Extreme Mg and Zn isotope fractionation recorded in the Himalayan leucogranites

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**Abstract**

High-silica granites (typically >70 wt. % SiO₂) represent the products of extreme crustal differentiation, but whether their distinctive chemical compositions reflect source lithology or are produced via magmatic differentiation is commonly difficult to discriminate. To provide new insights into this issue, here we present high-precision Mg and Zn isotope data for high-silica, peraluminous leucogranites from the Himalayan orogen. The samples are subdivided into two-mica leucogranite, tourmaline leucogranite and garnet-bearing leucogranite based on their mineral assemblages. The two-mica leucogranites, representing the least evolved Himalayan leucogranites, have similar \( \delta^{66}Zn_{JMC \, 3-0749L} \) (mean = 0.31 ± 0.06‰) but heavier \( \delta^{26}Mg_{DSM-3} \) values (mean = -0.01 ± 0.12‰) relative to more mafic igneous rocks. This indicates that they were formed by anatexis of weathered silicates, consistent with the well-acknowledged metasedimentary source of the Himalayan leucogranites. In contrast, the more evolved tourmaline leucogranites and garnet-bearing leucogranites have lower \( \delta^{26}Mg \) and higher \( \delta^{66}Zn \) values compared with the two-mica leucogranites. The extremely low \( \delta^{26}Mg \) (-1.32‰ to -0.54‰) and high \( \delta^{66}Zn \) values (0.35‰ to 0.69‰) of garnet-bearing leucogranites vary systematically with indices of granitic differentiation (e.g., Zr/Hf, K/Rb, Eu/Eu*, 1/TiO₂). Although exsolution of chlorine-rich fluid may result in elevated \( \delta^{66}Zn \) values, it is unlikely to explain the low \( \delta^{26}Mg \) signatures of the same samples. Analysis of major Mg-bearing minerals suggests that substantial segregation of tourmaline and/or Fe-Ti oxide could have driven the differentiated leucogranites towards very low \( \delta^{26}Mg \) values. In this regard, the slightly lower \( \delta^{26}Mg \) values (mean = -0.17 ± 0.06‰) of tourmaline leucogranites relative to the two-mica leucogranites may also reflect that the former was more differentiated than the latter. Thus, although source heterogeneity may be responsible for the Mg isotopic variations observed in some high-silica granites, our study implies that high-silica granites could be remarkably heterogeneous in terms of Mg...
isotopes primarily as a result of prolonged fractional crystallization at the late stage of melt evolution. The anomalously light Mg and heavy Zn isotopic signatures of garnet-bearing leucogranites highlight that Mg and Zn isotopes may be treated as important makers of highly fractionated granites in future studies.

**Keywords:** High-silica granite; Mg isotopes; Zn isotopes; Himalayan leucogranite; Granite differentiation.

1. **INTRODUCTION**

High-silica granites, here referred to those with SiO$_2$ > 70% wt. %, comprise one of the most evolved igneous rocks of the Earth’s continental crust. They have high concentrations of highly incompatible elements such as K and Rb (Deering and Bachmann, 2010; Lee and Morton, 2015) and are often associated with mineralization of many economic elements (e.g., W, Sn, Nb, Ta; Linnen, 1998; Ballouard et al., 2016; Wang et al., 2017a). Therefore, considerable attention has been paid to unraveling their petrogenesis. One key issue worthy of investigation is whether the strongly peraluminous feature of high-silica granites is produced by partial melting of peraluminous sources (e.g., weathered supracrustal materials) or via extreme crystal-melt fractionation (Chappell and White, 1992; Chappell, 1999; Wu et al., 2017). Discriminating between the two mechanisms is essential for a better understanding of the compositional evolution of the continental crust. However, melts generated in these two ways may have very similar chemical compositions, especially if the source rocks have a wide range of compositions that are comparable to those of a differentiated sequence (e.g., Stevens et al., 2007). Furthermore, major elements (e.g., Si, Al, K, Na, Mg) do not vary greatly during prolonged fractionation of high-silica melts since the crystallizing assemblage is compositionally similar to the remaining melt (Chappell, 1999; Lee and Morton, 2015), making it difficult to distinguish strongly fractionated granites from unFractionated or weakly fractionated ones.

The stable isotopes of rock-forming metal elements have been widely applied to investigating the origin of high-silica granites in recent years. The best-known example is the unique Fe isotopic signature of high-silica igneous rocks that is shifted towards higher values relative to less evolved rocks, explained as a result of change in the valence states of iron in melts during fluid exsolution, fractional crystallization, and/or partial melting (e.g., Poitrasson and Freydier, 2005; Heimann et al., 2008; Sossi et al., 2012; Telus et al., 2012; Xia et al., 2017). If redox change is the principal factor driving isotope fractionation in high-silica granites, the magnitude of isotope fractionation of monovalent metal elements such as Mg and Zn should be expected to be limited. However, available data show that high-silica granites also have highly variable Mg and Zn isotopic compositions.
The broad range of Mg isotopic compositions observed in high-silica granites has been attributed to source heterogeneity caused by involvement of supracrustal weathered materials in earlier studies (Shen et al., 2009; Li et al., 2010; Liu et al., 2010; Jo et al., 2016). Since continental weathering preferentially incorporates lighter Mg isotopes into the hydrosphere and leaves isotopically heavy Mg in the weathered residue (Teng et al., 2010b; Liu et al., 2014b), anatexis of the weathered materials potentially generates high-$\delta^{26}$Mg granites. On the other hand, extremely low $\delta^{26}$Mg values (down to -1.00‰) have been reported in some highly fractionated granites (Ke et al., 2009; Telus et al., 2012; Lee et al., 2018), but its origin is poorly known and may be related to magmatic differentiation.

There is also limited data which suggests that pegmatites and a few high-silica granites are enriched in heavier Zn isotopes relative to low-silica granites (Telus et al., 2012; Doucet et al., 2018; Xu et al., 2019). The origin of high-$\delta^{66}$Zn granites is controversial and has commonly been interpreted as being due to the removal of $^{66}$Zn-depleted chloride by fluid exsolution and/or isotope fractionation during crustal melting and magmatic differentiation (Telus et al., 2012; Doucet et al., 2018; Xu et al., 2019). Collectively, the causes of Mg and Zn isotope fractionation in high-silica granites are still not fully understood.

The mid-Eocene–Miocene leucogranites in the Himalayan orogen comprise one of the largest granite belts in the world and represent classic examples of high-silica granites. They thus provide an outstanding opportunity for studying the origin of Mg and Zn isotopic variations in high-silica granites. These granites are generally acknowledged as pure crustal melts derived from anatexis of metasedimentary rocks given the strongly peraluminous compositions (Deniel et al., 1987; France-Lanord et al., 1988; Patiño Douce and Harris, 1998; Searle et al., 2009; Guo and Wilson, 2012; Weinberg, 2016; Gao et al., 2017). The origin of compositional variations in the Himalayan leucogranites was commonly attributed to source heterogeneity, types of crustal anatexis, or variable degrees of partial melting (Harris et al., 1993; Guillot and Le Fort, 1995; Patiño Douce and Harris, 1998; Visonà and Lombardo, 2002; Gao et al., 2017). However, several recent studies have argued an alternative model of extreme fractional crystallization (Liu et al., 2014c, 2016b, 2019; Wu et al., 2017). In this study, we report the first Mg and Zn isotope data on the Himalayan leucogranites. To determine the individual effect of magmatic differentiation and source heterogeneity, samples with diverse mineralogical and geochemical properties and emplaced in different sites were selected to investigate. The new Mg and Zn isotope data demonstrate that the Himalayan leucogranites were derived from partial melting of weathered silicate materials and underwent extensive magmatic differentiation before emplacement.
2. GEOLOGY AND SAMPLE DESCRIPTION

The Himalayan orogen, which is defined by the Indus-Yarlung suture (IYS) in the north and the Indian shield in the south, was formed following the closure of Neo-Tethys Ocean and subsequent Indian-Asian continental collision since the early Eocene (Fig. 1a; Yin, 2006). The orogen is further divided into three main sequences from south to north: the Lesser Himalayan Sequence (LHS), the High (Greater) Himalayan Sequence (HHS) and the Tethyan (North) Himalayan Sequence (THS). These sequences are separated by two north-dipping Late Cenozoic fault systems: the Main Central Thrust (MCT) and the South Tibetan Detachment System (STDS) (Fig. 1b).

