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蚀变洋壳和俯冲带变质流体的Fe-Mg同位素组成

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摘　要： 贫碳酸盐的蚀变洋壳具有与新鲜洋中脊玄武岩一致的Mg同位素组成，说明低温和高温洋壳蚀变不会导致Mg同位素分馏．大别山港河和花凉亭的早期变质脉比榴辉岩具有偏高的δ^{56}Fe-δ^{26}Mg值，而且早期到晚期变质脉的δ^{56}Fe-δ^{26}Mg值逐渐降低．这些结果说明，在流体－岩石反应和流体演化过程中，Fe-Mg同位素发生了显著的分馏，且矿物溶解－再沉淀是同位素分馏的控制因素．相比洋中脊玄武岩，蚀变洋壳和变质脉具有相似或偏高的δ^{56}Fe-δ^{26}Mg值，说明蚀变洋壳脱水产生的流体富集重Fe-Mg同位素，不能解释弧岩浆岩的轻Fe/重Mg同位素组成．因此，弧岩浆岩异常的Fe-Mg同位素组成是熔体提取和富集^{54}Fe-^{26}Mg的蛇纹岩流体交代地幔楔两个过程共同作用的结果．

关键词： 铁－镁同位素；蚀变洋壳；榴辉岩；变质流体；流体演化；弧岩浆岩；地球化学．
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Fe-Mg Isotopic Compositions of Altered Oceanic Crust and Subduction-Zone Fluids

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Abstract: The origin of the light Fe and heavy Mg isotope enrichments in arc lavas remains unclear because of the lack of constraints on the Fe-Mg isotope compositions of altered oceanic crust (AOC) and metamorphic fluids in subduction zones. Carbonate-barren AOC has Mg isotope compositions similar to those of fresh mid-ocean ridge basalts, suggesting that low-to-high temperature alteration of oceanic crust by seawater and hydrothermal fluids results in limited Mg isotope fractionation. Fe-Mg isotope measurements show that the early omphacite-epidote veins have higher δ^{56}Fe and δ^{26}Mg compared to the host eclogites and that the δ^{56}Fe and δ^{26}Mg gradually decrease from the early omphacite-epidote through epidote-quartz to the late kyanite-epidote-quartz veins. These results indicate significant Fe-Mg isotope fractionation during fluid-rock interaction and fluid evolution due to the dissolution-precipitation processes of minerals in subduction zones. Compared to mid-ocean ridge basalts, the similar or higher δ^{56}Fe and δ^{26}Mg of AOC and metamorphic veins suggest that AOC-derived fluids are probably enriched in heavy Fe-Mg isotopes. Thus, contribution from AOC-derived fluids is unlikely to explain the light Fe and heavy Mg isotope compositions of arc lavas. We propose that the Fe-Mg isotope anomaly of arc lavas may result from a combination of prior melt depletion and addition of serpentinite-derived ^{54}Fe-^{26}Mg-rich fluids into the overlying mantle wedge.

Key words: Fe-Mg isotopes; altered oceanic crust; eclogite; metamorphic fluids; fluid evolution; arc lavas; geochemistry.

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0　引言

在板块汇聚边缘，俯冲板片脱水或熔融产生流体或熔体，交代上覆地幔楔，引发地幔楔部分熔融，产生弧岩浆岩.板片熔流体是俯冲带壳－幔相互作用的重要媒介，会导致弧岩浆岩相比洋中脊玄武岩富集大离子亲石（如K、Rb、Sr和Ba）和轻稀土（如La和Ce）元素，亏损高场强元素（如Nb和Ta）（Elliott *et al.*，1997）.板片熔流体的主要源区为沉积物、蚀变洋壳和蛇纹岩（Schmidt and Poli，1998）.对板片熔流体来源的认识将有助于阐明壳－幔相互作用的过程以及俯冲带板片物质进入地幔楔的方式.

