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Why magnesium isotope fractionation is absent from basaltic melts under thermal gradients in natural settings

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Abstract

Laboratory experiments have shown that thermal gradients in silicate melts can lead to isotopic fractionation; this is known as the Richter effect. However, it is perplexing that the Richter effect has not been documented in natural samples as thermal gradients commonly exist within natural igneous systems. To resolve this discrepancy, theoretical analysis and calculations were undertaken. We found that the Richter effect, commonly seen in experiments with wholly molten silicates, cannot be applied to natural systems because natural igneous samples are more likely to be formed out of partially molten magma and the presence of minerals adds complexity to the behaviour of the isotope. In this study, we consider two related diffusion-rate kinetic isotope effects that originate from chemical diffusion, which are absent from experiments with wholly molten samples. We performed detailed calculations for magnesium isotopes, and the results indicated that the Richter effect for magnesium isotopes is buffered by kinetic isotope effects and the total value of magnesium isotope fractionation can be zero or even undetectable. Our study provides a new understanding of isotopic behaviour during the processes of cooling and solidification in natural magmatic systems.

1. Introduction

Significant chemical and isotopic compositional changes can occur in thermal gradients while conducting laboratory experiments with silicate melts, known as the Soret effect (Walker & Delong, 1982; Leshner & Walker, 1986). Specifically, when the concerned species only focuses on isotopes associated with the Soret effect, it is called a Richter effect (Dauphas *et al.* 2010). Both the Soret effect and the Richter effect have been widely investigated in high-temperature experiments using a thermal gradient where both ends of the experimental silicate melt were set at a higher temperature than the liquidus temperature of silicate samples (Leshner & Walker, 1986; Richter *et al.* 2003, 2009). Although thermal gradients must exist in the cooling and solidification processes of magma bodies, the role of thermal gradients in magmatic differentiation has been regarded as unimportant since heat dissipation occurs faster than mass diffusion (Bowen, 1915). However, recent geochronological observations and numerical results have revealed that the cooling time for plutonic rocks may be 10–100 Ma; Soret and Richter effects may therefore be expected (Coleman *et al.* 2004; Huang *et al.* 2009, 2010; Xu *et al.* 2014). For example, Bouquain *et al.* (2009) explained the reversal of the crystallizing sequence of pigeonite and augite along the investigated rock profile by referring to the Soret diffusion in Alexo komatiites in Canada. However, the large isotopic variations (Richter effect) that were expected for magnesium and iron were not observed (Dauphas *et al.* 2010). Moreover, no isotopic fractionation that has been caused by the Richter effect has been found in the natural samples that have been studied to date (Teng, 2017). The reason for this discrepancy is not clear and needs further investigation.

Thermal migration is another process that is different from the Soret and Richter effects, and it occurs when the temperatures at both ends of a thermal gradient are lower than the liquidus temperature (Leshner & Walker, 1988; Huang *et al.* 2009). The mechanism for thermal migration is that crystal solubilities vary with changes in temperature, which establishes a chemical gradient in an interstitial melt and also induces mass transportation. As a result, minerals keep crystallizing at the cold end and dissolving at the hot end because of the local equilibrium between minerals and interstitial melts, the composition of which changes. Overall, two potentials drive the species to diffuse within interstitial melts in partially molten silicates: one is the Soret effect, which tends to concentrate in cooler or warmer regions depending on its component properties; and the other is thermal migration, which is derived from the chemical gradient (Leshner & Walker, 1988). Both the Soret effect and thermal migration are achieved through mass diffusion during which kinetic isotope fractionation occurs because light isotopes diffuse at a faster rate than heavy isotopes. The kinetic isotope effects mentioned here could theoretically

weaken or eliminate the Richter effect. However, the degree to which the Richter effect is weakened has not been evaluated before.

Here, we first emphasized the experimental differences between wholly molten and partially molten basaltic silicates, and theoretically elaborated on the mechanisms of isotopic fractionation in partially molten silicates. We then conducted quantitative Mg kinetic isotope fractionations to assess the influence of isotopic fractionation on the Richter effect. We aim to provide a new explanation for the absence of the Richter effect in Alexo komatiites in Canada, and to further our understanding of isotopic behaviour during the cooling and solidification processes of natural magmatic systems.