Voluminous and discontinuous leucogranite plutons were emplaced within the HHS and THS during the mid-Eocene–Miocene, constituting two subparallel east-west trending granite belts extending ~2200 km in length (Searle et al., 2009; Zeng et al., 2015; Weinberg, 2016). These leucogranites are generally classified into three subtypes according to their mineral assemblages, i.e., two-mica leucogranite, tourmaline leucogranite and garnet-bearing leucogranite (e.g., King et al., 2011; Guo and Wilson, 2012; Liu et al., 2014c; Weinberg, 2016). The samples investigated in this study are from five locations, including all of the three subtypes. Detailed petrological and geochemical descriptions are provided below.

2.1 Xiaru

The Xiaru dome, exposed in the middle section of the THS (Fig. 1b), is formed of a two-episode granitic pluton with subordinate Paleozoic granitic orthogneiss (~480 Ma) at the margin and dominant younger leucogranite (~35 Ma) intruded into the granitic orthogneiss in the center. The granitic orthogneiss is mainly composed of quartz, plagioclase, K-feldspar, biotite and muscovite, while the leucogranite typically consists of quartz, plagioclase, K-feldspar, muscovite, tourmaline and minor garnet. Four orthogneisses and six garnet-bearing leucogranites were chosen in this study. These samples have been well characterized by Liu et al. (2016b). In general, the leucogranites have high SiO$_2$ contents (73.46–74.55 wt. %) and very low MgO contents (0.06–0.18 wt. %), characterized by strongly peraluminous compositions with A/CNK (molar ratio of Al$_2$O$_3$/(CaO+Na$_2$O+K$_2$O)) = 1.13–1.30 (Fig. 2b). They display relatively flat and tetrad effect-like REE patterns with pronounced negative Eu anomalies with Eu/Eu* (Eu$^\text{N}$/\sqrt{Sm^N+Gd^N}, where N denotes chondrite-normalized value) = 0.18–0.59 (Fig. S1). In particular, the Xiaru leucogranites are characterized by subchondritic Zr/Hf, K/Rb, Nb/Ta and Y/Ho ratios typical of highly evolved granitic melt (Liu et al., 2016b). The orthogneisses also have very high SiO$_2$ contents (73.45–75.35 wt. %) and many geochemical signatures analogous to the leucogranites, implying a certain extent of localized assimilation of orthogneiss during emplacement of the leucogranites (Liu et al., 2016b).
2.2 Ramba and Langkazi

The Ramba and Langkazi domes are located in the vicinity of the north-south trending Yadong rift within the THS (Fig. 1b). The Miocene (8-12 Ma) two-mica leucogranites, composed of quartz, plagioclase, K-feldspar, muscovite and biotite, were emplaced in the core of these domes (Liu et al., 2014c; Zheng et al., 2016). Five samples from Ramba and six samples from Langkazi were studied here. The Ramba samples have $\text{SiO}_2$ of 73.02–74.55 wt. % and MgO of 0.16–0.24 wt. % and are strongly peraluminous with A/CNK = 1.16-1.20 (Fig. 2). The Ramba two-mica leucogranites were thought to originate from muscovite and biotite dehydration melting of metapelites (Liu et al., 2014c) and have undergone fractional crystallization in a crystal mush environment (Liu et al., 2019). In comparison, the Langkazi samples have lower $\text{SiO}_2$ contents (68.82–70.13 wt. %), higher MgO contents (0.58–0.83 wt. %) and less peraluminous compositions (A/CNK = 1.00–1.14). All two-mica leucogranites from the two regions are enriched in LREE relative to HREE, but the Ramba samples display lower Eu/Eu* values relative to the Langkazi samples (0.39–0.50 vs. 0.57-0.85) (Fig. S1). The distinctive geochemical signatures of the Langkazi leucogranites, together with the finding of dioritic enclaves in these samples, indicate derivation of them by mixing of crustal melt with mantle-derived dioritic melt (Zheng et al., 2016).

2.3 Yadong

The Yadong leucogranites occurring as dikes, sills and small intrusions were emplaced in both the HHS and THS during the Miocene (12–23 Ma) (Fig. 1b). Two types of leucogranites were identified based on the modal proportions of tourmaline and biotite, i.e., two-mica leucogranite and tourmaline leucogranite (Guo and Wilson, 2012; Gou et al., 2016). Five tourmaline leucogranites investigated here typically contain plagioclase, K-feldspar, quartz, muscovite, tourmaline and rare biotite. They have high $\text{SiO}_2$ (72.33–74.04 wt. %) and low MgO (0.07–0.18 wt. %) contents and are strongly peraluminous (A/CNK = 1.13-1.20) (Fig. 2). They display LREE-enriched patterns with Eu/Eu* = 0.27–0.50 (Fig. S1). The Yadong tourmaline leucogranites were considered to form by muscovite-dehydration melting in previous studies (Gou et al., 2016).

2.4 Luozha

The Luozha leucogranites crop out along the STDS in the eastern Himalaya (Fig. 1b), and were intruded into the THS in the north and into the HHS in the south during the Miocene (12-20 Ma) (Guo and Wilson, 2012; Huang et al., 2017; Tian et al., 2017). The leucogranites include the two subtypes of two-mica leucogranite and tourmaline leucogranite. Four samples from Luozha were selected for Mg-Zn isotope analysis. Among them, one is two-mica leucogranite (CB-295) consisting of quartz, plagioclase, K-feldspar, biotite and muscovite, and three are tourmaline leucogranites consisting of quartz, plagioclase, K-feldspar,
muscovite, tourmaline and rare biotite. The two-mica leucogranite has slightly lower SiO$_2$ (72.33 wt. %) and higher MgO (0.54 wt. %) contents relative to the tourmaline leucogranites with SiO$_2$ of 73.76–73.93 wt. % and MgO of 0.13–0.16 wt. %. Both the two types of granites are strongly peraluminous (A/CNK = 1.14–1.18) (Fig. 2) and show LREE-enriched patterns with Eu/Eu* = 0.36–0.87 (Fig. S1). These leucogranites were proposed to form by muscovite dehydration melting of metasediments previously (Huang et al., 2017).

3. METHODS

Sample digestion, ion chromatographic separation and isotope ratio measurement follow the procedure detailed in previous studies (Liu et al., 2014a, 2016a; Ke et al., 2016), and only a brief description is presented below. To ensure ~10 µg Mg and ~1 µg Zn for isotope analyses, 5–50 milligrams of whole-rock powders or pre-cleaned mineral separates containing over 20 grains were dissolved in a mixture of HF-HNO$_3$-HCl acids. After complete dissolution, samples were transferred into 1 ml of 1 M HNO$_3$ or 8 M HCl for Mg and Zn purification, respectively. Magnesium was purified using Bio-Rad® AG50W-X8 resin. For samples with high Mn/Mg (e.g., garnet), an additional column filled with 1 ml AG50W-X8 resin in 0.5 M HCl+95% acetone was used after the method of Bizzarro et al. (2011). Separation of Zn was achieved using Bio-Rad® AG-MP-1M resin. The purified solutions were first analyzed for Mg or Zn concentration using MC-ICP-MS by comparing $^{24}$Mg and $^{64}$Zn signals with those of standards with known concentrations. Total procedure blank was <10 ng for Mg and <6 ng for Zn, which accounts for negligible portions of the collected Mg and Zn (<0.001% and <0.006%, respectively).

Magnesium and zinc isotopic ratios were measured separately using a Thermo Scientific Neptune plus multicollector inductively coupled plasma mass spectrometry (MC-ICP-MS) at the Isotope Geochemistry Laboratory, China University of Geosciences, Beijing. Isotope measurements were carried out in low resolution modes under wet plasma conditions. About 7 V signal of $^{24}$Mg and 3 V signal of $^{64}$Zn (Faraday cups with resistors of 10$^{11}$ Ω) can be yielded for solutions containing ~400 ng/g Mg and ~200 ng/g Zn, respectively. Instrumental mass fractionation was corrected by using sample-standard bracketing (SSB) method for Mg isotopes and by using both SSB and Cu-doped regression methods for Zn isotopes. The Cu-doped regression method was modified after that reported by Maréchal et al. (1999) and 100 ng/g of Cu was added to each sample and Zn standard solution as the external element for mass bias correction. Potential isobaric interferences from $^{23}$NaH, $^{25}$MgH, $^{24}$MgAr and $^{26}$MgAr were monitored and they are always unresolvable by the signal of blank acid. The results are reported in delta notation against reference Mg standard DSM-3 and Zn standard Lyon JMC 3-0749L, expressed as:

$$\delta^{26, 25}\text{Mg (‰)} = \left(\frac{^{26, 25}\text{Mg} / ^{24}\text{Mg}}{^{26, 25}\text{Mg} / ^{24}\text{Mg}}\right)_{\text{DSM3}} - 1 \right) \times 1000$$
The long-term external reproducibility (2SD) is better than ±0.06‰ for δ²⁶Mg (Ke et al., 2016) and ±0.04‰ for δ⁶⁶Zn (Wang et al., 2017b) based on repeated analyses of synthetic solutions and rock standards. Three USGS reference materials BHVO-2, AGV-2 and GSP-2 processed through the whole chemical procedure yielded results consistent with the literature values (Table 1) (e.g., Sossi et al., 2015; Chen et al., 2016; Teng, 2017). The uncertainties are given as 2SD throughout the paper, unless noted otherwise.

