程,侧重介绍岩浆过程的相关实验,对国内外相关研究予以归纳总结,以期帮助我们所有人对这种矿床的成岩成矿过程有更加深入的理解。

1 液态不混溶作用实验研究

IOA 型矿床具有独特的矿体形态和矿石结构构造,其块状矿石呈现出类似火山岩的特征(Park, 1961; Henríquez et al., 1978; 宋学信等, 1981; Nystrom et al., 1994; Henríquez et al., 1998; Naslund, 2002; Chen et al., 2010),比如智利拉科铁矿产出的致密块状矿石具有流动构造,树枝状和绳状构造等,以及可以指示层面的定向排列的气孔;磁铁矿熔岩流层面平行火山斜坡,构成完整的熔岩单元,即顶部气孔发育,往下气孔减少等。在中国宁芜盆地的姑山铁矿不但可以观察到这些类似的现象,还可以观察到斑状、菊花状和熔渣状矿石等(翟裕生等, 1992)。因此,国内外学者早期的研究根据这些野外现象提出了铁矿浆贯入的成矿模式,即致密块状铁矿石直接结晶于纯的铁氧化物熔体(Park, 1961; Frietsch, 1978; 宁芜研究项目编写小组, 1978; 常印佛等, 1991; 翟裕生等, 1992)。由于只有在简单体系不混溶实验中才能获得纯的氧化物熔体,如 $\text{FeO-Fe}_2\text{O}_3\text{-SiO}_2\text{-H}_2\text{O}$ 体系(Gibbon et al., 1967), Fe-C-O 体系(Weidner, 1982)和铁橄榄石-石榴石-石英体系(Lester et al., 2013),所以将铁氧化物熔体作为铁矿浆的模式遭到了很多研究者的反对。从热力学的角度来说,如果要在岩浆温度下形成熔体,则必须有足够的网络形成子(成网离子, network former)形成熔体结构来“溶解”或“承载”网络修饰子(变网离子, network modifier)。虽然 Fe^{3+} 是网络形成子, Fe^{2+} 是网络修饰子,但是在岩浆温度下,显然前者不足以聚合成熔体来“溶解”后者。具体的表现就是在 $\text{FeO-Fe}_2\text{O}_3\text{-TiO}_2$ 三元系中,不论铁的价态和钛含量的高低,铁钛氧化物的熔点都要高于玄武岩的液相线温度($\sim 1200^\circ\text{C}$; 图 1a)。所以铁氧化物熔体在 IOA 矿床的形成过程中,甚至在所有的岩浆系统成岩成矿过程中,存在的可能性极低(Lindsley et al., 2017)。

既然如此,考虑到矿石中往往不同程度富磷,那么铁矿浆会不会是以熔融态的磷灰石-磁铁矿的形式存在呢? 实际上,岩浆体系的液态不混溶实验所产生的富铁相基本都是富磷的(Ryerson et al., 1978; Naslund, 1983; 李九玲等, 1986; 袁家铮, 1990; Longhi, 1990; Hess et al., 1975; Rutherford et al., 1974; Dixon et al., 1979; Philpotts et al., 1983; Bogaerts et al., 2006; Veksler et al., 2007; Charlier et al., 2012),但