除了微量元素和放射成因（如Sr-Nd-Pb）同位素，金属Fe-Mg稳定同位素也能制约板片熔流体的性质和来源.相比亏损地幔，原始弧岩浆岩（$Mg^{\#}\geqslant 0.60$）具有较高的δ^{26}Mg（－0.35‰～0.06‰；Teng *et al.*，2016；Li *et al.*，2017）和较大的δ^{56}Fe变化范围（－0.15‰～0.14‰；Dauphas *et al.*，2009；Nebel *et al.*，2013，2015；Foden *et al.*，2018）.原始弧岩浆岩的轻Fe/重Mg同位素组成被认为是富集^{54}Fe-^{26}Mg的板片熔流体交代弧下地幔楔所致（Nebel *et al.*，2015；Sossi *et al.*，2016；Teng *et al.*，2016）.Fe-Mg-Sr-Nd-Pb-Hf同位素联合模拟显示，沉积物来源的熔流体不能解释原始弧岩浆岩富集轻Fe/重Mg同位素的特征（Nebel *et al.*，2015；Teng *et al.*，2016；Foden *et al.*，2018）.因此，蚀变洋壳或蛇纹岩可能是富集^{54}Fe-^{26}Mg板片熔流体的主要来源（Chen *et al.*，2016；Debret *et al.*，2016；Sossi *et al.*，2016；Teng *et al.*，2016）.然而，由于不清楚蚀变洋壳和变质流体的Fe-Mg同位素组成以及变质流体形成和演化过程中Fe-Mg同位素的分馏行为，极大地制约了我们对原始弧岩浆岩Fe-Mg同位素异常的理解.针对上述问题，本文综述了东太平洋IODP 1256钻孔蚀变洋壳以及大别山超高压榴辉岩和变质脉的Fe-Mg同位素的研究.

1　蚀变洋壳

蚀变洋壳样品取自位于东太平洋的IODP 1256钻孔.洋壳的年龄约为15 Ma，是在洋中脊超快速扩张时形成（200 mm/a）.IODP 1256钻孔恢复了迄今为止最为完整的洋壳，包括火山岩、过渡带、席状岩墙和侵入岩等4个部分（图1）.低－高温蚀变矿物的出现和高度变化的δ^{18}O（图1a，1b）显示，洋壳经历低温海水和高温热液蚀变作用.我们精心挑选的44个蚀变洋壳样品涵盖了4个不同的部分.Mg同位素分析结果显示，除了一个样品具有偏高的δ^{26}Mg，其他样品具有相对均一的δ^{26}Mg（－0.36‰～－0.14‰）（图1c）.

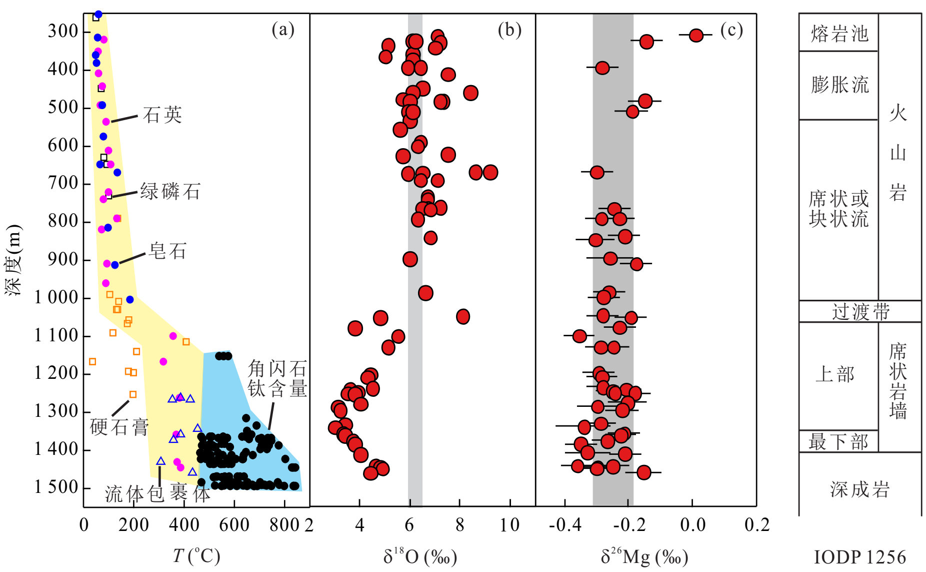

图1　IODP 1256钻孔洋壳的蚀变温度(a)，δ^{18}O(b)和δ^{26}Mg(c)的空间变化

Fig.1　Down-hole variations in alteration temperatures，δ^{18}O，and δ^{26}Mg of oceanic crust from IODP site 1256