4. RESULTS

Whole-rock Mg and Zn isotope data are presented in Table 1, along with SiO₂, MgO, and Zn contents. The data for mineral separates are shown in Table 2. The complete dataset of major and trace elements are given in the appendix Table A3.

4.1 Magnesium isotopic compositions

The δ²⁶Mg values of the investigated leucogranites vary widely from -1.32‰ to 0.13‰, exceeding the δ²⁶Mg range of granitoids reported in previous studies (-1.00‰ to 0.44‰; Fig. 3a). Two-mica leucogranites from Ramba, Langkazi and Luoza define a narrow range of δ²⁶Mg values (-0.07‰ to 0.13‰) that are all above the average value of global basalts (-0.26 ± 0.07‰; Teng et al., 2010a). Tourmaline leucogranites from Yadong and Luoza have δ²⁶Mg values ranging from -0.22‰ to -0.14‰, lower than those of the two-mica leucogranites. Garnet-bearing leucogranites from Xiaru show large δ²⁶Mg variations (-1.32‰ to -0.54‰) and have the lowest δ²⁶Mg value among granites reported so far. The δ²⁶Mg of orthogneisses from Xiaru are from -0.36‰ to -0.29‰. Garnets (-2.35‰ to -1.20‰), tourmalines (-0.73‰ to -0.38‰) and muscovites (-0.83‰ to -0.65‰) in garnet-bearing leucogranites are also highly variable (Fig. 4a). In a given sample, inter-mineral isotope fractionation follows the order: δ²⁶Mg_tourmaline > δ²⁶Mg_muscovite > δ²⁶Mg_garnet. The garnet has much lower δ²⁶Mg values relative to other minerals with Δ²⁶Mg_garnet-tourmaline of -1.63‰ to -0.82‰ and Δ²⁶Mg_garnet-muscovite of -1.53‰ to -0.49‰ (ΔX_A–B = δX_A − δX_B, where X is the isotope of interest). Resolvable Mg isotope fractionation between tourmaline and muscovite also exists with Δ²⁶Mg_tourmaline–muscovite = 0.10‰ to 0.33‰. Similar Mg isotope fractionation (0.15‰ to 0.24‰) has also been observed between tourmaline and other silicates such as chlorite, omphacite and glaucophane (Pogge von Strandmann et al., 2015).

4.2 Zinc concentrations and isotopic compositions

The Zn concentrations are 31‒61 μg/g for two-mica leucogranites, 33‒49 μg/g for tourmaline leucogranites, 22–37 μg/g for garnet-bearing leucogranites, and 15‒25 μg/g for orthogneisses (Fig. 2d). The δ⁶⁶Zn values range from 0.28‰ to 0.46‰ for two-mica leucogranites, 0.34‰ to 0.54‰ for tourmaline
leucogranites, 0.35‰ to 0.69‰ for garnet-bearing leucogranites, and 0.36‰ to 0.48‰ for orthogneisses (Fig. 3b). Several samples have \( \delta^{66}\text{Zn} \) values higher than the average value of global basalts (0.26 ± 0.08‰; Chen et al., 2013; Wang et al., 2017b; Huang et al., 2018; McCoy-West et al., 2018; Sossi et al., 2018), and also exceed the \( \delta^{66}\text{Zn} \) range of granitoids reported so far (0.12‰–0.49‰; Telus et al., 2012, Doucet et al., 2018; Xu et al., 2019). Compositions which approach the highest \( \delta^{66}\text{Zn} \) values of the garnet-bearing leucogranites have also been observed in pegmatites from Black Hills, South Dakota, USA (0.53‰–0.88‰; Telus et al., 2012) and leucosomes in migmatites from Dabie orogen, central China (up to 0.81‰; Xu et al., 2019). Mineral separates from garnet-bearing leucogranites have \( \delta^{66}\text{Zn} \) values varying from 0.34‰ to 0.63‰ for garnet, 0.39‰ to 0.59‰ for tourmaline, and 0.42‰ to 0.92‰ for muscovite (Fig. 4b). In a given sample, inter-mineral isotope fractionation follows the order: \( \delta^{66}\text{Zn}_\text{muscovite} > \delta^{66}\text{Zn}_\text{tourmaline} \approx \delta^{66}\text{Zn}_\text{garnet} \), with \( \Delta^{66}\text{Zn}_\text{garnet–tourmaline} = -0.05‰ \) to -0.04‰, \( \Delta^{66}\text{Zn}_\text{garnet–muscovite} = -0.28‰ \) to -0.12‰, and \( \Delta^{66}\text{Zn}_\text{tourmaline–muscovite} = -0.32‰ \) to -0.07‰.

5. DISCUSSION

5.1 Inter-mineral Mg and Zn isotope fractionation in garnet-bearing leucogranites

The equilibrium inter-mineral isotope fractionation is primarily dependent on the bond strength and typically, heavier isotopes prefer to incorporation into stronger bonds (e.g., Urey, 1947; Schaubule, 2004; Young et al., 2015). In general, the bond strength is inversely related to the bond length or coordination number of cations in minerals. The Mg\(^{2+}\) is 8-fold coordinated in garnet ([X\(_3\)Y\(_2\)(SiO\(_4\))]\(_3\), X = Mg\(^{2+}\), Fe\(^{2+}\), Ca\(^{2+}\), Mn\(^{2+}\); Y = Al\(^{3+}\), Fe\(^{3+}\)) and 6-fold coordinated in tourmaline ([XY\(_3\)Z\(_6\)(SiO\(_4\))\(_3\)(BO\(_3\))\(_3\)(OH, F)\(_4\)], X = Na\(^+,\) K\(^+\), Ca\(^{2+}\); Y = Fe\(^{2+}\), Mg\(^{2+}\), Mn\(^{2+}\); Z = Al\(^{3+}\), Fe\(^{3+}\), Mg\(^{2+}\)) and muscovite ([XY\(_2\) (AlSi\(_3\)O\(_10\))(OH)\(_2\)], X = Na\(^+\), K\(^+\); Y = Al\(^{3+}\), Fe\(^{3+}\), Mg\(^{2+}\)). Therefore, the different bonding environments result in remarkably lighter Mg isotopic composition of garnet relative to tourmaline and muscovite, which was similarly observed between garnet and other minerals with 6-fold coordinated Mg\(^{2+}\) (e.g., olivine, pyroxene, mica; Li et al., 2011; Huang et al., 2013; Wang et al., 2015; Pogge von Strandmann et al., 2015; Wang et al., 2016; An et al., 2017; Stracke et al., 2018). As Zn\(^{2+}\) mainly substitutes for Mg\(^{2+}\) in the crystal lattices of minerals due to their similar ionic radius (Le Roux et al., 2010), inter-mineral Zn isotope fractionation is expected to be similar to Mg isotopes. However, the relative mass difference of different Zn isotopes is much smaller compared with Mg isotopes, rendering the magnitude of Zn isotope fractionation significantly reduced since mass-dependent isotope fractionation generally becomes smaller for elements with greater atomic mass.

Notably, the Mg and Zn isotope fractionation between tourmaline and muscovite is in an opposite direction. Tourmaline is a cyclic silicate with Mg\(^{2+}\) accommodated in both the Y and Z sites, while Zn\(^{2+}\) appears to only replace Mg\(^{2+}\) in the Y site (Bosi and Lucchesi, 2007; Watenphul et al., 2016). The Z-O bond
is generally shorter than the Y-O bond (Hawthorne, 2002; Ertl et al., 2006), meaning that cation in the Z site is likely enriched in heavier isotopes relative to that in the Y site. This perhaps explains the different Mg and Zn isotopic signatures observed in tourmaline, but it requires confirmation by theoretical studies.