2. Theoretical analysis

2.a. Richter effect (δ_1)

Thermal isotope fractionation was initially found in oxygen isotopes during thermal gradient experiments (Kyser *et al.* 1998). Further studies have been undertaken using wholly molten silicate experiments, measured using the application of a multicollector inductively coupled plasma mass spectrometer (MC-ICP-MS) to non-traditional stable isotopic measurements (Richter *et al.* 2003, 2008, 2009, 2014; Lacks *et al.* 2012). Isotope fractionation under thermal gradient in silicate liquids is termed the ‘Richter effect’ to highlight Frank M. Richter’s contribution (Dauphas *et al.* 2010). The Richter effect is denoted δ_1 in this paper.

Within the Richter effect, light isotopes tend to enrich the hot end of a thermal gradient, while heavy isotopes tend to enrich the cold end (Fig. 1). The typical Richter effect is a steady state that is between equilibrium and kinetic isotope fractionation (Yun, 2015). Although the elemental and isotopic compositions of the thermal ends differ, they do not vary with time and no net chemical flux occurs across the section. Although the governing mechanism of the Richter effect has not been fully elucidated (Dominguez

et al. 2011; Lacks *et al.* 2012; Li & Liu, 2015), the value of the Richter effect can be calculated from experiments and the following recommended equation (Lacks *et al.* 2012):

$$\delta_1 = -\Omega \times (m_h - m_l)(T_h - T_c), \quad (1)$$

where Ω is an experimental parameter, defined as the fractionation in per mil per 100°C per atomic mass unit (amu), m_h and m_l are the atomic mass of the heavy and light isotopes, respectively, and T_h and T_c are the temperatures of the hot end and cold end, respectively.

2.b. Kinetic isotope fractionation caused by thermal migration (δ_2)

Thermal gradients occur in partially molten silicates. Figure 2 demonstrates thermal migration in a binary system comprising phases A and B (Leshner & Walker, 1988). The system is open to heat but is isolated from mass transfers. The bulk composition X is placed under a thermal gradient, as shown in Figure 2a. The system quickly establishes a local chemical and isotopic equilibrium along the temperature gradient, and the cold end of the gradient becomes enriched in heavy isotopes (the Richter effect in partially molten silicates, δ_1). Solid-phase A is on liquidus throughout, and the amount increases with decreasing temperature as per the level rule (William & Javad, 2005). As a result, the interstitial melt becomes poorer in component A as the temperature decreases. Although this initial configuration corresponds to an equilibrium state within a closed system, it is not actually stable since mass communication is permitted through the melts. Figure 2b shows that component A begins to diffuse from a high-temperature to a low-temperature region, and the diffusing process enriches the number of light isotopes of A in the cold end as light isotopes diffuse at a faster rate than heavy isotopes. This kinetic isotope fractionation (denoted δ_2) occurs in the opposite direction to that of the Richter effect. The mass transfer throughout the melts disrupts the previous local equilibrium between solid and liquid states. Furthermore, an increasing amount of component A in the cold melts is consumed by the crystallization of phase A, and a reduced amount of component A in the hot melts are replenished by the dissolution of phase A. The isotopic composition of the newly crystallized phase A consists of a combination of δ_1 and δ_2 . Figure 2c is the final state, when the solid and liquid fronts meet. The Richter effect remains only within the molten region.

Zhu *et al.* (2015) provided the following equation to calculate kinetic isotopic fractionation associated with the diffusing process:

$$\delta_2 = 1000 \times \left(1 - \left(\frac{m_2}{m_1} \right)^\beta \right) \left(\frac{C_0}{C_\infty} - 1 \right), \quad (2)$$

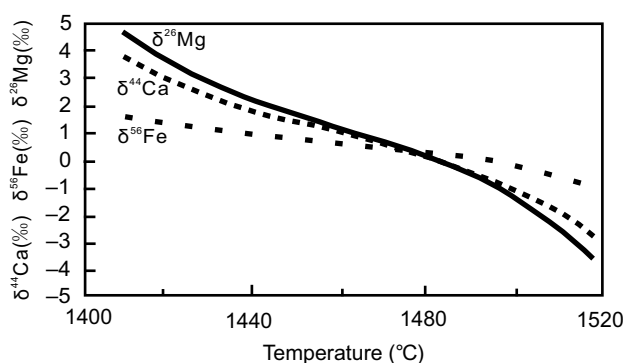


Fig. 1. Richter effect for Mg, Ca and Fe isotopes (Richter *et al.* 2009).