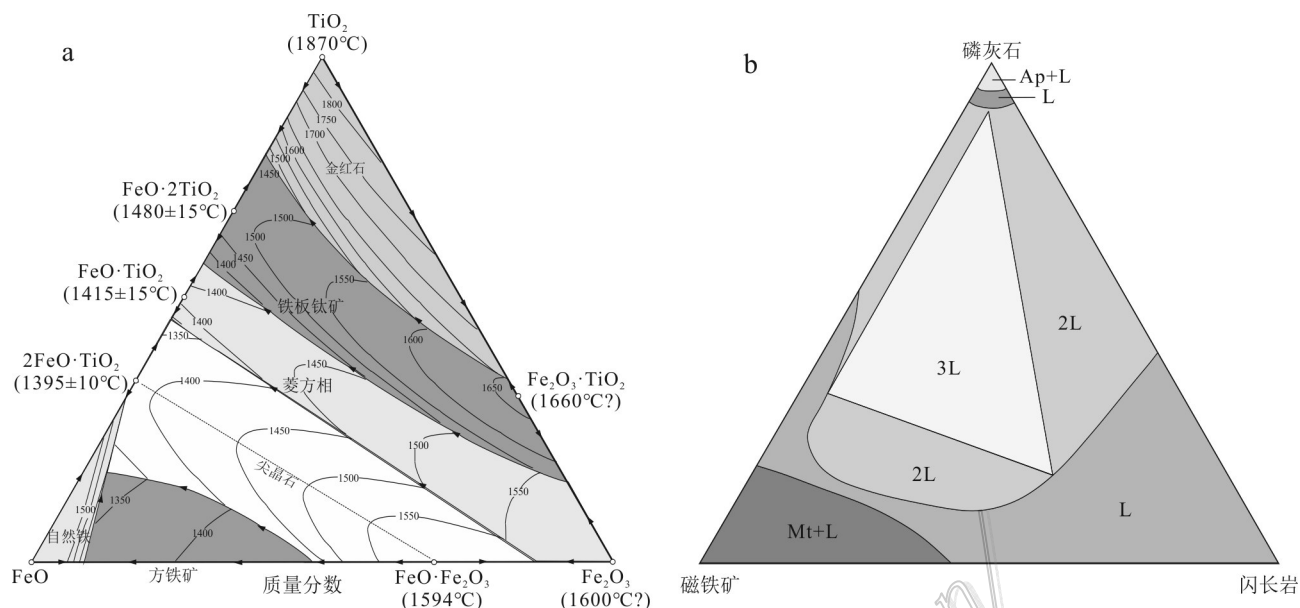


图1 常压下(1 atm)FeO-Fe₂O₃-TiO₂体系相图(a, 据 Taylor, 1963)及常压下(1 atm)磁铁矿-磷灰石-闪长岩体系相图(b, 据 Philpotts, 1967, 实验温度为 1420°C)

Fig. 1 Liquidus (melting) diagram for oxides in the system FeO-Fe₂O₃-TiO₂ at one atm (a, modified after Taylor, 1964) and liquidus (melting) diagram in the system diorite-magnetite-apatite at one atm (b, modified after Philpotts, 1967, experiments were carried out at 1420°C)

是富铁相除了富磷以外,还含有至少20%的SiO₂ (Wang et al., 2017),此成分与磁铁矿+磷灰石成分仍相去甚远。虽然 Philpotts (1967)的实验(图1b)在磁铁矿-磷灰石-闪长岩体系中获得不混溶的磁铁矿+磷灰石成分熔体,但是实验温度高达1420°C,依然远高于玄武岩的液相线温度,所以类似铁氧化物熔体,很难将实验结果外推到地质环境中。最近,Hou 等(2018)针对IOA成矿体系,进行了含氟、水和硫的复杂体系实验,温度和压力条件是最接近自然界可发生的条件,即压力为100 MPa,温度为1000~1040°C。在不同条件下,实验分别获得了2种基本不含硅的不混溶的富铁磷熔体,一种是基本接近矿石组分的Fe-Ca-P-O熔体;另一种是Fe-P-O熔体。这些熔体的矿物亲湿性都很好,实验产物中磁铁矿和磷灰石都优先被其包裹(图2),形成的熔体-矿物混合物的化学成分可以解释不同品位矿石的形成。特别是Fe-P-O熔体,与Xie 等(2019)在拉科铁矿块状矿石中磁铁矿粒间发现的铁的磷酸盐(Fe phosphate)的成分基本是一致的,与Mungall 等(2018)的实验获得的铁的磷酸盐熔体成分也是非常类似的。因此,上述2种熔体与磁铁矿和磷灰石以不同比例的混合形成铁矿浆可以解

