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不同地质储库中的镁同位素组成及碳酸盐矿物形成过程中的镁同位素分馏控制因素

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摘要: 镁同位素在低温地球化学过程中显著的分馏效应, 是其示踪地球表生环境演化及物质循环的基础。本文在前人研究的基础上, 对地球上不同地质储库中的镁同位素组成及碳酸盐矿物形成过程中的镁同位素分馏控制因素进行了总结: 火成岩的镁同位素组成较均一; 风化产物总体富集重的镁同位素, 且变化较大; 碳酸盐岩中灰岩相对白云岩富集轻的镁同位素, 但总体上富集轻的镁同位素; 岩石类型、风化强度以及植被等因素对河流地表水的镁同位素组成影响较大, 导致地表水的镁同位素组成总体变化较大; 海水的镁同位素组成均一, 平均值约为 $-0.83\text{\textperthousand}$; 低温条件下, 控制碳酸盐矿物无机成因过程中镁同位素分馏的因素有矿物相、沉淀速率和温度, 其中矿物相是主要控制因素; 生物成因碳酸盐矿物镁同位素组成与生物体对含镁碳酸盐矿物的利用形式有关, 除了需考虑与无机碳酸盐沉淀类似的控制因素外, 还需考虑不同物种对轻、重镁同位素的选择性吸收能力; 因生物成因海相碳酸盐矿物几乎都是由最初的无定形相碳酸盐转变而来, 故生物成因海相碳酸盐矿物的镁同位素特征不能代表生成无定形相碳酸盐的流体的镁同位素特征。镁同位素在低温条件下具有良好的分馏效应, 随着分析测试技术的发展及不同地质储库中镁同位素组成数据的积累和完善, 有关表生环境中镁同位素分馏机制的许多问题将逐步得到解决, 镁同位素在揭示地球表生环境演化及物质循环方面将发挥更大的作用。

关键词: 地质储库; 镁同位素组成; 碳酸盐矿物; 无定形相碳酸盐; 控制因素

要点:

- (1) 镁同位素在低温地球化学过程中具有显著的分馏作用。
- (2) 碳酸盐岩总体上富集轻的镁同位素, 灰岩的镁同位素组成比白云岩稍轻。
- (3) 生物成因海相碳酸盐矿物几乎都是由最初的无定形相碳酸盐转变而来。

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自 21 世纪初以来, 随着多接收电感耦合等离子体质谱仪 (MC-ICP-MS)、二次离子质谱仪 (SIMS) 等高精度测试仪器在非传统稳定同位素分

析领域的应用, 非传统同位素体系在地学领域展现出广阔的应用前景^[1-13]。

镁 (Mg) 属非传统稳定同位素, 同时也是主要的

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造岩元素,广泛参与地球各圈层的相互作用^[14]。自21世纪以来,有关镁同位素在自然界分馏机制的研究取得了一系列成果^[14-17],其在低温地球化学过程中显著的同位素分馏效应,使之在揭示表生地质作用过程(风化作用、河流作用、水岩反应)^[2,18-31],白云岩成因^[32-37],壳幔物质循环^[38-39],甚至在探讨低温矿床流体性质及来源^[40-41]等方面都表现出独特的优势。

1 镁的基本化学特征

除大气圈外,镁在地球各圈层中广泛存在^[14]。地表水体(河流、湖泊、海洋)中的镁离子主要来源于岩石风化和生物体的降解,在水体中又可以碳酸盐矿物、水岩反应或生物体吸收的形式从水体中移出。镁在海水中的性质趋于保守,其在海洋中驻留时间达13Ma,是表生地球化学循环中的主要代表性元素^[20]。

镁在自然界有²⁴Mg、²⁵Mg、²⁶Mg三种稳定同位素,三者的相对丰度分别为78.99%、10.00%和11.01%^[42]。值得一提的是,镁的三种稳定同位素间的相对质量差达4%~8%^[21],使之在表生地质作用中能产生明显的同位素分馏。