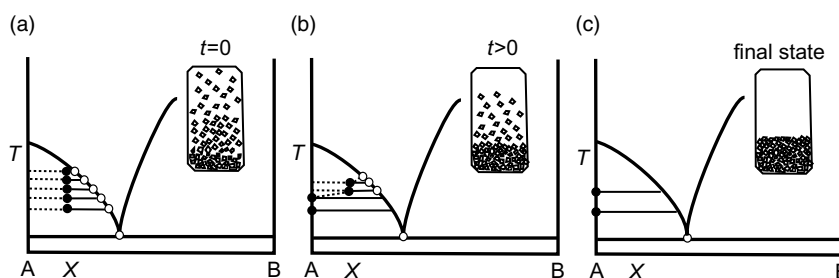


Fig. 2. Schematic illustration of thermal migration in binary system A-B (Leshner & Walker, 1988).

where m_1 and m_2 are masses of heavy and light isotopes, respectively; β is an empirical parameter obtained from experiments (Richter *et al.* 2003) and theoretical computation (Watson & Muller, 2009); and C_0 and C_∞ are the elemental concentrations of an interface of crystals and melts and of far-field melts, respectively. C_0 and C_∞ provide a chemical potential for diffusion occurrence. In this paper, these C_0 and C_∞ may be viewed as elemental concentrations of the hot and cold ends, respectively.

2.c. Kinetic isotope fractionation caused by Soret diffusion (δ_3)

In addition to δ_2 in partially molten silicates, another kinetic isotope fractionation influences the final result. This effect is caused by a non-equilibrium Soret diffusion (denoted δ_3). The elemental and isotopic equilibrium states can be reached if Soret diffusion takes place within wholly molten silicates; this is conditional upon whether they are given enough time and whether their variations are associated with temperature differences. However, in partially molten silicates, an elemental Soret equilibrium cannot be achieved because the distribution in the melts is not only influenced by the Soret effect, but also constrained by the local equilibrium between liquids and solids; on other words, more components are consumed upon crystallization, as shown in Figure 2b. Consequently, the Soret equilibrium has the potential to keep acting and driving the diffusion of species, during which kinetic isotope fractionation occurs. Using Mg as an example in partially molten basaltic silicates, heavy Mg isotopes initially concentrate in melts at the cold end (δ_1). Thermal migration then drives Mg to migrate from the hot end to the cold end, because the concentration of Mg is higher at the hot end. This process enriches light Mg isotopes in the cold end (δ_2); meanwhile, Mg spontaneously diffuses to the cold end, even if there is no chemical migration; this is because the Soret effect requires the enrichment of Mg at the cold end, and this also enriches the light Mg isotopes at the cold end (δ_3). Therefore, δ_2 and δ_3 play roles opposite to that of δ_1 .

The chemical distribution for Soret equilibrium is described (Walker & Delong, 1982):

$$\sigma = \frac{X_{\text{cold}} - X_{\text{hot}}}{\bar{X}(1 - \bar{X}) \times \Delta T} \quad (3)$$

where σ is the Soret coefficient; X_{cold} , X_{hot} and \bar{X} are the concentrations at the cold end, hot end and the bulk value, respectively; and ΔT is the temperature difference. X_{cold} and X_{hot} can be calculated if \bar{X} , ΔT and σ are known. X_{cold} and X_{hot} provide the diffusion potential, and δ_3 can be obtained from:

$$\delta_3 = 1000 \times \left(1 - \left(\frac{m_2}{m_1} \right)^\beta \right) \left(\frac{X_{\text{cold}}}{X_{\text{hot}}} - 1 \right) \quad (4)$$