蚀变温度、O和Mg同位素数据引自Alt *et al.*（2010）、Gao *et al.*（2012）和Huang *et al.*（2015）.灰色条带表示新鲜洋中脊玄武岩的O和Mg同位素组成（Harmon and Hoefs，1995；Teng *et al.*，2010）

2 大别山榴辉岩和变质脉

大别山超高压变质带位于大别—苏鲁超高压造山带的西南部(Zheng *et al*.,2003).榴辉岩和变质脉采自大别山港河和花凉亭.根据矿物组合,变质脉分为绿辉石—绿帘石、绿帘石—石英和蓝晶石—绿帘石—石英脉.详细的岩相学、矿物学和地球化学特征请见Guo *et al*.(2012,2013,2014,2015).两地榴辉岩含有硬柱石脱水后形成的绿帘石+蓝晶石+石英柱状集合体,且它们的模式丰度向脉体方向逐渐降低;同时,在蓝晶石中发现柯石英;另外,榴辉岩与变质脉具有相似的Sr同位素组成.最后,榴辉岩具有系统变化的主微量元素含量.这些证据说明,初始流体是硬柱石在超高压变质阶段脱水产生,然后与榴辉岩反应,溶解其中的矿物(如绿帘石、蓝晶石、石英和绿辉石等),最后形成溶质富集的成脉流体.绿辉石—绿帘石脉最早从成脉流体中固结形成,接着形成的是绿帘石—石英脉,最后形成的是蓝晶石—绿帘石—石英脉.

图2和图3总结了大别山港河和花凉亭榴辉岩、变质脉和单矿物的Fe-Mg同位素分析结果.如图2所示,相比远离脉的榴辉岩,靠近脉的榴辉岩具有相似或略微偏轻的Fe-Mg同位素组成;相比榴辉岩,绿辉石—绿帘石脉富集重Fe-Mg同位素;从早期绿辉石—绿帘石脉到晚期蓝晶石—绿帘石—石英脉,δ^{56}Fe和δ^{26}Mg值逐渐降低.如图3所示,相比石榴石,绿辉石、绿帘石和多硅白云母富集重Fe-Mg同位素.