2 镁同位素表示方法及标准样品

镁同位素组成用 $\delta^{25}\text{Mg}$ 或 $\delta^{26}\text{Mg}$ 表示:

$$\delta^x\text{Mg} = \left[\frac{(^x\text{Mg}/^{24}\text{Mg})_{\text{样品}}}{(^x\text{Mg}/^{24}\text{Mg})_{\text{标准}}} - 1 \right] \times 1000$$

式中:^xMg为²⁵Mg或²⁶Mg。如无特殊说明,本文的镁同位素组成均指 $\delta^{26}\text{Mg}$ 。

早期镁同位素分析中使用的标准样品为美国国家标准技术研究院(NIST)研制的SRM980,该样品的同位素组成用热电离质谱法(TIMS)测定^[16],在早期的分析精度范围内能满足使用要求。但随着高精度多接收电感耦合等离子体质谱仪(MC-ICP-MS)的使用^[43],发现SRM980标样内部镁同位素组成具有不均一性,其作为镁同位素标样的适宜性受到广泛质疑^[44-45]。为此,剑桥大学Galy等^[44]制备了2个镁同位素溶液标样:DSM3和Cambridge1。其中,因标样DSM3的镁同位素组成与碳质球粒陨石(Orgueil和Allende)相同,自2003年以后,DSM3逐渐取代了SRM980成为国际通用的镁同位素标准物质。

早期采用不同批次SRM980标样报道的镁同位素不具对比性,但若其采用的标样为牛津大学配制

的单一批次SRM980-O溶液标样^[44],均可据以下公式换算成相对于DSM3标样的值来进行对比^[46]:

$$\delta^{25}\text{Mg}(\text{样品})_{\text{DSM3}} = \delta^{25}\text{Mg}(\text{样品})_{\text{SRM980-O}} + \delta^{25}\text{Mg}(\text{SRM980-O})_{\text{DSM3}} + 0.001 \times \delta^{25}\text{Mg}(\text{样品})_{\text{SRM980-O}} \times \delta^{25}\text{Mg}(\text{SRM980-O})_{\text{DSM3}}$$

$$\delta^{26}\text{Mg}(\text{样品})_{\text{DSM3}} = \delta^{26}\text{Mg}(\text{样品})_{\text{SRM980-O}} + \delta^{26}\text{Mg}(\text{SRM980-O})_{\text{DSM3}} + 0.001 \times \delta^{26}\text{Mg}(\text{样品})_{\text{SRM980-O}} \times \delta^{26}\text{Mg}(\text{SRM980-O})_{\text{DSM3}}$$

式中: $\delta^{25}\text{Mg}(\text{SRM980-O})_{\text{DSM3}} = -1.744$, $\delta^{26}\text{Mg}(\text{SRM980-O})_{\text{DSM3}} = -3.405$ 。

3 镁同位素测试技术

热电离质谱法(TIMS)、多接收电感耦合等离子体质谱法(MC-ICP-MS)和激光剥蚀多接收电感耦合等离子体质谱法(LA-MC-ICP-MS)是目前被广为应用的3种镁同位素高精度测量方法。

3.1 热电离质谱法

TIMS法在早期镁同位素分析中被普遍采用,但样品前期耗时长,且分析精度仅能达到1‰~2‰,不能有效地辨识地质体中镁同位素组成的变化,因而该方法应用范围有限。

3.2 多接收电感耦合等离子体质谱法

Galy最早将MC-ICP-MS法应用于镁同位素测试^[15]。与TAMS法相比,该方法的分析精度提高了一个数量级,普遍能达到0.1‰甚至更高($\delta^{25}\text{Mg} = 0.06\%$; $\delta^{25}\text{Mg} = 0.12\%$),而且测试速度也比TAMS法大大提高,每个样品测试时间只需约200s^[16]。采用MC-ICP-MS方法时,为避免样品纯化过程中造成的镁同位素分馏^[47],要求镁的化学分离回收率接近100%。实际操作中为最大程度地减小测试过程中仪器引起的分馏效应,通常采用“样品-标样”交叉技术进行校正,即在分析每一个未知样品的前后分别分析一次标样,用标样实测值与理论值之间的差异来校正仪器引起的分馏效应,进而对样品的实测值进行校正。