3. Calculations and results

δ_1 , δ_2 and δ_3 for Mg are obtained from Equations (1), (2) and (4). The results may vary if different parameters are used for the calculations (temperature gradient, β_{Mg} , σ_{Mg} , etc.). In this paper, the temperature gradient is quoted from a portion of the crystallizing komatiitic lava in Alexo, Canada, which is suggested to be 1200–1270°C across a 22 cm profile (Bouquain *et al.* 2009). β_{Mg} , the ratio of the effective diffusing coefficient of ^{24}Mg to that of ^{26}Mg , is 0.05, which is obtained from Richter *et al.* (2008). The Soret coefficient σ_{Mg} acts as a proxy for the Mg Soret effect, and can span four orders of magnitude ranging from 10^{-2} (Walker & Delong, 1982) to 10^{-5} (Latypov, 2003). The reason for this wide range is partially due to the differing water content between the experimental samples. In addition, the experiment itself has a high level of uncertainty. We arrive at the calculated results by using σ_{Mg} over several orders of magnitude. The MgO content under liquidus temperature is also needed for these calculations. Multiple equations have been provided in previously published literature (e.g. Niu *et al.* 2002; Chen & Zhang, 2008) to calculate the relationship between MgO and the lava liquidus temperature; the results are similar, and we used the equation provided by Niu *et al.* (2002):

$$T_{\text{liquidus}} = 1026 e^{0.01894\text{MgO}}, \quad (5)$$

where T is in °C and MgO is in wt%.

Results for δ_1 , δ_2 and δ_3 with different σ_{Mg} are listed in Table 1.

Table 1. Calculations of Mg isotope fractionation with a thermal gradient ($T_{\text{cold}}-T_{\text{hot}}$) of 1200–1270°C and β_{Mg} of 0.05 for different values of σ_{Mg} .

| σ_{Mg} | 10^{-2} | 10^{-3} | 10^{-4} | 10^{-5} | 6.75×10^{-3} |
|--------------------------------------|-----------|-----------|-----------|-----------|-----------------------|
| C_{cold} (%) | 8.27 | 8.27 | 8.27 | 8.27 | 8.27 |
| C_{hot} (%) | 11.26 | 11.26 | 11.26 | 11.26 | 11.26 |
| Bulk (%) | 9.77 | 9.77 | 9.77 | 9.77 | 9.77 |
| δ_1 (‰) | 5.04 | 5.04 | 5.04 | 5.04 | 5.04 |
| δ_2 (‰) | -1.45 | -1.45 | -1.45 | -1.45 | -1.45 |
| $C_{\text{Soret,cold}}$ (%) | 12.85 | 10.08 | 9.80 | 9.77 | 11.85 |
| $C_{\text{Soret,hot}}$ (%) | 6.68 | 9.46 | 9.74 | 9.76 | 7.69 |
| δ_3 (‰) | -4.95 | -1.63 | -1.36 | -1.34 | -3.59 |
| $\delta_1 + \delta_2 + \delta_3$ (‰) | -1.36 | 1.96 | 2.23 | 2.25 | 0 |

Notes: T_{cold} and T_{hot} are temperatures of the cold and hot ends, respectively; β is an empirical parameter obtained from experiments (Richter *et al.* 2008); σ_{Mg} is the Soret coefficient of Mg; C_{cold} and C_{hot} are Mg concentrations at the cold and hot ends, respectively (Equation (5)); Bulk is the theoretical mean Mg concentration, estimated as $(C_{\text{cold}} + C_{\text{hot}})/2$; δ_1 is the Richter effect; δ_2 and δ_3 are the values of kinetic isotope fractionation caused by thermal migration and Soret diffusion, respectively; $C_{\text{Soret,cold}}$ and $C_{\text{Soret,hot}}$ are the theoretical Soret steady values of Mg; and $\delta_1 + \delta_2 + \delta_3$ is the final value of Mg isotope fractionation.