5.2 Origin of bulk Mg and Zn isotopic variations in the Himalayan leucogranites

The leucogranites studied are quite fresh in both hand specimen and thin section (Liu et al., 2016b; Zheng et al., 2016), and have very low LOI contents (≤ 1 wt. %; appendix Table A3) that are not correlated with either δ²⁶Mg or δ⁶⁶Zn, ruling out the influence of post-magmatic alteration. Both field observations and geochemical features imply a certain extent of localized assimilation of country rock (granitic orthogneiss) during the emplacement of Xiaru garnet-bearing leucogranites (Liu et al., 2016b). These granites have much lower δ²⁶Mg and higher δ⁶⁶Zn values than those of the orthogneisses (Fig. 3), indicating that country rock contamination is unlikely to explain the extreme Mg and Zn isotopic compositions. As mentioned in section 5.1, garnet is strongly ²⁶Mg-depleted relative to other coexisting minerals owing to its much weaker Mg-O bond strength. It is thus necessary to assess whether the low δ²⁶Mg values of garnet-bearing leucogranites are a result of garnet enrichment due to heterogeneous mineral distribution. Notably, other major Mg-bearing minerals such as tourmaline and muscovite in the garnet-bearing leucogranites also have remarkably lower δ²⁶Mg values than the garnet-free granites, providing strong evidence that these minerals crystallized from low-δ²⁶Mg melts. In addition, garnet is also depleted in ⁶⁶Zn relative to other phases (Fig. 4b), but the garnet-bearing leucogranites have higher δ⁶⁶Zn values compared to the garnet-free granites, which is also contrary to the effect of garnet enrichment. Thus, the large Mg and Zn isotopic variations in the Himalayan leucogranites must reflect either isotope fractionation during igneous processes or compositional heterogeneity of sources.

5.2.1 Magmatic differentiation

Isotope fractionation during fractional crystallization of basaltic magma is almost indiscernible within current analytical uncertainties for Mg (Teng et al., 2007; Schiller et al., 2017), and is also limited for Zn (<0.1‰; Chen et al., 2013; Wang et al., 2017b; McCoy-West et al., 2018). This is primarily due to the absence of Mg and Zn isotope difference between the major Mg- and Zn-bearing minerals (e.g., olivine, pyroxene) and the bulk rock (Liu et al., 2011; Wang et al., 2017b). However, our results imply that minerals crystallizing from evolved, high-Si melts have more variable isotopic compositions, with concomitantly large fractionation factors, compared to minerals that typically crystallize from mafic melts. The large inter-mineral Mg and Zn isotope fractionation among minerals (i.e., garnet, tourmaline, and muscovite) in the garnet-bearing leucogranites may lead to significant isotope fractionation during granite differentiation.

Even if there is no obvious relationship between SiO₂ and MgO or Zn contents for the garnet-bearing leucogranites (Fig. 2), many geochemical characteristics indicate that they have undergone strong
differentiation. For example, the very low \( \text{Eu}/\text{Eu}^* \), \( \text{Zr}/\text{Hf} \) and \( \text{K}/\text{Rb} \) ratios and high \( 1/\text{TiO}_2 \) ratios indicate late-stage differentiation of granitic melt due to aqueous fluid-melt interaction or fractional crystallization of specific minerals such as feldspar and zircon (Bau, 1996; Förster et al., 1999; Irber, 1999; Jahn et al., 2001; Deering and Bachmann, 2010). Good correlations between \( \delta^{26}\text{Mg} \) or \( \delta^{66}\text{Zn} \) values and the above indicators are observed in the Xiaru garnet-bearing leucogranites (Figs. 5-6), implying that Mg and Zn isotopic variations in these samples can be caused by magmatic differentiation.

Zinc is highly compatible in mafic minerals and oxides (\( D_{\text{Zn}}^{\text{mafic mineral or Fe-Ti oxide/melt}} = 7.8-58.9 \)), but incompatible in feldspars (\( D_{\text{Zn}}^{\text{feldspar/melt}} = 0.04-0.23 \)) during differentiation of high-silica melt (Ewart and Griffin, 1994). Therefore, fractional crystallization of mafic minerals and oxides is the most likely mechanism lowering the Mg and Zn contents and driving isotope fractionation in the melt during magmatic differentiation.

There are good correlations between \( \delta^{66}\text{Zn} \) or \( \delta^{26}\text{Mg} \) values with proxies for feldspar fractionation (i.e., \( \text{Eu}/\text{Eu}^* \) and \( \text{K}/\text{Rb} \)) in the garnet-bearing leucogranites. Feldspar and quartz dominate the crystallizing assemblage, while mafic minerals and oxides, the major Mg and Zn budget, only account for minor proportion during differentiation of high-silica granites (Deering and Bachmann, 2010). Thus, the MgO and Zn contents of residual melt will not change dramatically if the crystallizing assemblage is compositionally similar to the melt with respect to Mg and Zn. This probably explains the lack of obvious correlations between isotope composition and element abundance of Mg and Zn.

The light Mg and heavy Zn isotope signatures of the garnet-bearing leucogranites require the fractionating minerals to possess higher \( \delta^{26}\text{Mg} \) and lower \( \delta^{66}\text{Zn} \) values than the melt. Tourmaline with the highest \( \delta^{26}\text{Mg} \) and lowest \( \delta^{66}\text{Zn} \) among minerals analyzed is the most likely candidate. Also, magnetite and ilmenite are isotopically enriched in \( ^{26}\text{Mg} \) relative to coexisting melts (Chen et al., 2018; Xie et al., 2018; Sedaghatpour and Jacobsen, 2018), and tend to have lower \( \delta^{66}\text{Zn} \) than coexisting melts (Chen et al., 2013; Xu et al., 2019). The excellent negative correlation between \( \delta^{26}\text{Mg} \) and \( 1/\text{TiO}_2 \) (\( R^2 = 0.99 \)) supports the fractionation of Fe-Ti oxides in controlling the Mg isotopic variation of the garnet-bearing leucogranites (Fig. 5d). Another mechanism that can result in heavier Zn isotopic compositions in the residual melt is fluid exsolution (Telus et al., 2012; Xu et al., 2019). The details of this mechanism will be discussed in section 5.3.

5.2.2 Source composition effect

As discussed above, the large Mg and Zn isotope fractionation during differentiation of garnet-bearing leucogranites makes it impossible to infer source compositions based on Mg and Zn isotopes of these rocks. Two-mica leucogranites are considered to be less differentiated than garnet-bearing and tourmaline leucogranites (Liu et al., 2019). Despite their variable contents of major and trace elements, the two-mica leucogranites have relatively homogenous Mg isotopic compositions (mean = \(-0.01 \pm 0.12\%_o\); \( n = 10 \)). Except
for sample LKZ-2 with $\delta^{66}$Zn of 0.46‰, other two-mica leucogranites have Zn isotopic compositions (mean $= 0.31 \pm 0.06$‰; n = 10) indistinguishable from those of more mafic rocks (e.g., global basalts). By contrast, some tourmaline leucogranites have high $\delta^{66}$Zn, low Zr/Hf ratios and Zn concentrations, similar to the garnet-bearing leucogranites (Figs. 5-7), indicating that their Zn isotopic systematics have been influenced by fluid exsolution or fractional crystallization. The slightly lower $\delta^{26}$Mg values of tourmaline leucogranites than the two-mica leucogranites, coupled with lower contents of MgO and other compatible trace elements such as Sr and Ba (Fig. 8), may also reflect a higher degrees of fractional crystallization of the tourmaline leucogranites than the two-mica leucogranites. Therefore, we conclude that the Mg and Zn isotopic compositions of two-mica leucogranites are least affected by magmatic differentiation and closest to those of the primary melts.

The $\delta^{26}$Mg values of two-mica leucogranites are apparently higher than those of more mafic rocks and the average upper continental crust (~0.22‰; Li et al., 2010). Given that Mg isotope fractionation during crustal anatexis is limited (Liu et al., 2010), the elevated $\delta^{26}$Mg values of two-mica leucogranites should inherit from their source rocks. Unlike igneous rocks (except for high-silica granites) that show limited variations in $\delta^{26}$Mg (Teng, 2017), weathered siliciclastic sediments or rocks often have more variable and heavier Mg isotopic compositions relative to the igneous protoliths (Teng et al., 2010b; Liu et al., 2014b). The Zn isotopic compositions of siliciclastic sediments or rocks are similar to or slightly lighter than those of igneous rocks owing to the small Zn isotope fractionation during chemical weathering (Moynier et al., 2017 and references therein; Suhr et al., 2018). The heavy Mg isotope signatures of two-mica leucogranites thus indicate the presence of weathered siliciclastic materials in their sources. This view is consistent with the source of Himalayan leucogranites consisting dominantly of metasedimentary rocks, as supported by the strongly peraluminous compositions and other isotope signatures such as the extremely high $^{87}$Sr/$^{86}$Sr ratios (~0.72–0.88) and heavy $\delta^{18}$O values (~10‰–13‰) in the Himalayan leucogranites (Deniel et al., 1987; France-Lanord et al., 1988; Guillot and Le Fort, 1995; Patiño Douce and Harris, 1998; Guo and Wilson, 2012; Liu et al., 2018). Furthermore, the limited Mg and Zn isotopic variations in two-mica leucogranites from different locations suggest that the source of Himalayan leucogranites is homogenous in terms of Mg and Zn isotopic compositions.