3.3 激光剥蚀多接收电感耦合等离子体质谱法

与单纯的MC-ICP-MS方法不同,LA-MC-ICP-MS法将激光剥蚀技术与MC-ICP-MS相结合,最早由Young等^[48-49]和Pearson等^[50]应用于天体和地幔中富镁矿物的镁同位素分析。虽然现阶段LA-MC-ICP-MS法的测试精度与单纯的MC-ICP-MS法相比略显不足,但其优势在于能进行矿物颗粒尺度上的原位分析。因而,随着实验方

法的改进和分析精度的提高,LA-MC-ICP-MS法的应用前景势必越来越广泛。

4 自然界镁同位素的组成

众多学者在大量测试数据的基础上,对自然界不同储库中的镁同位素组成特征进行了详细总结(图1),为研究自然界镁同位素的分馏机制奠定了基础。

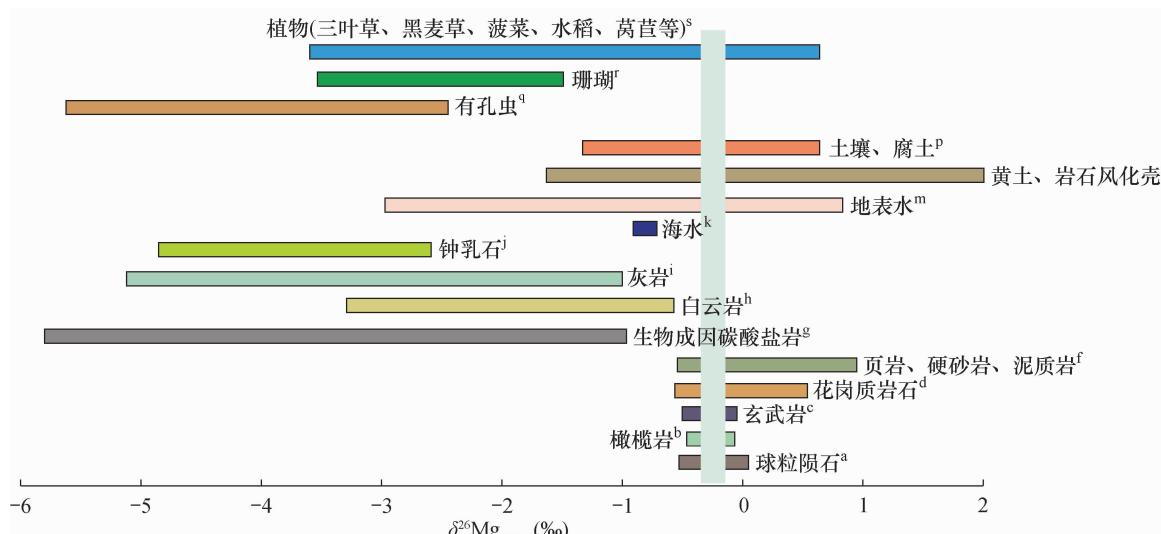
球粒陨石与地球同时诞生于原始太阳星云,两者的总元素丰度相近,但一般认为球粒陨石没有经历过后期的熔融分异作用,其物质组分保留了最初状态。因此,认为球粒陨石的镁同位素组代表了地球形成初期的镁同位素组成。研究发现,地球上火成岩(橄榄岩、玄武岩、花岗岩)具有与地幔相近的镁同位素组成,且与球粒陨石基本一致^[51],这表明在地球形成以后的熔融分异演化中镁同位素的分馏非常小;另外,地幔橄榄岩、玄武岩具有相对均一的镁同位素组成,由此估算地球镁同位素组成的平均值约为-0.25‰^[51](图1中的浅绿色柱子),也有数据是-0.3‰^[6]。黄土、土壤、岩石风化壳等风化产物总体上相对于火成岩富集重的镁同位素,且变化较大,一方面与物质中镁的总量有关,同时还取决于其中“结构镁”与“吸附镁”的相对含量^[2]。整体上碳酸盐岩富集轻的镁同位素,生物成因碳酸盐岩镁同位素组成的变化范围最大,灰岩镁同位素组成比