4. Discussion

4.a. Influence of Mg kinetic isotope effects on the Richter effect

As the Richter effect is approximately one order of magnitude larger than the equilibrium fractionation in magmatic rocks for Mg isotopes, and since a temperature difference of 70°C can fractionate Mg isotopes by 5.04‰ (Table 1), the conventional view states that the Richter effect should be observed if it manifests in natural samples. However, the calculated results indicate that the Mg kinetic isotope effects are the same order of magnitude as δ_1 but of the opposite sign; they therefore have the potential to theoretically counterbalance the Richter effect. The value of δ_2 was determined by the saturation gradient of MgO between the hot and cold ends, which was fixed according to the given local temperature. For 1200–1270°C, δ_2 is -1.45‰ and δ_3 is set by the MgO chemical gradient under the assumed ideal Soret steady state. If the Soret effect could demonstrate the fractionation of Mg isotopes of a large value, the diffusing potential would correspondingly increase, leading to a large δ_3 value. The results show that δ_3 can vary from -4.95‰ to -1.34‰ with the variation of σ_{Mg} . The total $\delta^{26}\text{Mg}$ (e.g. $\delta_1 + \delta_2 + \delta_3$) varies from -1.36‰ to $+2.25\text{‰}$. If σ_{Mg} is arbitrarily adopted as 6.75×10^{-3} , then the total $\delta^{26}\text{Mg}$ is calculated as 0 and the Mg isotope fractionation that is caused by the thermal gradient would not be observed in natural samples. We acknowledge that the calculations are currently not precise enough due to uncertainty regarding σ_{Mg} , which leads to a wide range of values for δ_3 . However, the calculations provide a theoretical possibility that the Richter effect could be counterbalanced by the kinetic isotope effects and could further be

understood as the reason for the absence of the Richter effect in natural samples.

4.b. Time of thermal gradient existence

It is important to consider whether the thermal gradient can be maintained long enough for mass diffusion to occur. We estimated the existence interval of a thermal gradient and the time required to achieve a Richter 'steady state'. A thermal gradient may be maintained for a longer period in erupted lava than in intrusions. This is because the quenched surface of the lava flow is held to be near the value of 0, while the thermal gradient may disappear after boundary rocks are heated by intrusions. It has been suggested that the cooling rate is near 0.1°C/hour for komatiite flows in the case of liquids under a 4 m thick solid crust (Turner *et al.* 1986). In 1200–1270°C across 22 cm, the thermal gradient might exist for at least 29 days.

The time taken for the silicate melts to reach the Richter steady state can be estimated from the equation $\theta = L^2/D\pi^2$ (Richter *et al.* 2008), where θ is the time needed, L is the diffusing distance and D an effective binary diffusing coefficient, reported as $5.10^{-5} \text{ cm}^2 \text{ s}^{-1}$ from Bouquain *et al.* (2009). Here, $L = 22 \text{ cm}$, and θ is approximately 11 days. This result indicates that the thermal gradient interval is sufficient for the Richter effect to occur.

4.c. Mg isotopes in Alexo komatiites

In the Alexo komatiites of Canada, a temperature gradient has been reported to explain the reverse order of pigeonite and augite crystallization that occurred during the processes of cooling and solidifying (Bouquain *et al.* 2009). Furthermore, no obvious Mg isotope