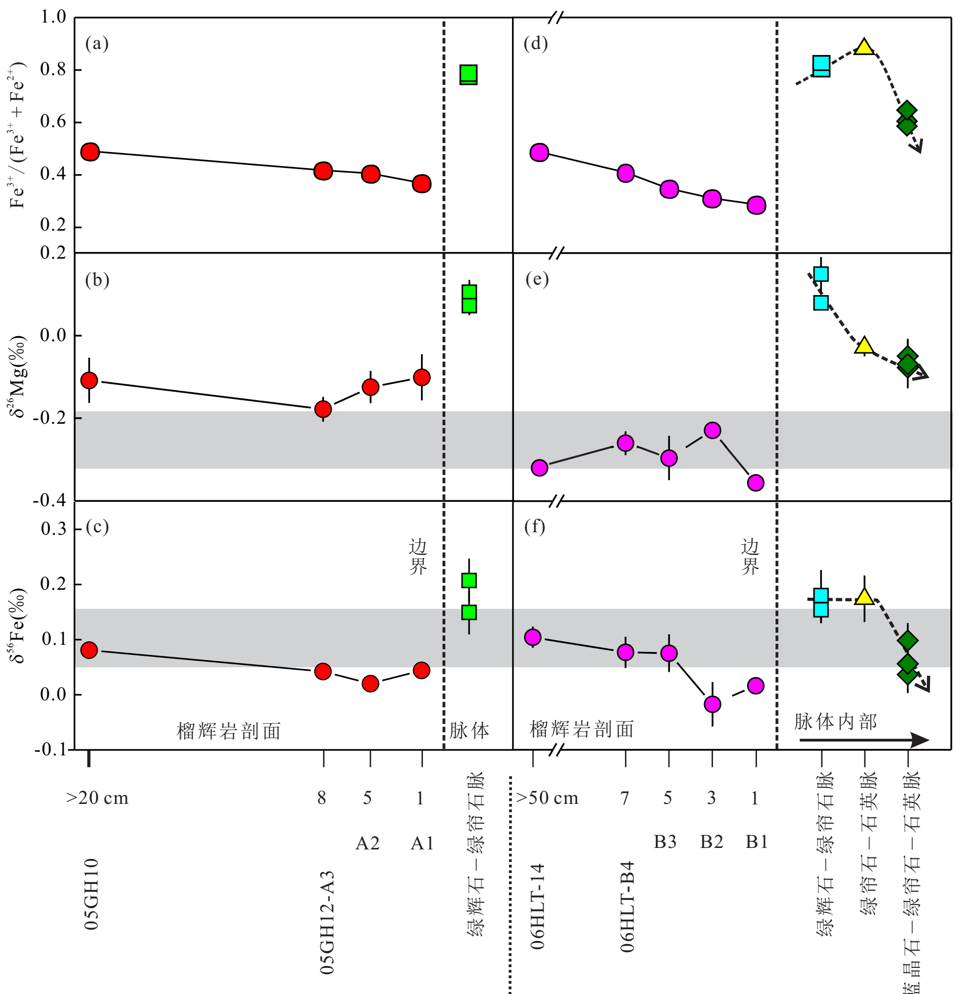

图2 大别山港河和花凉亭榴辉岩和变质脉的$Fe^{3+}/\Sigma Fe$、δ^{26}Mg和δ^{56}Fe变化

Fig.2 $Fe^{3+}/\Sigma Fe$, δ^{26}Mg, and δ^{56}Fe in ecoligites and veins at Ganghe and Hualiangting in the Dabie orogen

据Huang *et al*.(2019).灰色条带表示新鲜洋中脊玄武岩的Fe-Mg同位素组成(Weyer and Ionov,2007;Teng *et al*.,2010;Nebel *et al*.,2013)

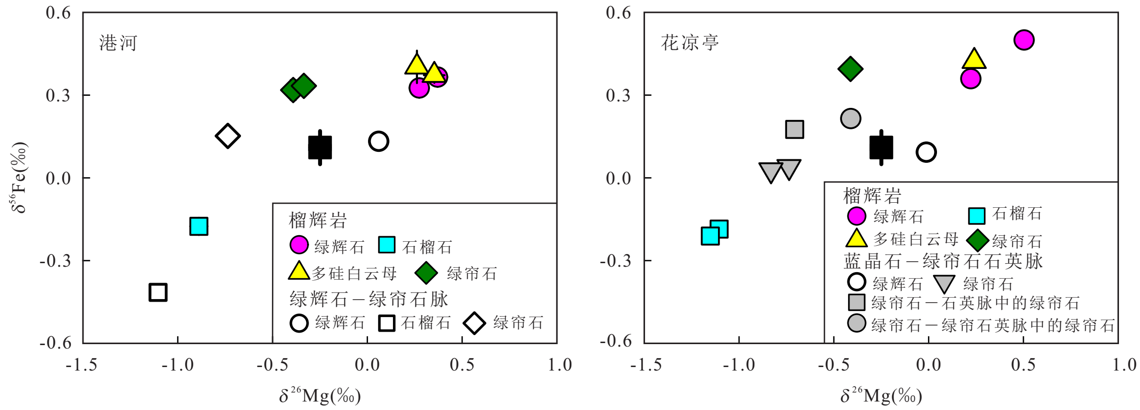

图 3　大别山港河和花凉亭超高压榴辉岩和变质脉中矿物的Fe-Mg同位素组成

Fig.3　δ^{26}Mg and δ^{56}Fe of minerals from ecoligites and veins at Ganghe and Hualiangting in the Dabie orogen

据Huang *et al*.(2019).黑色正方形表示新鲜洋中脊玄武岩的Fe-Mg同位素组成(Weyer and Ionov,2007;Teng *et al*.,2010;Nebel *et al*.,2013)