白云岩稍轻^[34,37,69,73,88,91]。总体上生物体(包括动物、植物)富集轻的镁同位素,且变化范围大,取决于生物体种类以及对镁的利用方式。地表水(不包括海水)镁同位素组成的变化范围也较大,控制因素主要有流域的岩石类型、风化强度、植被类型等;估算的世界主要河流的镁同位素组成平均为-1.09‰±0.05‰^[74]。海水的镁同位素组成均一,δ²⁶Mg值范围为-0.86‰~ -0.77‰,平均值约为-0.83‰^[46-47,79]。

总之,当前自然界不同储库中的镁同位素组成是对不同储库中已有镁同位素测试数据的总结。而总结成果与实际情况的差异,则取决于测试数据的体量^[18,21]、测试方法^[92],甚至不同实验室之间的系统误差^[51]。因此,随着研究的不断深入、实验方法的不断改进以及测试数据的不断积累,对自然界不同储库中镁同位素组成认识将逐渐完善,与自然界实际情况的差异也将日趋减小。

5 碳酸盐矿物形成过程中的镁同位素分馏控制因素

碳酸盐矿物的形成过程是地球各圈层间镁同位素循环的重要环节。碳酸盐矿物的成因可分为无机成因和生物成因两种类型。不同类型的成因过程对镁同位素分馏的控制因素不同,使得镁同位素的分馏程度也不同。



本图修改自参考文献[6,20]。图中各样品涉及的参考文献,a:[51-54];b:[51-52,55];c:[46,51-52,54,56-57];d:[6];f:[58];g:[59-68];h:[33-34,37,47,69-72];i:[37,65,69,73-76];j:[69];k:[46-47,77-79];m:[17,29,69,71,75,77,80-83];n:[24,28-30,77,81,84-87];p:[30,75];q:[68,88];r:[68,88];s:[46,67,89-90]。

图1 不同地质储库中的镁同位素(δ²⁶Mg)组成

Fig. 1 Composition of Mg isotopes (δ^{26} Mg) in major terrestrial materials

5.1 无机成因过程中的镁同位素分馏控制因素

不论是实验模拟还是野外观测,均发现不同的碳酸盐矿物的镁同位素组成不同,这表明不同的碳酸盐矿物在形成过程中,矿物相控制了镁同位素分馏^[59]。自然界常见的碳酸盐矿物依次有方解石(CaCO_3)、白云石[$\text{MgCa}(\text{CO}_3)_2$]、文石(CaCO_3)、菱镁矿(MgCO_3)和水菱镁矿[$\text{Mg}_5(\text{CO}_3)_4(\text{OH})_2 \cdot 4\text{H}_2\text{O}$]。能够在常温常压下自然沉淀形成的有方解石、文石和水菱镁矿。通过实验模拟观测到,当温度为25℃,且沉淀过程达到平衡时,文石、水菱镁矿相对于溶液的分馏程度 $\Delta^{26}\text{Mg}_{\text{文石}-\text{溶液}}$ 、 $\Delta^{26}\text{Mg}_{\text{水菱镁矿}-\text{溶液}}$ 大致相等,均约为-1‰,而 $\Delta^{26}\text{Mg}_{\text{方解石}-\text{溶液}}$ 小于-2‰,最小可达到-3.5‰^[93-94],与野外直接观测基本一致^[95-96]。白云石与菱镁矿在常温常压下不能形成,通过实验室模拟及反演推算,得到25℃时 $\Delta^{26}\text{Mg}_{\text{白云石}-\text{溶液}}$ 约为-1.75‰^[97], $\Delta^{26}\text{Mg}_{\text{菱镁矿}-\text{溶液}}$ 约为-2‰^[98]。由以上可知,碳酸盐矿物形成过程中总体上倾向于富集轻的镁同位素,矿物在形成过程中分馏程度最大的是方解石,其次是菱镁矿、白云石,最小的是文石与水菱镁矿。