释绝大部分IOA矿石的形成。值得指出的是,实验结果还表明,氟是非常重要的挥发分,它不但可以降低

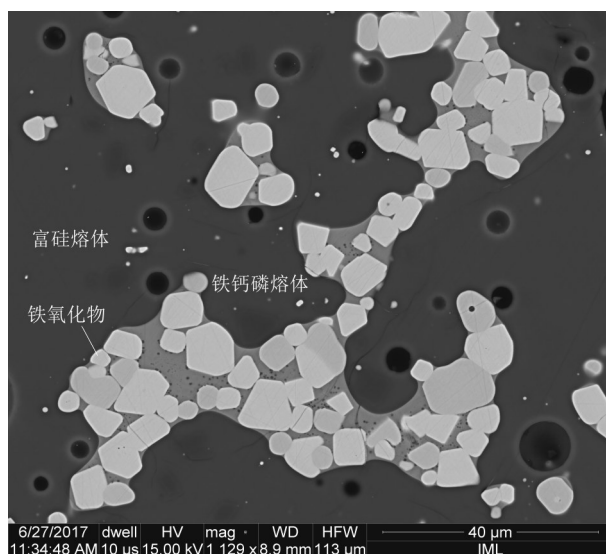


图2 铁氧化物被Fe-Ca-P熔体包裹形成铁矿浆(1020°C,具体实验细节详见 Hou et al., 2018)

Fig. 2 Iron oxides preferentially enclosed by Fe-Ca-P liquid due to wetting property at 1020°C (Experiment, details see Hou et al., 2018)

低系统的固相线温度,延长岩浆演化的时间,更可以通过与熔体中铝的结合提高铁的活度,使不混溶富铁相中铁的含量进一步增大,所形成的氟磷灰石也与IOA型矿床的实际情况是一致的。另外一个重要发现是当不混溶作用发生后,水优先富集在富硅熔体中,这与前人推断的富铁熔体富水是不同的(Lester et al., 2013; Tornøse et al., 2016; 2017),但这恰好与岩浆岩广泛发育的热液蚀变是一致的。

2 岩浆磁铁矿-气泡悬浮模式及其实验证据

智利铁矿带中的IOA型矿床多数都是贫磷灰石的。因此,美国密歇根大学Adam Simon团队(Knipping et al., 2015a; 2015b; 2019a; Ovalle et al., 2018; Simon et al., 2018)针对其中的El Laco和Los Colorados矿床开展研究。根据矿床不同深度矿石的结构和化学成分、磁铁矿的核幔显微结构、地球化学成分及Fe-O同位素特征,提出了岩浆磁铁矿-气泡悬浮模

型。为了检验这个模型,Knipping等(2019b)使用含水和氧化的岩浆开展了岩浆脱气作用的模拟实验,通过对比250 MPa和250~150 MPa的等温(1050℃)减压实验,发现磁铁矿可以作为早期液相线矿物先结晶而出,同时由于压力降低而出现流体出溶产生的大量气泡。由于气泡对磁铁矿的亲湿性较好而附着在磁铁矿表面成核并生长,形成密度较小的、富含FeCl₂的磁铁矿-气泡对(magnetite-bubble pair)悬浮液而上升。依据这样的实验结果,他们提出了磁铁矿-气泡悬浮成矿模式(图3a~c),即总体密度较小的磁铁矿-气泡对悬浮液在浮力作用下,在岩浆房中不断上升而聚集;当达到一定位置后,因构造应力的变化形成水压致裂(hydraulic fracture),悬浮液迅速上升,并且在冷却过程中岩浆磁铁矿微晶外围叠加形成热液磁铁矿。这些悬浮液作为铁矿浆,为成矿奠定了重要的物质基础。这个模式可以解释矿石中的磁铁矿发育核幔环带结构,且内部磁铁矿的Ti/V等元素偏高的现象,也可以解释广泛发育的热液蚀变以及矿物中的高盐度包裹体。