5.3 Modelling Mg and Zn isotope fractionation during magmatic evolution

5.3.1 Fluid exsolution

Volatile-rich fluid exsolution can occur as magma ascends and cools down. Considerable amounts of elements are sequestered from the melt into the vapor phase during this process (e.g., Audétat and Pettke, 2003; Williams-Jones and Heinrich, 2005; Zajacz et al., 2008), leading to compositional change and perhaps isotope fractionation in the residual melt. Fluids or vapors in equilibrium with rhyolitic or granitic melts are
commonly enriched in halogens, particularly chlorine with concentrations up to 50 wt. % (Urabe, 1987; Heinrich et al., 1992; Audétat and Pettke, 2003; Zajacz et al., 2008). Zinc readily bonds with Cl and thus prefers to incorporate into the vapor phase during fluid exsolution. This is supported by the very high fluid/melt partitioning coefficient of Zn \((D_{\text{Zn}}^{\text{fluid/melt}})\), which is strongly dependent on the Cl content of fluids \((D_{\text{Zn}}^{\text{fluid/melt}} = 8.2 + 232.7 \times [\text{Cl}])\); Zajacz et al., 2008). Provided that the initial melt has a Zn concentration of 55 \(\mu\)g/g, the highest value among most of the Himalayan leucogranites (Figs. 2c, 6a), the mass fraction of exsolved fluids can be calculated using the Rayleigh fractionation equation \([[\text{Zn}]]_{\text{melt}} = [[\text{Zn}]]_{\text{initial melt}} \cdot (1-F)^{(D_{\text{Zn}}^{\text{fluid/melt}}-1)}\), where \(F\) is the mass fraction of exsolved fluids). The results show that only 1–3% mass fraction of Cl-rich fluid \(([\text{Cl}] = 10–50\text{ wt. %})\) exsolved from melt is sufficient to generate the lowest Zn concentration of the studied leucogranites, while for pure aqueous fluid the fraction could be as high as 13% (Fig. 7c).

In the absence of experimentally determined Zn isotope fractionation factor between fluid and melt, a fractionation factor of \(\Delta^{66}\text{Zn}_{\text{fluid-silicate}} = -0.099 \cdot 10^6 \cdot T^{-2}\) or \(\Delta^{66}\text{Zn}_{\text{fluid-silicate}} = -0.084 \cdot 10^6 \cdot T^{-2}\) (\(T\) in K) is roughly estimated from the reduced partition function ratios of aqueous Zn\(^{2+}\), ZnCl\(^{+}\), ZnCl\(_2\), and Zn-silicate (hemimorphite) determined by ab initio calculations (Fig. 7b; Fujii et al., 2011; Ducher et al., 2018). The \(\Delta^{66}\text{Zn}_{\text{fluid-melt}}\) is then calculated to be about -0.15‰ when fluid exsolution occurs at ~600 °C and ZnCl\(_2\) is the major vapor phase. The evolution of melt \(\delta^{66}\text{Zn}\) was modelled using the following Rayleigh equation: \(\frac{\delta^{66}\text{Zn}_{\text{melt}}}{\delta^{66}\text{Zn}_{\text{initial melt}}} = f_{\text{Zn}} \cdot \frac{\delta^{66}\text{Zn}_{\text{fluid-melt}}}{\delta^{66}\text{Zn}_{\text{initial melt}}} \cdot \ln(f_{\text{Zn}})\), where \(f_{\text{Zn}} \left(1-F \cdot \frac{[[\text{Zn}]]_{\text{melt}}}{[[\text{Zn}]]_{\text{initial melt}}}\right)\) is the fraction of Zn remained in residual melt. The results show that \(\leq 4.5\%\) mass fraction of Cl-rich fluid \(([\text{Cl}] = 20–50\text{ wt. %})\) exsolved from the melt is able to increase the \(\delta^{66}\text{Zn}\) of residual melt to values comparable to that of the garnet-bearing leucogranite (Fig. 7d). Accordingly, fluid exsolution is a viable mechanism for generating high-\(\delta^{66}\text{Zn}\) values in the Himalayan leucogranites. In contrast to Zn, Mg is barely soluble in fluids exsolved from high-silica melts (Audétat and Pettke, 2003; Zajacz et al., 2008) and light Mg isotopes appear to be preferentially incorporated into the fluid (e.g., Tipper et al., 2010). Therefore, it is unlikely to produce the extremely variable and low Mg isotopic ratios of some Himalayan leucogranites by fluid exsolution.

5.3.2 Crystal segregation

As discussed above (section 5.2.1), crystal-melt separation may exert an important role in Mg isotope fractionation in the Himalayan leucogranites, especially the garnet-bearing leucogranites. Although the high viscosity and the low-density contrast of granitic melts make it difficult for crystal to isolate from melt like crystal-basalt segregation, the differentiated granitic melt could be extracted from the out layer of a crystal
mush in the form of hindered settling (Lee and Morton, 2015). A recent study on the Himalayan leucogranite suggested that the muscovite leucogranite, same as tourmaline leucogranite and garnet-bearing leucogranite, could have been extracted from a crystal mush during high-degree crystallization (≈59%–64%), while the two-mica leucogranite represents the residual crystal mush composed of cumulate and trapped interstitial melt (Liu et al., 2019). Here, we adopted the "crystal mush" model simulating the Mg isotope fractionation in the Himalayan leucogranites. The details of the calculations are shown below and all modelling parameters are given in Table 3.

It is difficult to determine the composition of the initial or primary melt directly and we can only make assumptions here. Although the garnet-bearing leucogranites and two-mica leucogranites differ in age and site of emplacement, their chemical compositions are broadly similar to those of the same type of rocks observed in other Himalayan leucogranites. For example, the Xiaru and Ramba garnet-bearing leucogranites are quite similar in terms of major and trace elements, even if the age difference is ≈27 Ma (Liu et al., 2014c, 2016b). As stable isotopes do not change with time, we speculate that the Mg isotopic compositions of our samples are common features of other Himalayan leucogranites with similar chemical compositions. Therefore, using the same method employed by Liu et al. (2019), we assume the average composition of the studied two-mica leucogranites to be similar to that of the residual crystal mush, and the most evolved garnet-bearing leucogranite (sample 110923) represents the pure melt extracted from the crystal mush.

The MgO content ([MgO]) and $\delta^{26}\text{Mg}$ value of the initial melt can be calculated using mass balance equations: $[\text{MgO}]_{\text{initial melt}} = [\text{MgO}]_{\text{extracted melt}} \cdot X + [\text{MgO}]_{\text{crystal mush}} \cdot (1-X)$ (1) and $\delta^{26}\text{Mg}_{\text{initial melt}} = \delta^{26}\text{Mg}_{\text{extracted melt}} \cdot [\text{MgO}]_{\text{extracted melt}} \cdot X + \delta^{26}\text{Mg}_{\text{crystal mush}} \cdot [\text{MgO}]_{\text{crystal mush}} \cdot (1-X)$ (2), where $X$ is the fraction of melt extracted from the crystal mush and assumed to be 0.2 based on the modelling result of Gelman et al. (2014). The evolution of melt [MgO] and $\delta^{26}\text{Mg}$ during fractional crystallization was modelled using the Raleigh equation: $[\text{MgO}]_{\text{melt}} = [\text{MgO}]_{\text{initial melt}} \cdot (1-F)^{D_{\text{Mg}}^{-1}}$ (3), where $F$ is the degree of fractional crystallization and $D_{\text{Mg}}$ is the portioning coefficient between crystal and melt; $\left(\frac{2^{26}\text{Mg}}{2^{25}\text{Mg}}\right)_{\text{melt}} = \left(\frac{2^{26}\text{Mg}}{2^{25}\text{Mg}}\right)_{\text{initial melt}} \cdot fMg \cdot a_{\text{crystal-melt}} \cdot 1$ (4), which can be transformed into: $\delta^{26}\text{Mg}_{\text{melt}} = \delta^{26}\text{Mg}_{\text{initial melt}} + \Delta^{26}\text{Mg}_{\text{crystal-melt}} \cdot \ln(fMg)$ (5), where $fMg \left(\frac{(1-F) \cdot [\text{MgO}]_{\text{melt}}}{[\text{MgO}]_{\text{initial melt}}}\right)$ is the fraction of Mg in residual melt, $a_{\text{crystal-melt}}$ is the Mg isotope fractionation factor between crystal and melt, and $\Delta^{26}\text{Mg}_{\text{crystal-melt}}$ is approximate to 1000ln($a_{\text{crystal-melt}}$).