据相关模拟实验(实验过程中化学参数基本保持不变)和实际观测,碳酸盐矿物的沉淀速率会对镁同位素的分馏产生影响^[73,93-94]。Mavromatis等^[94]的研究表明,沉淀速率与 $\Delta^{26}\text{Mg}_{\text{方解石}-\text{溶液}}$ 呈正相关:若沉淀速率加快,方解石将相对富集重的镁同位素,若沉淀速率足够小,方解石将相对富集轻的镁同位素,但总体上仍将富集轻的镁同位素。Immenhauser等^[73]、Galy等^[69]通过对洞穴钟乳石的研究,也观测到了与室内模拟实验相似的情况。而Saulnier等^[93]的实验模拟则表明,方解石的沉淀速率是控制镁同位素分馏的主要因素,温度、 Ca/Mg 及pH值的影响有限。上述现象与理论上推导的动力分馏过程中镁同位素分馏机制相反。理论上来说,轻的镁同位素迁移速度快,沉淀形成的碳酸盐矿物更富集轻的镁同位素,即沉淀速率应与 $\Delta^{26}\text{Mg}_{\text{方解石}-\text{溶液}}$ 呈负相关。对此现象,董爱国等^[20]认为,可能与水中的镁元素具有较高的吉布斯自由能,其更倾向于与水结合形成水合镁离子有关。

据Li等^[99]的模拟实验,在温度4~45℃之间,方解石相对于溶液的分馏程度 $\Delta^{26}\text{Mg}_{\text{方解石}-\text{溶液}}$ 与温度呈正相关;Wang等^[95]在常压下、22~55℃、与海水性质类似的水体中对文石沉淀的模拟实验也表明 $\Delta^{26}\text{Mg}_{\text{文石}-\text{溶液}}$ 与温度呈正相关;但据实验模拟推算的 $\Delta^{26}\text{Mg}_{\text{方解石}-\text{溶液}}$ 与 $\Delta^{26}\text{Mg}_{\text{文石}-\text{溶液}}$ 平衡分馏值与理论计

算结果均存在较大差异^[20],具体原因尚不明确。在白云石形成的模拟实验中^[97],温度为130℃、160℃和220℃时, $\Delta^{26}\text{Mg}_{\text{白云石}-\text{溶液}}$ 分别为-0.93‰、-0.84‰和-0.65‰,两者也表现为正相关关系,且实验估算的平衡分馏值介于理论计算范围之内^[99-100]。在Pearce等^[98]的菱镁矿合成实验中,在120~200℃,CO₂压力为15~30个大气压条件下,测得150℃、200℃时的 $\Delta^{26}\text{Mg}_{\text{菱镁矿}-\text{溶液}}$ 分别为-1.19‰和-0.88‰,也显示了温度与 $\Delta^{26}\text{Mg}_{\text{菱镁矿}-\text{溶液}}$ 之间的正相关关系,且实验模拟平衡分馏的值与理论计算值接近。由此可知,低温条件下碳酸盐矿物无机沉淀过程中的镁同位素分馏与温度成正比。