2.1　流体–岩石相互作用过程中的Fe-Mg同位素分馏

根据绿片岩到榴辉岩具有相似的Fe-Mg同位素组成，前人提出玄武质岩石脱水不会产生Fe-Mg同位素分馏(Wang *et al*.,2014;El Korh *et al*.,2017;Inglis *et al*.,2017).但是，我们发现绿辉石－绿帘石脉比围岩榴辉岩具有较高的δ^{56}Fe和δ^{26}Mg(图2)，说明流体－岩石反应能够导致显著的Fe-Mg同位素分馏.岩石学和地球化学证据证实，榴辉岩中硬柱石在～3.0 GPa和～670 ℃条件下，脱水产生初始流体，初始流体与榴辉岩反应，溶解其中的绿辉石、绿帘石、蓝晶石、石英、金红石、锆石和磷灰石等矿物，迁移大量的元素进入成脉流体中，最终形成绿辉石－绿帘石脉.与石榴石相比，绿辉石和绿帘石具有较高的$Fe^{3+}/\Sigma Fe$(Li *et al*.,2005)，且相对富集重Fe-Mg同位素(图3).因此，绿辉石和绿帘石溶解会导致榴辉岩的$Fe^{3+}/\Sigma Fe$逐渐降低，同时优先运移Fe^{3+}、^{56}Fe和^{26}Mg进入成脉流体中.绿辉石－绿帘石脉从这种成脉流体中结晶分离出来，从而具有高的$Fe^{3+}/\Sigma Fe$、δ^{56}Fe和δ^{26}Mg值(图2).

2.2　流体演化过程中的Fe-Mg同位素分馏

花凉亭多期变质脉记录了俯冲带流体的演化过程.绿辉石－绿帘石、绿帘石－石英和蓝晶石－绿帘石－石英脉相继从同一成脉流体中结晶沉淀出来(Guo *et al*.,2015).在花凉亭变质脉中，绿帘石的Eu/Eu*[$Eu_N/(Sm_N \cdot Gd_N)^{0.5}$]可以反映成脉流体的演化过程，因为绿帘石是轻稀土元素的主要寄主矿物，且绿帘石与流体之间Eu的分配系数大于Sm和Gd(Feineman *et al*.,2007;Martin *et al*.,2011).全岩和绿帘石系统变化的Eu/Eu*(图4)是矿物连续分离结晶的结果.δ^{56}Fe和δ^{26}Mg与Eu/Eu*呈正相关性，说明脉体变化的Fe-Mg同位素组成是流体演化过程中，矿物－流体之间Fe-Mg同位素平衡分馏的结果.因为绿辉石和绿帘石相对富集^{56}Fe和^{26}Mg(图3)，它们的分离结晶会降低残余流体的δ^{56}Fe和δ^{26}Mg，使得后期形成的绿帘石－石英和蓝晶石－绿帘石－石英脉具有较低的δ^{56}Fe和δ^{26}Mg.综合2.1和2.2节，我们认为矿物溶解－再沉淀导致了俯冲带变质流体的Fe-Mg同位素组成的高度变化.

3　弧岩浆岩Fe-Mg同位素异常的成因

相比洋中脊玄武岩(δ^{56}Fe=0.11‰±0.06‰;Weyer and Ionov,2007;Nebel *et al*.,2013;Teng *et al*.,2013)，弧岩浆岩的δ^{56}Fe具有较大的变化范围(－0.15‰～0.71‰;Dauphas *et al*.,2009;Nebel *et al*.,2013,2015;Foden *et al*.,2018).部分熔融会导致玄武质熔体相对地幔源区富集^{56}Fe;岩浆演化过程中，矿物的分离结晶致使残余熔体进一步富集^{56}Fe.因此，弧岩浆岩的重Fe同位素组成可能是部分熔融和岩浆演化共同作用的结果.但是，大部分原始弧岩浆($Mg^{\#} \geqslant 0.60$)富集轻Fe/重Mg同位素(Dauphas *et al*.,2009;Nebel *et al*.,2013,2015;Teng *et al*.,2016;Li *et al*.,2017;Foden *et al*.,2018)，不能用部分熔融和岩浆演化来解释.由于流体优先从沉积物中淋滤出^{54}Fe(Inglis *et al*.,2017;Debret *et al*.,2018)，沉积物脱水形成的流体具有轻Fe同位素组成.沉积物来源的流体可以具有轻或重Mg同位素组成，取决于富集^{24}Mg碳酸盐的溶解，还是富集^{26}Mg白云母和黑云母的溶解(Wang *et al*.,2017).但是，Fe-Mg-Sr-Nd-Pb-Hf同位素联合模拟显示，沉积物来源的流体不