The modelling results show that, in order to reproduce the observed MgO content and $\delta^{26}\text{Mg}$ value of the tourmaline leucogranites, a $\Delta^{26}\text{Mg}_{\text{crystal-melt}}$ value of ≈0.1‰ is required. For the garnet-bearing leucogranites, a $\Delta^{26}\text{Mg}_{\text{crystal-melt}}$ value of ≈0.2–0.3‰ is required (Fig. 8b). The required $\Delta^{26}\text{Mg}_{\text{crystal-melt}}$ values agree with the range of $\Delta^{26}\text{Mg}_{\text{tourmaline-bulk}}$ (0.10‰–0.46‰; this study) and $\Delta^{26}\text{Mg}_{\text{magnetite-bulk}}$ (0.25‰–0.50‰; Xie et al., 2018).
observed in natural samples. In the case of garnet-bearing leucogranite, the maximum $F$ reaches as high as $\sim 70\%$. The concentrations of Ba and Sr are good indicators of the degree of granitic differentiation as Ba and Sr are strongly partitioned into the major minerals such as feldspar and mica (Liu et al., 2019). The $F$ for the garnet-bearing leucogranite independently obtained from Ba and Sr element modelling (using the equations 1–3) is up to $\sim 70\%$ (Fig. 8a), which is almost the same as that inferred from the Mg isotopes-based modelling (Figs. 8b-d). It should be noted that the modelled isotope fractionation factor is strongly dependent on the $D_{\text{crystal/melt}}$ value and Mg concentration of primary melt, both of which are variable in fact. Even so, our modelling shows that a small $\Delta^{26}\text{Mg}_{\text{crystal-melt}}$ value can significantly shift the Mg isotopic compositions of melt during extreme differentiation. Therefore, a high degree of fractional crystallization is essential to account for the anomalously low $\delta^{26}\text{Mg}$ values observed in the studied garnet-bearing leucogranites. A similar model has been recently proposed to explain the very low $\delta^{26}\text{Mg}$ compositions observed in lunar mare basalts, which is ascribed to the extreme Mg isotope fractionation at the end of lunar magma ocean differentiation (Sedaghatpour and Jacobsen, 2018).

Once the $F$ is determined, the average $[\text{MgO}]$ of cumulate crystallized from the melt can be calculated using the following equation: $[\text{MgO}]_{\text{cumulate}} = [\text{MgO}]_{\text{initial melt}} \cdot (1-F_D^{\text{Mg}})/(1-F)$. If the trapped melt in the crystal mush is the same as the extracted melt, the $\delta^{26}\text{Mg}$ of the cumulate can be estimated based on mass balance equation: $\delta^{26}\text{Mg}_{\text{crystal mush}} \cdot [\text{MgO}]_{\text{crystal mush}} = \delta^{26}\text{Mg}_{\text{cumulate}} \cdot [\text{MgO}]_{\text{cumulate}} \cdot (1-f_{\text{trap}})$ $+ \delta^{26}\text{Mg}_{\text{trapped melt}} \cdot [\text{MgO}]_{\text{trapped melt}} \cdot f_{\text{trap}}$, where $f_{\text{trap}} = \frac{[\text{MgO}]_{\text{cumulate}} - [\text{MgO}]_{\text{crystal mush}}}{[\text{MgO}]_{\text{trapped melt}} - [\text{MgO}]_{\text{crystal mush}}}$. The calculated $\delta^{26}\text{Mg}$ value of the average cumulate is $0.09\%$, only slightly heavier than the average of two-mica leucogranites (Table 3). Mixing calculation suggests that even if $\sim 70\%$ melt is trapped, the shift in $\delta^{26}\text{Mg}$ of residual crystal mush will not exceed $0.18\%$. This explains why the two-mica leucogranites are limitedly variable in $\delta^{26}\text{Mg}$ irrespective of a wide range of MgO contents (Fig. 8b).

### 5.4 Implication for recognition of highly fractionated granite

Highly fractionated (evolved) granites are commonly referred to as residual melts after intensive crystal differentiation. These granites are favored to be produced in extensional tectonic regimes and therefore, they can provide important information on the tectonic evolution of orogeny (Wu et al., 2017). For instance, the appearance of highly fractionated Himalayan leucogranites may indicate that the leucogranite magmatism occurs in an extensional tectonic environment during the post-collisional stage of Himalayan orogeny (Sylvester, 1998), which is consistent with the distribution of Himalayan leucogranites along the normal faults.
(i.e., STDS) and rifts (Fig. 1b). However, how to distinguish highly fractionated granites from more primitive granites is still a matter of debate.

The subtle variation in major element contents of high-silica granite makes it difficult to identify highly differentiated melt simply based on major elements (e.g., SiO$_2$, MgO; Gelman et al., 2014; Lee and Morton, 2015). Instead, highly fractionated granites are often characterized by tetrads effect in their REE distribution patterns and non-CHARAC (charge and radius controlled) trace element abundance ratios (Bau, 1996; Irber, 1999; Jahn et al., 2001; Wu et al., 2003). For instance, trace element ratios such as Zr/Hf, K/Rb, Nb/Ta and Y/Ho may be significantly reduced via magmatic-hydrothermal interaction or segregation of accessory minerals at the very late stage of granitic differentiation (Bau, 1996; Irber, 1999; Deering and Bachmann, 2010; Ballouard et al., 2016). Nonetheless, many A-type granites, which may have not experienced significant crystal-melt differentiation, have trace element ratios overlapping with highly fractionated granites (Whalen et al., 1987; Wu et al., 2017). It is also a challenge to quantify the degree of granite differentiation using these trace element ratios since magmatic-hydrothermal interaction may happen in both strongly fractionated and weakly fractionated granitic melts when magma cools down. Hence, new geochemical criteria that can effectively identify highly fractionated granites are required in addition to these element indices.

The Xiaru garnet-bearing leucogranites investigated in this study share many trace element characteristics of highly fractionated granites (Liu et al., 2016b), e.g., non-CHARAC trace element behaviors and tetrads REE patterns. Their extremely light Mg and heavy Zn isotopic compositions imply that these granites were formed by strong fractional crystallization involving Mg-bearing minerals, accompanied by fluid exsolution. Similar Mg-Zn isotopic anomalies have also been observed in one sample of Harney Peak granite from Black Hills, South Dakota, USA (Sample HP49A: $\delta^{26}$Mg = -0.72±0.09‰, $\delta^{66}$Zn = 0.49±0.09‰, ; Telus et al., 2012), which is also one of the representative examples of highly fractionated S-type granite (Shearer et al., 1987; Nabelek et al., 1992). In a recent study, Lee et al. (2018) found that some I- and A-type granites with tetrads REE patterns from Korea , display very variable and low $\delta^{26}$Mg values (-1.00‰ to 0.01‰) and they ascribed the Mg isotopic signatures to heterogeneous source compositions. However, granite source with such light Mg isotopic composition is rarely reported and in fact, the $\delta^{26}$Mg values of Korean granites, similar to the garnet-bearing leucogranites in this study, are well correlated with indices of magmatic differentiation (e.g., 1/TiO$_2$, Fig. 5d). Together, the low-$\delta^{26}$Mg and high-$\delta^{66}$Zn anomalies appear to be a typical feature of highly fractionated granites regardless of granite types (I-, A- and S-type). Therefore, Mg and Zn isotopes can be treated as a novel indicator for identifying highly fractionated granites. In order to develop Mg and Zn isotopes as robust tools for quantifying the degree of granite differentiation, experimental and theoretical constraints on the Mg and Zn isotope fractionation factors between high-silica melt and Mg-Zn bearing minerals are demanded in future studies.
6. CONCLUSIONS

The Himalayan leucogranites analyzed in this study show a broad spectrum of Mg and Zn isotopic compositions that far exceeds the range of low-silica granitoids. Garnet-bearing leucogranites exhibit extremely low $\delta^{26}\text{Mg}$ and high $\delta^{66}\text{Zn}$ values that are well correlated with indices of granite differentiation (e.g., Zr/Hf, K/Rb, Eu/Eu*, 1/TiO$_2$). Although their elevated $\delta^{66}\text{Zn}$ values may be explained by exsolution of chlorine-rich fluids, fluid exsolution only has a negligible impact on melt $\delta^{26}\text{Mg}$ due to the very low solubility of Mg in the exsolved fluids. Instead, substantial removal of Mg-bearing mineral (e.g., tourmaline, Fe-Ti oxide) from the melt is predicted to play the dominant role in controlling the Mg isotope fractionation in the garnet-bearing leucogranites. In combination with similar Mg and Zn isotopic compositions observed in highly fractionated granites from other regions, we propose that Mg and Zn isotopes have a potential for distinguishing between strongly and weakly fractionated high-Si granites.