综上,在低温条件下,控制碳酸盐矿物无机成因过程中镁同位素分馏的因素有矿物相、沉淀速率和温度,其中矿物相应是主要控制因素。

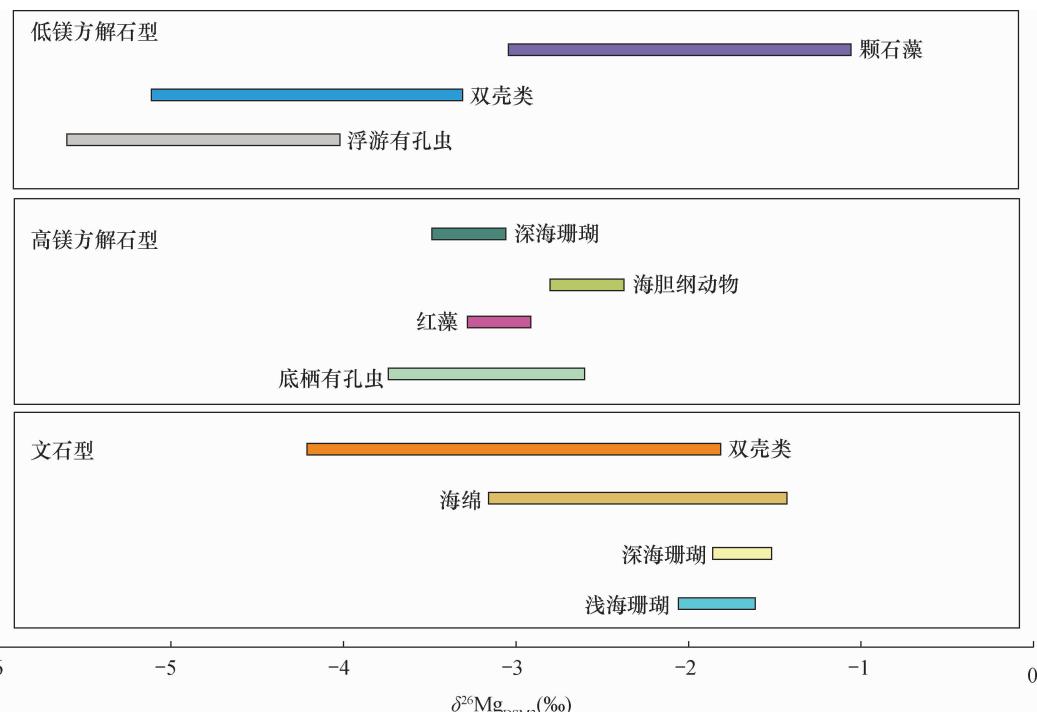
5.2 生物成因过程中的镁同位素分馏控制因素

与生物活动有关的碳酸盐矿物整体上富集轻的镁同位素(图2)。除矿物相以外,生物体对含镁碳酸盐矿物的利用形式也是控制生物成因碳酸盐矿物镁同位素组成的主要因素^[88]。

生物体对含镁碳酸盐矿物的利用存在两种形式:一种是直接组成生物体的一部分,如有孔虫;另一种是碳酸盐矿物被粘结于生物体周围,支撑生物体生长,如珊瑚。利用形式不同,生物体对镁同位素的利用机制也不同^[63]。在第一种形式中,生物体优先吸收轻的镁同位素(如部分有孔虫),产生的镁同位素分馏较大,且因物种不同而有差异,这可能与不同物种对轻、重镁同位素的选择性吸收能力有关;第二种形式则与无机碳酸盐沉淀过程类似,受矿物相、沉淀速率和温度等诸多因素控制。

生物成因碳酸盐矿物相可分为高镁方解石型、低镁方解石型和文石型三种。高镁方解石型碳酸盐矿物的镁同位素组成范围较窄;大多数低镁方解石型生物碳酸盐矿物都具有较宽范围的镁同位素组成,而部分文石型生物碳酸盐矿物的镁同位素组成与低镁方解石型生物碳酸盐矿物相似。但总体上,类方解石型生物碳酸盐矿物的镁同位素组成大多数要明显轻于文石型生物碳酸盐矿物。

综上,生物成因碳酸盐矿物镁同位素组成的影响因素与生物体对含镁碳酸盐矿物的利用形式有关。除了需考虑与无机碳酸盐沉淀类似的控制因素外,还需考虑不同物种对轻、重镁同位素的选择性吸收能力。



本图修改自文献[88]。

图2 生物成因碳酸盐矿物的镁同位素($\delta^{26}\text{Mg}$)组成

Fig. 2 Mg isotope composition ($\delta^{26}\text{Mg}$) of biogenic carbonate minerals