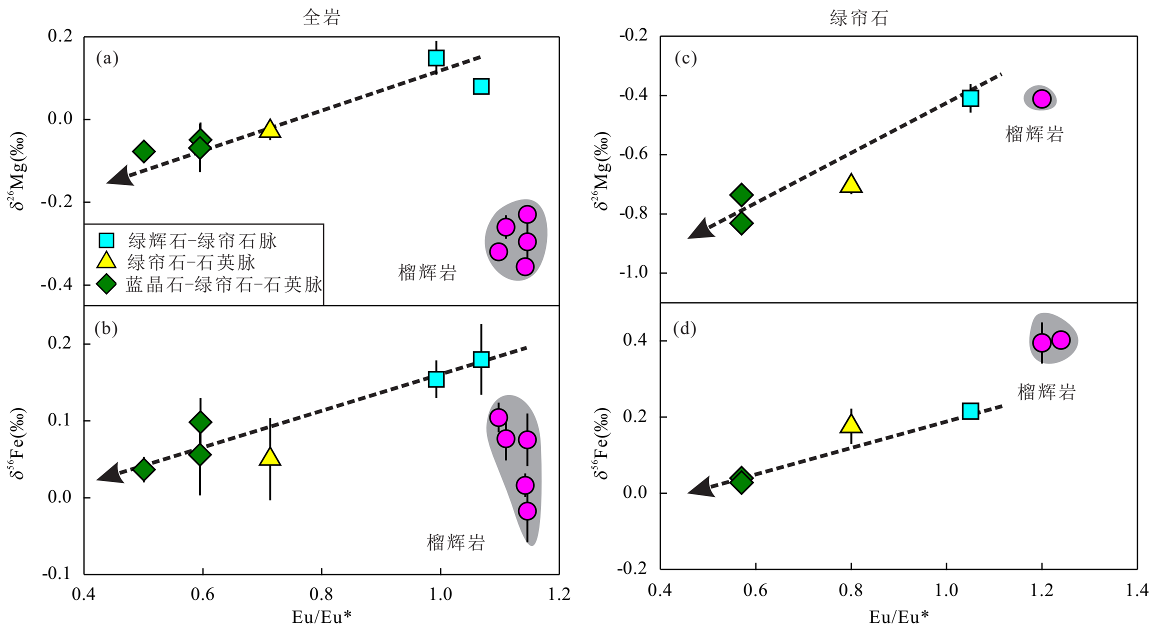

图4　大别山花凉亭三期变质脉全岩(a,b)和绿帘石(c,d)的 Eu/Eu*、δ^{26}Mg 和 δ^{56}Fe 协变图解

Fig.4　Eu/Eu*, δ^{26}Mg, and δ^{56}Fe in whole-rocks (a, b) and epidotes (c, d) of multi-stage veins at Hualiangting in the Dabie orogen

据 Huang *et al*.(2019)

能解释原始弧岩浆岩 Fe-Mg 同位素异常(Nebel *et al*., 2015; Teng *et al*., 2016; Foden *et al*., 2018). 因此,蛇纹岩或蚀变洋壳来源的熔流体成为一种选择(Sossi *et al*., 2016; Teng *et al*., 2016; Li *et al*., 2017).