然而,由于目前没有实验表明磁铁矿和磷灰石

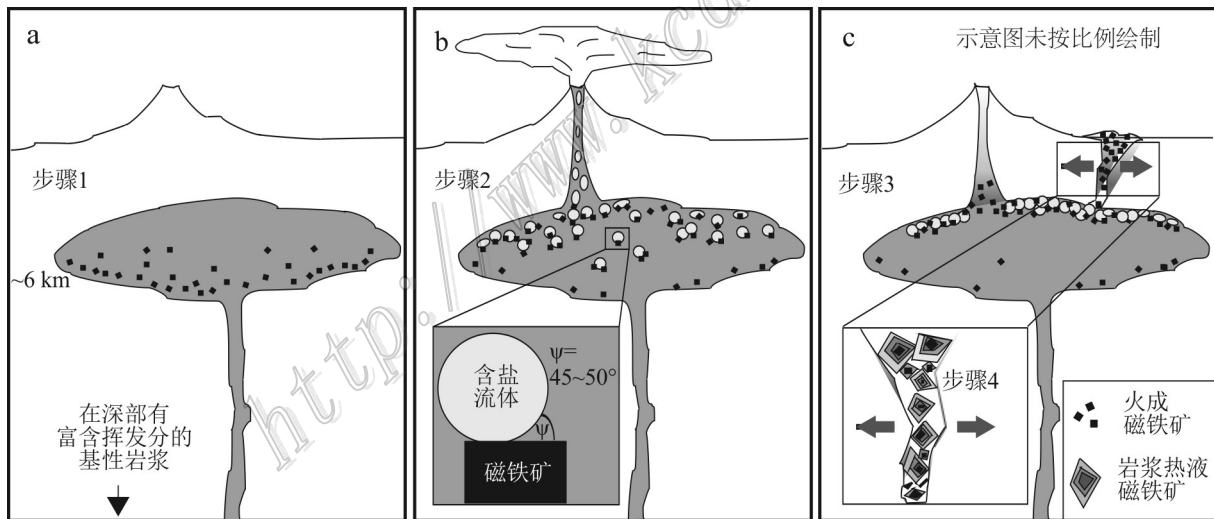


图3 基鲁纳型IOA矿床的磁铁矿-气泡对悬浮成矿模型简图(未按比例绘制;据Knipping et al., 2019b)

- a. 原生磁铁矿作为早期液相线矿物结晶,由于密度比硅酸盐熔体高,在岩浆房内下沉;b. 如果富盐流体因为压力降低而出溶产生气泡,由于磁铁矿亲湿性较好,这些气泡会附着在磁铁矿表面形成磁铁矿-气泡对;c. 磁铁矿-气泡对总体密度较小,便会在岩浆房内上升、聚合形成磁铁矿-流体悬浮液,如果拉张的构造应力产生了裂隙,悬浮液便会沿裂隙迅速上升直至地表,在低温和低压下,次生热液作用便会围绕原生磁铁矿叠加形成热液磁铁矿

Fig. 3 Schematic illustration of the magnetite-flotation model for Kiruna-type iron oxide-apatite deposits(after Knipping et al., 2019b)
a. Primary igneous magnetite crystallizes from silicate melt in a magma reservoir and should gravitationally settle owing to its higher density relative to melt. However, b. If saline fluid exsolves during decompression and bubbles nucleate on magnetite crystals owing to favorable wetting properties; c. Magnetite-bubble pairs form and buoyantly ascend, coalesce and separate as a magnetite-fluid suspension within the magma, and can escape the magma if extensional tectonic stress opens crustal fractures wherein secondary magmatic-hydrothermal magnetite can precipitate, at lower pressures and temperatures, and surround primary igneous magnetite crystals

会在含水岩浆中同时以最早的液相线矿物出现,故无法形成磁铁矿+磷灰石-气泡悬浮对。所以虽然这个模式可以用来解释一些贫磷灰石 IOA 型矿床的形成,但是仍然无法用来解释磁铁矿和磷灰石的同步富集,也无法解释 El Laco 块状磁铁矿矿石粒间铁的磷酸盐的产出(Mungall et al., 2018)。