5.3 高镁无定形碳酸钙向高镁方解石转变过程中 的镁同位素分馏

Mavromatis 等^[101]在给定实验条件下(温度 25°C, pH 8.3), 分别将 50mL 浓度均为 0.6mol/L, 但 Mg/Ca 比值分别为 1:4、1:5、1:6、1:8 的 (Ca,Mg)Cl₂ 溶液以 2mL/min 的速率滴入浓度为 1mol/L 的碳酸氢钠溶液中。当滴定溶液的 Mg/Ca 比值为 1:8 时, 高镁方解石(Mg-Calcite)将直接从反应溶液中晶出, 且晶出的高镁方解石相对于反应溶液的分馏程度 $\Delta^{26}\text{Mg}_{\text{Calcite}-\text{fluid}}$ 随时间波动不大。

相比之下, 当滴定溶液的 Mg/Ca 比值分别为 1:4、1:5、1:6 时, 反应溶液中将最先晶出高镁无定形碳酸钙(Mg-ACC), 然后再转变为高镁方解石。在滴定初期, 高镁无定形碳酸钙相对于反应溶液的分馏程度 $\Delta^{26}\text{Mg}_{\text{ACC}-\text{fluid}}$ 维持在 $-1.0\text{‰} \pm 0.1\text{‰}$, 随着实验过程中高镁无定形碳酸钙向高镁方解石转变, 镁同位素发生明显迁移, 新生成的高镁方解石相对于反应溶液的分馏程度 $\Delta^{26}\text{Mg}_{\text{Calcite}-\text{fluid}}$ 随时间发生显著变化, 下降至最低 -3.6‰ 。

海相碳酸盐矿物可分为非生物成因和生物成因, 其中生物成因的海相碳酸盐矿物几乎都是由无定形碳酸盐相转变而来。实验研究提示, 对于非生

物成因的海相碳酸盐胶结物, 其 $\delta^{26}\text{Mg}$ 特征基本上可代表其形成时的流体同位素组成; 而现代稳定生物成因海相碳酸盐矿物的 $\delta^{26}\text{Mg}$ 特征已随时间发生显著变化, 不能代表其前期无定形碳酸盐相形成时的流体同位素组成。因此, 能否利用生物成因碳酸盐矿物的 $\delta^{26}\text{Mg}$ 特征恢复古海洋环境, 仍需商榷。

6 存在问题和发展方向

关于不同地质储库中的镁同位素组成, 尽管目前已取得一系列成果, 但仍有一些数据需进一步补充、完善。如: 目前对地球下地壳的镁同位素组成仍缺乏直接研究; 相当多的学者对世界河流的镁同位素组成的平均值存在质疑, 认为需进一步限定。完善的镁同位素储库数据是探讨不同储库间镁同位素地球化学循环的基础, 因此, 在现有研究成果的基础上, 补充、完善地球各储库中镁同位素组成的数据显得尤为重要。

由于白云石在表生作用下不能直接通过沉淀形成, 目前有关白云石沉淀过程中的镁同位素分馏研究都是在热液条件下通过实验模拟进行, 完全不同于表生环境中的白云岩形成机制。因此, 表生条件下白云岩形成过程中的镁同位素分馏还是研究空

白。开展表生条件下白云岩形成过程中的镁同位素分馏研究,不仅能进一步揭示表生作用下的镁同位素分馏机制,还有助于揭开长久以来困扰地学界的“白云岩”问题。

镁是地球上广泛存在的造岩元素,由于镁同位素在低温条件下良好的分馏效应,使其具备示踪地球表生环境演化及物质循环的潜力。可以预料,随着分析测试技术的发展及不同地质储库中镁同位素组成数据的积累和完善,有关表生环境中镁同位素分馏机制的许多问题将逐步得到解决,镁同位素在揭示地球表生环境演化及物质循环方面将发挥更大的作用。

7 参考文献

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Magnesium Isotope Composition of Different Geological Reservoirs and Controlling Factors of Magnesium Isotope Fractionation in the Formation of Carbonate Minerals—A Summary of Previous Results