For the less differentiated two-mica leucogranites analyzed in this study, their Mg and Zn isotopic compositions are more representative of the source signatures of the Himalayan leucogranites. The heavy Mg isotopic compositions of two-mica leucogranites relative to less felsic rocks agree with the hypothesis that their sources are mainly composed of supracrustal materials subjected to chemical weathering. This supports the derivation of Himalayan leucogranites from a metasedimentary source as previously proposed on the basis of other isotope systems (e.g., Sr, O). The slightly higher $\delta^{66}\text{Zn}$ and lower $\delta^{26}\text{Mg}$ values of tourmaline leucogranites compared to two-mica leucogranites perhaps implies that the former underwent more magmatic differentiation than the latter. Our study further confirms that Mg isotopes may be an excellent tracer of weathered crustal materials in granite sources, but the effect of magmatic differentiation should be evaluated when interpreting the data of high-silica granite.

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Table 1. Whole-rock Mg and Zn isotope compositions of samples investigated in this study.

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<th>Sample No.</th>
<th>Location</th>
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<th>δ²⁶Mg</th>
<th>2SD</th>
<th>δ⁸⁷Zn</th>
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<td>n.d.</td>
<td>0.30</td>
<td>0.02</td>
<td>0.58</td>
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<tr>
<td>CB-295</td>
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<td>72.54</td>
<td>0.54</td>
<td>56</td>
<td>0.07</td>
<td>0.01</td>
<td>0.13</td>
<td>0.02</td>
<td>0.34</td>
<td>0.03</td>
<td>0.68</td>
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<tr>
<td>YAD12-1-2</td>
<td>Yadong</td>
<td>72.33</td>
<td>0.18</td>
<td>42</td>
<td>n.d.</td>
<td>n.d.</td>
<td>0.36</td>
<td>0.03</td>
<td>0.71</td>
<td>0.08</td>
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<td>YAD12-1-3</td>
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<td>YAD12-3-1</td>
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<td>0.07</td>
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<td>0.01</td>
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<td>0.03</td>
<td>0.71</td>
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<td>0.07</td>
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<td>0.02</td>
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<td>YAD12-4-2</td>
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<td>CB-292</td>
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<td>0.02</td>
<td>0.76</td>
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</tr>
<tr>
<td>110918</td>
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<td>0.11</td>
<td>25</td>
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<td>0.06</td>
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<tr>
<td>12FW10</td>
<td>Xiaru</td>
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<td>0.06</td>
<td>24</td>
<td>-0.44</td>
<td>0.01</td>
<td>-0.84</td>
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<td>0.01</td>
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<tr>
<td>12FW14</td>
<td>Xiaru</td>
<td>74.55</td>
<td>0.18</td>
<td>27</td>
<td>-0.32</td>
<td>0.01</td>
<td>-0.63</td>
<td>0.03</td>
<td>0.35</td>
<td>0.03</td>
<td>0.70</td>
<td>0.04</td>
</tr>
<tr>
<td>12FW15</td>
<td>Xiaru</td>
<td>74.04</td>
<td>0.16</td>
<td>23</td>
<td>-0.42</td>
<td>0.05</td>
<td>-0.83</td>
<td>0.06</td>
<td>0.40</td>
<td>0.05</td>
<td>0.79</td>
<td>0.08</td>
</tr>
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<td>110922</td>
<td>Xiaru</td>
<td>73.45</td>
<td>0.54</td>
<td>22</td>
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<td>n.d.</td>
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<td>0.39</td>
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<td>-0.15</td>
<td>0.02</td>
<td>-0.34</td>
<td>0.02</td>
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<td>0.72</td>
<td>0.07</td>
</tr>
<tr>
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<td>0.01</td>
<td>-0.36</td>
<td>0.02</td>
<td>0.37</td>
<td>0.05</td>
<td>0.73</td>
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</tr>
<tr>
<td>GSP-2</td>
<td></td>
<td>0.04</td>
<td>0.01</td>
<td>0.04</td>
<td>0.02</td>
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<td>AGV-2</td>
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<td>BHVO-2</td>
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<td>0.05</td>
<td>-0.21</td>
<td>0.06</td>
<td>0.32</td>
<td>0.06</td>
<td>0.64</td>
<td>0.11</td>
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<td></td>
</tr>
</tbody>
</table>

Major element is in wt.%. Zn concentration is in μg/g. Mg and Zn isotope ratios are in permil. The SiO₂ and MgO contents of Xiaru and Langkazi samples are from Liu et al. (2016b) and Zheng et al. (2016), respectively. Other data are reported in this study for the first time. 2SD represents two standard deviations of the population of more than three batches of repeated measurement on the same solution.
Table 2. Mg and Zn isotopic data of mineral separates from Xiaru leucogranite.

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Mineral</th>
<th>MgO</th>
<th>$\delta^{25}$Mg</th>
<th>2SD</th>
<th>$\delta^{26}$Mg</th>
<th>2SD</th>
<th>Zn</th>
<th>$\delta^{66}$Zn</th>
<th>2SD</th>
<th>$\delta^{68}$Zn</th>
<th>2SD</th>
</tr>
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<tbody>
<tr>
<td>12FW10</td>
<td>Garnet</td>
<td>0.05</td>
<td>-1.22</td>
<td>0.02</td>
<td>-2.35</td>
<td>0.03</td>
<td>92</td>
<td>0.63</td>
<td>0.05</td>
<td>1.23</td>
<td>0.10</td>
</tr>
<tr>
<td></td>
<td>Tourmaline</td>
<td>0.88</td>
<td>-0.37</td>
<td>0.03</td>
<td>-0.73</td>
<td>0.04</td>
<td>1014</td>
<td>0.59</td>
<td>0.04</td>
<td>1.16</td>
<td>0.08</td>
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<tr>
<td></td>
<td>Muscovite</td>
<td>0.08</td>
<td>-0.42</td>
<td>0.05</td>
<td>-0.83</td>
<td>0.08</td>
<td>167</td>
<td>0.92</td>
<td>0.04</td>
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<tr>
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<td>Garnet</td>
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<td>-0.78</td>
<td>0.02</td>
<td>-1.50</td>
<td>0.03</td>
<td>75</td>
<td>0.36</td>
<td>0.03</td>
<td>0.71</td>
<td>0.11</td>
</tr>
<tr>
<td></td>
<td>Tourmaline</td>
<td>0.43</td>
<td>-0.28</td>
<td>0.03</td>
<td>-0.54</td>
<td>0.04</td>
<td>593</td>
<td>0.37</td>
<td>0.04</td>
<td>0.74</td>
<td>0.08</td>
</tr>
<tr>
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<td>Muscovite</td>
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<td>-0.35</td>
<td>0.05</td>
<td>-0.65</td>
<td>0.04</td>
<td>90</td>
<td>0.54</td>
<td>0.02</td>
<td>1.06</td>
<td>0.08</td>
</tr>
<tr>
<td>12FW15</td>
<td>Garnet</td>
<td>0.18</td>
<td>-0.69</td>
<td>0.02</td>
<td>-1.20</td>
<td>0.04</td>
<td>71</td>
<td>0.34</td>
<td>0.04</td>
<td>0.67</td>
<td>0.08</td>
</tr>
<tr>
<td></td>
<td>Tourmaline</td>
<td>1.30</td>
<td>-0.20</td>
<td>0.03</td>
<td>-0.38</td>
<td>0.07</td>
<td>515</td>
<td>0.39</td>
<td>0.04</td>
<td>0.77</td>
<td>0.08</td>
</tr>
<tr>
<td></td>
<td>Muscovite</td>
<td>0.09</td>
<td>-0.40</td>
<td>0.03</td>
<td>-0.71</td>
<td>0.07</td>
<td>110</td>
<td>0.46</td>
<td>0.04</td>
<td>0.90</td>
<td>0.08</td>
</tr>
</tbody>
</table>