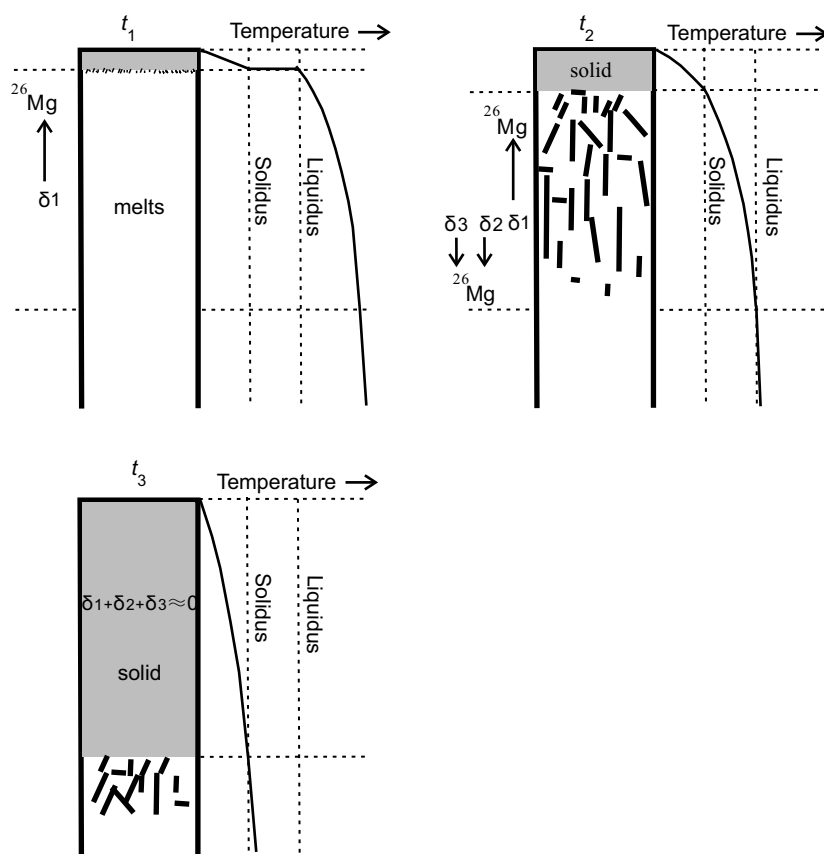


Fig. 3. Mg isotope fractionation after magma eruption. During t_1 , a quenched lid forms at the surface to be near 0°C and the bottom becomes the solidus of silicate lava. A thin layer exists under the lid, within which temperature jumps from solidus to liquidus. Under this thin layer, the temperature distribution is adiabatic and super-liquidus; this zone can therefore be assumed to comprise wholly molten silicates. The Richter effect drives heavy Mg isotopes to the cold surface and light Mg isotopes to the hot interior. During t_2 , with lava cooling progressively, a partial crystallizing zone forms, where the presence of crystals decrease from the surface to the interior, leading to Mg chemical gradients in interstitial melts. Mg migrates to the cold surface, responding to thermal migration and Soret diffusion, and this diffusing process enriches light Mg isotopes in the cold surface. During t_3 , with the lava becoming cooler, the crystallized zone moves downwards. The solidified rocks record the sum of the Richter effect and kinetic effects.

fractionation was observed along the profile (Dauphas *et al.* 2010). The Mg isotope fractionation which considered only the occurrence of the Richter effect during magma cooling in previous calculations was too large. However, we have interpreted the observation as a combination of the Richter effect and kinetic isotope fractionations. We divided the magma-evolving process after an eruption into three stages (Figure 3). Over t_1 , conducted crust forms quickly because the surface temperature remains near 0°C and a nearly wholly molten interior is under the crust (where the adiabatic thermal gradient exists). The Soret effect for elements and isotopes are active, enriching the upper layer (cooler end) with high Mg concentration and heavier isotopes. During t_2 , a partial crystallizing zone forms with a degree of solidification that ranges from 100% at the top to 0% at the bottom. Mg accumulated in melts during stage t_1 is consumed by the crystallization of mafic minerals with a high Mg content. An Mg chemical gradient forms within the interstitial melts, with the Mg content in each section maintained at the local temperature. Thermal gradients and Soret diffusion continue driving Mg to migrate upwards and provides a buffer against the heavy Mg that accumulated during t_1 . The crystallized Mg minerals reflect the mixed isotopic contributions of the Richter effect and kinetic effects. During t_3 , crystallizing fronts move downwards, and no Mg isotope variation can be found on the rock profile.

5. Conclusions

The conclusions of this study are that the Richter effect obtained from wholly molten silicate experiments cannot be directly applied to Mg isotope fractionation under a thermal gradient in natural systems. Furthermore, kinetic isotope fractionation during thermal migration and a non-steady-state Soret diffusion should be considered for future investigations. The calculations indicate that thermal migration and non-steady-state Soret diffusion are qualitatively sufficient to offset the Richter effect, which may be taken as a reason for the absence of Mg isotope fractionation in natural samples.

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