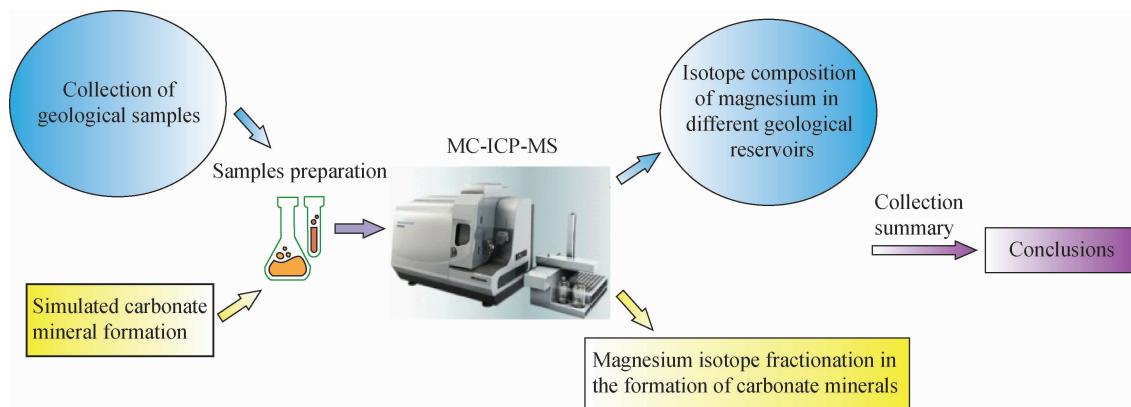
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HIGHLIGHTS

- (1) Magnesium isotopes showed significant fractionation during low – temperature geochemical processes.
- (2) Carbonate rocks showed enrichment in light isotopes of magnesium in general; magnesium isotopes of limestone were lighter than those of dolomite.
- (3) Almost all biogenic marine carbonate minerals were transformed from the original amorphous phase carbonate precursor.



ABSTRACT

BACKGROUND: Magnesium isotope fractionation effect during low – temperature geochemical processes is the foundation of tracing supergene evolution and material cycle of the earth.

OBJECTIVES: To summarize the magnesium isotope composition of different geological reservoirs and investigate the controlling factors of the magnesium isotope fractionation during the formation of carbonate minerals.

METHODS: Systematic collection and summary of previous research results.

RESULTS: Magnesium isotope compositions of igneous rocks were relatively homogeneous. The weathering products were relatively enriched in heavy isotopes of magnesium with significant variation. Carbonate rocks showed enrichment in light isotopes of magnesium in general. The large variation of magnesium isotope composition of river water was affected by lithology, weathering degree and vegetation. Magnesium isotope composition of seawater was homogeneous with an average of $-0.83\text{\textperthousand}$. At low temperature, the factors controlling the fractionation of magnesium isotopes in the inorganic process of carbonate minerals were mineral phase, precipitation rate and temperature, of which mineral phase was the main controlling factor. The factors influencing the magnesium isotope composition of biogenic carbonate minerals were forms of utilization of magnesium carbonate minerals by organisms. In addition to considering the mechanisms that were similar to inorganic carbonate precipitation, the selective absorption of light and heavy magnesium isotopes by different species should be considered. Almost all biogenic marine carbonate minerals were transformed from the original amorphous phase carbonate precursor, and their original magnesium isotope composition was masked by the later magnesium isotope composition during transformation. Therefore, the magnesium isotope composition of biogenic marine carbonate minerals cannot represent the fluid isotope composition from which the original amorphous carbonate precursor formed.

CONCLUSIONS: Magnesium isotopes have a good fractionation effect at low temperature. With the development of analytical technology and the accumulation and improvement of magnesium isotope composition data in different geological reservoirs, many problems related to the mechanism of magnesium isotope fractionation in the supergene environment will be solved gradually. Magnesium isotopes will play a greater role in revealing the evolution of the supergene environment and the material cycle of the earth.

KEY WORDS: geological reservoirs; magnesium isotope composition; carbonate minerals; amorphous carbonate; controlling factors