大别山港河和花凉亭榴辉岩的原岩是大陆玄武岩,它们的主量元素含量与蚀变洋壳相似. 因此,港河和花凉亭榴辉岩脱水可以类比为蚀变洋壳在榴辉岩相变质脱水. 变质脉的 δ^{56}Fe(0.04‰~0.21‰)和 δ^{26}Mg(−0.08‰~0.15‰)高于亏损地幔(δ^{56}Fe=0.03‰±0.03‰, Craddock *et al*., 2013; δ^{26}Mg=−0.25‰±0.07‰, Teng *et al*., 2013),暗示蚀变洋壳来源的流体具有偏重的 Fe-Mg 同位素组成,不能解释弧岩浆岩的轻 Fe 和重 Mg 同位素特征. 蚀变洋壳具有与洋中脊玄武岩一致或偏重的 Mg 同位素组成(图1)也支持这一结论.

蛇纹石和滑石是蛇纹岩中重要的富水和富 Mg 矿物. 滑石非常富集 ^{26}Mg,其脱水会导致蛇纹岩流体具有较高的 δ^{26}Mg 值(0.42‰~0.95‰; Chen *et al*., 2016). 蛇纹石富集 ^{54}Fe(Scott *et al*., 2017),同时蛇纹岩来源的流体一般富集 $Fe^{2+}SO_x$ 或 $Fe^{2+}Cl_2$(Debret *et al*., 2016),而 $Fe^{2+}SO_x$ 或 $Fe^{2+}Cl_2$ 相对富集 ^{54}Fe(Dauphas *et al*., 2017). 因此,蛇纹岩脱水产生的流体富集 ^{54}Fe(Debret *et al*., 2016). 在 Kohistan 弧下地幔中,从富 S-Cl 的蛇纹岩流体中结晶的橄榄石的 δ^{56}Fe 低至−0.36‰(Debret *et al*., 2018). 结合橄榄石与富 S-Cl 流体之间的 Fe 同位素分馏系数(即500 ℃时,$\Delta^{56}Fe_{橄榄石-Fe_2+SO_4(H_2O)_5}$=0.09‰, $\Delta^{56}Fe_{橄榄石-Fe_2+Cl_2(H_2O)_5}$=0.22‰; Dauphas *et al*., 2017),估计得到蛇纹岩流体的 δ^{56}Fe 可以低至−0.58‰~−0.45‰. 同时,多相固体和高压变质脉研究结果显示,蛇纹岩来源的流体具有变化较大但总体偏高的 MgO(1.05‰~39.8%)和 FeO(1.02‰~15.9%)含量(Debret *et al*., 2016). 因此,弧岩浆岩 Fe-Mg 同位素异常是由富集 ^{54}Fe-^{26}Mg 的蛇纹岩流体交代弧下地幔楔导致的. 这一解释与地幔楔橄榄岩具有高 δ^{26}Mg(−0.26‰~−0.06‰)和低 δ^{56}Fe(−0.38‰~0‰)的结果相吻合(Williams *et al*., 2005; Pogge von Strandmann *et al*., 2011; Turner *et al*., 2018).

4　结论

根据贫碳酸盐的蚀变洋壳以及大别山超高压榴辉岩和变质脉的高精度 Fe-Mg 同位素数据,可以得出以下结论.

(1)东太平洋 IODP 1256 钻孔贫碳酸盐的蚀变洋壳具有与新鲜洋中脊玄武岩一致的 Mg 同位素组成,说明低温海水和高温热液蚀变不会导致 Mg 同位素分馏.

(2)相比围岩榴辉岩,早期绿辉石—绿帘石脉富集重 Fe-Mg 同位素;相比绿辉石—绿帘石脉,晚期绿帘

石—石英和蓝晶石—绿帘石—石英脉的δ^{56}Fe和δ^{26}Mg值逐渐降低.这些结果说明，在流体—岩石反应和流体演化过程中，Fe-Mg同位素发生了显著的分馏，且矿物溶解-再沉淀是Fe-Mg同位素分馏的控制因素.

（3）蚀变洋壳来源的流体比亏损地幔具有偏重的Fe-Mg同位素组成，不能解释弧岩浆岩富集轻Fe/重Mg同位素的特征.因此，弧岩浆岩Fe-Mg同位素异常可能是熔体提取和富集^{54}Fe-^{26}Mg的蛇纹岩流体交代地幔楔两个过程共同作用的结果.

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