Major element is in wt. %. Zn concentration is in µg/g. Mg and Zn isotope ratios are in per mil. 2SD represents two standard deviations of the population of more than three batches of repeated measurement on the same solution.
Table 3 Parameters for modelling Mg isotope fractionation during fractional crystallization

<table>
<thead>
<tr>
<th></th>
<th>MgO (wt.%)</th>
<th>Ba (µg/g)</th>
<th>Sr (µg/g)</th>
<th>δ²⁶Mg (‰)</th>
<th>Bulk partitioning coefficient</th>
</tr>
</thead>
<tbody>
<tr>
<td>Extracted melt</td>
<td>0.06</td>
<td>4</td>
<td>12</td>
<td>-1.36</td>
<td>$D_{\text{Mg}}$ = 4.0</td>
</tr>
<tr>
<td>Residual crystal mush</td>
<td>0.47</td>
<td>525</td>
<td>212</td>
<td>-0.01</td>
<td>$D_{\text{Ba}}$ = 4.6</td>
</tr>
<tr>
<td>Initial melt</td>
<td>0.39</td>
<td>421</td>
<td>172</td>
<td>-0.05</td>
<td>$D_{\text{Sr}}$ = 3.5</td>
</tr>
<tr>
<td>Cumulate</td>
<td>0.98</td>
<td></td>
<td>0.09</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The extracted melt and residual crystal mush are assumed to have compositions same as sample 110923 and the average two-mica leucogranite, respectively. The compositions of initial melt and cumulate are calculated using mass balance equations (see details in the main text). Bulk partitioning coefficient for Mg is roughly estimated assuming that the fractionating phases contain 10% mafic minerals with $D_{\text{Mg}}^{\text{melt/mineral}} \approx 40$ (Nagasawa and Schnetzler, 1971). Bulk partitioning coefficients for Ba and Sr are from Liu et al. (2019).
**Figure 1:** Schematic maps showing the major tectonic units of the Tibetan-Himalayan orogenic belt (a) and the distribution of Himalayan leucogranites (after Liu et al., 2014c) (b). Abbreviations: MBT–Main Boundary Thrust; MCT–Main Central Thrust; STDS–South Tibetan Detachment System; IYS–Indus-Yarlung Suture.

**Figure 2:** (a) Na$_2$O + K$_2$O versus SiO$_2$ diagram for plutonic rocks classification (after Wilson, 2007). (b) A/NK (molar ratio of Al$_2$O$_3$/ (Na$_2$O+K$_2$O)) versus A/CNK (molar ratio of Al$_2$O$_3$/ (CaO + Na$_2$O + K$_2$O)) diagram (after Maniar and Piccoli, 1989). (c) MgO versus SiO$_2$ diagram. (d) Zn versus SiO$_2$. The SiO$_2$, MgO and Zn contents of the studied samples are shown in Table 1 and other element abundances are given in the appendix. Literature data of Himalayan leucogranites are also shown for comparison and the data sources are given in Wu et al. (2017). Abbreviations are as same as the following figures: 2MG–two-mica leucogranite; TG–tourmaline leucogranite; GG–garnet-bearing leucogranites.

**Figure 3:** Variations of bulk rock $\delta^{26}$Mg (a) and $\delta^{66}$Zn (b) with SiO$_2$. The blue band represents the average value of global basalts with two standard deviations ($\delta^{26}$Mg = -0.26 ± 0.07‰, Teng et al., 2010a; $\delta^{66}$Zn = 0.26 ± 0.08‰, Chen et al., 2013; Wang et al., 2017b; Huang et al., 2018; McCoy-West et al., 2018; Sossi et al., 2018). Data of the studied samples are presented in Table 1. Data of volcanic rocks and granitoids reported in literature are also shown for comparison. The full dataset of rocks with SiO$_2$ ≥ 50 wt. % is given in the appendix Tables A1 and A2.

**Figure 4:** Comparisons of $\delta^{26}$Mg (a) and $\delta^{66}$Zn (b) values among mineral separates of Xiaru garnet-bearing leucogranite. The blue band represents the average isotopic composition of global basalts with two standard deviations ($\delta^{26}$Mg = -0.26 ± 0.07‰, Teng et al., 2010a; $\delta^{66}$Zn = 0.26 ± 0.08‰, Chen et al., 2013; Wang et al., 2017b; Huang et al., 2018; McCoy-West et al., 2018; Sossi et al., 2018). Data are shown in Tables 1 and 2.

**Figure 5:** Variations of $\delta^{26}$Mg with Zr/Hf (a), K/Rb (b), Eu/Eu$^*$ (c) and 1/TiO$_2$ (d). The $\delta^{26}$Mg values are presented in Table 1 and element ratios are given in the appendix. Data for Korean granitoids are from Lee et al. (2018).

**Figure 6:** Variations of $\delta^{66}$Zn with Zr/Hf (a), K/Rb (b), Eu/Eu$^*$ (c) and 1/TiO$_2$ (d). The $\delta^{66}$Zn values are presented in Table 1 and element ratios are given in the appendix.

**Figure 7:** (a) Relationship between $\delta^{66}$Zn and Zn concentration for the Himalayan leucogranites. The grey
Temperature dependence of Zn isotope fractionation factors obtained from first-principal calculation (Fujii et al., 2011; Ducher et al., 2018). The yellow band corresponds to the range of calculated zircon saturation temperatures ($T_{Zr}$, data shown in the appendix) of the studied Himalayan leucogranites. (c, d) Modelled Zn concentrations and $\delta^{66}$Zn values for residual granitic magmas as a function of the mass fraction of fluid exsolved from melt. Curves with different colors refer to scenarios under which the exsolved fluids have different Cl contents. Zinc partitioning coefficient ($D_{\text{Zn}}^{\text{fluid/melt}}$) is after Zajacz et al. (2008). Modelling details are described in the main text.

**Figure 8:** (a) Relationship between Ba and Sr concentration for the Himalayan leucogranites. The black line shows the evolution of melt compositions during fractional crystallization of an assumed granitic melt. (b-d) Relationship between $\delta^{26}$Mg and MgO, Ba or Sr content for the Himalayan leucogranites. The dashed curves in b, c and d represent the $\delta^{26}$Mg evolution during fractional crystallization of an assumed granitic melt based on a Rayleigh fractionation model with $\Delta^{26}$Mg$_{\text{crystal-melt}}$ varying from 0.1‰ to 0.3. The red solid curve in b represents mixing of the most evolved melt, represented by sample 110923, with the estimated cumulates. The grey band refers to the average Mg isotopic composition of two-mica leucogranites. Numbers shown in a, c and d refer to the degree of fractional crystallization. Modelling details are given in the main text. The $\delta^{26}$Mg values and MgO contents are presented in Table 1. The Ba and Sr concentrations are given in the appendix.
Figure 3

(a) Average δ²⁶Mg values for different localities in China:
- Ramba (2MG)
- Langkazi (2MG)
- Luozha (2MG)
- Yadong (TG)
- Luozha (TG)
- Xiuru (GG)
- Xiuru (orthogneiss)

Average basalt (-0.26± 0.07‰)

(b) Average δ⁶⁶Zn values for different rock types:
- Volcanic rocks (literature)
- Granitoids (literature)
- Pegmatites (literature)
- Leucosomes in migmatites (literature)

Average basalt (0.26± 0.08‰)
Figure 4

(a) δ²⁶Mg (‰)

(b) δ⁶⁶Zn (‰)

Average basalt

12FW10 12FW14 12FW15

Grt Tur Mus Bulk
Figure 8

(a) Sr (µg/g) vs. Ba (µg/g) for various samples: Ramba (2MG), Langkazi (2MG), Luozha (2MG), Yadong (TG), Luozha (TG), and Xiara (GG).

(b) MgO (wt.%) vs. δ²⁶Mg (‰) for Cumulates and Two-mica leucogranite. Lines indicate different Δ²⁶Mg values: -0.3‰, -0.2‰, and -0.1‰.

(c) δ²⁶Mg (‰) vs. Ba (µg/g) for Initial melt.

(d) δ²⁶Mg (‰) vs. Sr (µg/g) for Initial melt.

Cumulates
110923

Two-mica leucogranite

Δ²⁶Mg_{crystal−melt} = -0.3‰
Δ²⁶Mg_{crystal−melt} = -0.2‰
Δ²⁶Mg_{crystal−melt} = -0.1‰