3 阳起石的岩浆成因实验验证

阳起石为硅酸盐类矿物,它是透闪石中的镁离子2%以上被二价铁离子置换而成的矿物,所以比透闪石富铁,也是闪石系列中的一员。阳起石作为 IOA 型矿床中重要的矿石矿物组成之一,其成因对阐明成矿机制和构建成矿模式至关重要。然而,过去很多研究往往直接默认矿石中的阳起石为热液蚀变产物,类似矽卡岩矿床中的湿矽卡岩阶段形成的阳起石,从而默认它们为交代透辉石的产物,并由此推断 IOA 型矿床为热液成因。然而,在很多矿石中是观察不到阳起石交代透辉石的,阳起石常常以自型柱状或者纤维状与磁铁矿和磷灰石共生。在智利的一些 IOA 型矿床的矿区范围内,新鲜的火山熔岩和碎屑岩中可以看到自形的阳起石斑晶或者集合体(Lledo et al., 2008)。这些证据表明,至少有一部分阳起石,尤其是块状矿石中与磁铁矿和磷灰石共生的那一部分,很可能是从岩浆中直接结晶出来的。

因此,几十年来国外学者对不同成分的透闪石-阳起石系列开展过一系列的热稳定性上限(upper thermal stability)检验实验,以明确其能否在岩浆温度下稳定存在,从而检验其是否为岩浆结晶的产物。然而,绝大部分的实验初始物质都使用的是富镁端员透闪石(Jenkins et al., 1990, 1991; Welch et al., 1991; Chernosky et al., 1998),或者是人造的富铁阳起石端员成分(Ernst, 1966; Hellner et al., 1966; Jenkins et al., 2003)。目前只有 Lledo 等(2008)的实验初始物质($\text{Fe 值} = n(\text{Fe}) / (n(\text{Fe}) + n(\text{Mg})) = 0.5$)最为接近 IOA 矿床中比较中性的相对富镁阳起石的化学成分($\text{Fe 值} = 0 \sim 0.4$; Verkouteren et al., 2000)。实验表明,在 750~900℃ 的岩浆温度下,100~400 MPa 的压力范围内,相对富镁的中性阳起石是可以稳定存在的,从而证实了 IOA 矿床中在该成分范围内的阳起石是岩浆结晶矿物。值得指出的是,如图 4 所示,阳起石的稳定域(stability field)与富水安山质岩浆的温压条件(固相线和液相线之间的范围;Stern et al., 1975)存在明显的重叠。

这进一步表明 IOA 型矿床中常见的阳起石是可以从富水的安山质岩浆中直接结晶而成的。除此以外,如上所述,如果富水的安山岩通过液态不混溶作用形成铁矿浆的话,就可以很好地解释磁铁矿-磷灰石-阳起石为什么是 IOA 型矿床的典型共生矿物组合。

4 存在问题及未来研究展望

综上所述,相关实验为研究者理解 IOA 矿床形成的原生过程提供了重要的思路和理论基础,充分表明岩浆过程,包括分离结晶、液态不混溶、岩浆脱气等作用对 IOA 型矿床的形成至关重要。然而由于实验模拟本身受条件局限,会在一定程度上偏离真

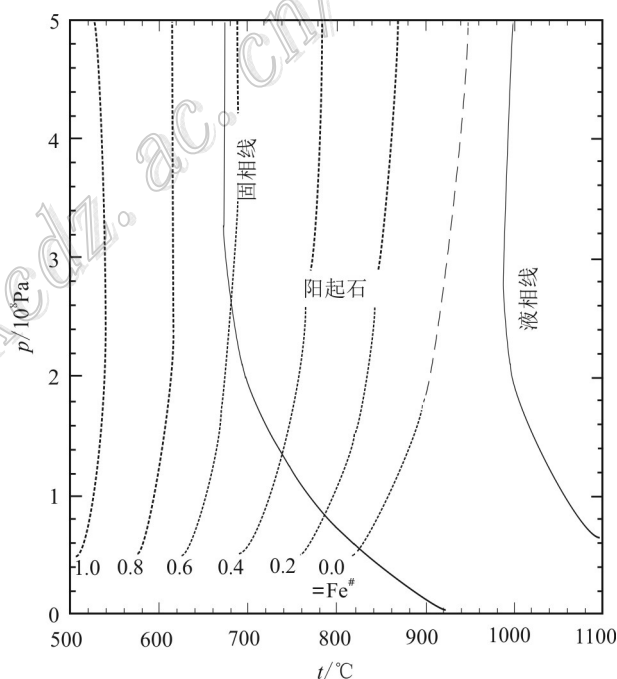


图4 富水条件下阳起石Fe值等值线 p - t 关系图(体系中存在单斜辉石、斜方辉石和石英,忽略低温下可能出现的镁铁闪石)。水饱和条件下英云闪长岩的固相线和液相线引自 Stern 等(1975)。贫Fe阳起石稳定域与安山岩熔融区域存在大范围重叠

Fig. 4 p - t diagram showing calculated isopleths of constant Fe-number in actinolite coexisting with clinopyroxene, orthopyroxene, quartz, and water (ignoring the probable appearance of cummingtonite at lower temperatures) compared with the water-saturated solidus and liquidus of tonalite (andesite) reported by Stern et al., 1975. The large extent of overlap in the upper thermal stability of Fe-poor actinolite and the onset of melting in an andesite should be noted

实的地质环境和过程。因此,相关实验还存在一些问题,仍然需要进一步加强工作。

(1) 在IOA成矿体系中需要进一步加强其他挥发分作用的研究,查明它们对岩浆阶段相平衡的影响,以及熔体-流体分配系数及其影响因素。例如,有证据表明岩浆演化晚期出溶的流体是一种富氯的高盐度流体,但是氯的作用仍不明确,所以需要进一步的实验来回答,含氯体系是否还会发生不混溶作用,是否还会产生Fe-P-O和Fe-Ca-P-O熔体? 如果会继续诱发液态不混溶作用的话,是否会改变相关元素比如Ca和P的分配系数? 能否产生相对贫Ca和P的熔体? 如果氯会抑制液态不混溶作用,那么它会对磁铁矿以及磷灰石的结晶产生什么样的影响? 岩浆演化到晚期氯又会如何影响出溶流体的成分? Fe在熔体-流体之间的分配及其影响因素的实验研究仍然是相当薄弱的。

(2) 在IOA成矿体系中需要进一步开放体系实验研究,查明岩浆脱气作用(去气作用;magma degassing)的具体影响。由于IOA型矿床多形成在浅成-超浅成的环境中,压力的降低,脱气作用在所难免,这将对岩浆的氧逸度等物理化学性质产生重要影响,比如氧逸度的变化不但会影响不混溶作用的发生(Naslund, 1983),也会影响磁铁矿的结晶(Botcharnikov et al., 2008)。此外,脱气作用对气泡-磁铁矿悬浮模式的可行性至关重要。因此,岩浆脱气作用实验需要与岩浆演化实验相结合,来查明在岩浆演化过程中,是否存在一个同时结晶磁铁矿和磷灰石并与脱气作用发生耦合的阶段。

(3) 除少量矿床以外,绝大部分IOA型矿床都是富磷的,甚至在某些矿床中可以形成独立的磷矿体。因此,笔者认为对IOA型矿床成因的考虑应该放在一个更为宽泛的范围来考虑,即为什么磁铁矿和磷灰石这2种密度不同、成分迥异的矿物总是产出在一起? 不但在IOA型矿床,在与斜长岩体型有关的铁钛磷灰岩(nelsonite; Chen et al., 2013),在与碱性岩有关的岩浆型磷矿(河北矾山; Hou et al., 2015),甚至在一般的全晶质的岩浆岩中,二者也往往是共生关系。除了液态不混溶作用可以同步富集磁铁矿和磷灰石以外, Tollari等(2006)的实验表明,岩浆中磷灰石和磁铁矿的溶解度是正相关的,也就是说一种矿物的结晶是会降低另一种矿物的溶解度的。所以不论怎样,针对IOA型矿床的成因,任何一种成矿模式的提出,都必须建立在将这个问题考虑在内的基础上,才是令人信服的。

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