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Petrogenesis of Himalayan leucogranites: perspective from a combined elemental and Fe-Sr-Nd isotope study

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Key Points:
- The first integrated Fe-Sr-Nd isotopic and geochemical dataset of Himalayan leucogranites and metasedimentary rocks are presented.
- Isotopic and geochemical data suggest that two-mica and tourmaline leucogranites have not experienced intensive fractional crystallization.
- The low δ⁵⁶Fe values of garnet leucogranites were likely derived from garnet accumulation.
Abstract

The petrogenesis of Himalayan leucogranites remains crucial for understanding the thermal and tectonic evolution of the Himalayan orogen. In order to understand whether they are largely pristine melts of crustal anatexis or have experienced intensive fractional crystallization, we present Fe isotopic data of 30 representative Himalayan leucogranites and 9 local metasedimentary rocks. Excepting three garnet leucogranites with low $\delta^{56}Fe$ (-0.04‰ to 0.06‰) that are likely affected by garnet accumulation, tourmaline and two-mica leucogranites have largely homogeneous $\delta^{56}Fe$ from 0.13‰ to 0.24‰ irrespective of their highly variable SiO$_2$, MgO and FeOt contents. Combined with observed mineral assemblages and available fractionation factors, this does not support intensive fractional crystallization (with or without assimilation) in their petrogenesis. The elevated $\delta^{56}Fe$ elevation relative to the supposed source rocks, represented by metasedimentary rocks and/or metabasite with a $\delta^{56}Fe$ value of 0.10, by ~0.07‰, may reflect Fe isotope fractionation during crustal anatexis. This study indicates most leucogranites can provide robust constraints on the conditions of crustal anatexis and thus the thermal and tectonic evolution of the Himalayan orogen.

Keywords: Himalaya; Leucogranite; Iron isotopes; Petrogenesis
Plain Language Summary

The Himalayas have been formed from the collision between two tectonic plates. During their formation the rocks of the continental crust have melted to form leucogranites which potentially provide important information on how the collision process evolves. Several recent studies of rare-element mineralization associated with these granites have argued that the magmas result from extensive removal of early formed minerals during the cooling of the magma (fractional crystallization) which, if true, would undermine their usefulness as monitors of the collisional process. In this study we address this issue through a geochemical approach that combines isotopic data from iron, strontium and neodymium. Whereas Sr and Nd give information on the source of the magmas, the isotopes of Fe will remain largely unfractionated if the granites result simply from melting the crust but fractionate significantly during fractional crystallization. Our results reveal very limited fractionation of Fe isotopic compositions for two types of leucogranites, which is inconsistent with an intensive fractional crystallization model but supports the interpretation that they represent largely unfracionated crustal melts. Our study therefore confirms that Himalayan leucogranites can provide reliable probes for the thermal and tectonic evolution of the Himalayan crust.
1. Introduction

Himalayan leucogranites are silica-rich and strongly peraluminous; they have been widely proposed to be primary melts of crustal anatexis, given their compositions comparable to experimental melts of local meta-sediments and crustal radiogenic (Sr-Nd-Pb-Hf) and stable isotopic (H-O-Si) compositions (Deniel et al., 1987; France-Lanord et al., 1988; Hopkinson et al., 2017; Liu et al., 2018; Patiño Douce and Harris, 1998). Their petrological and geochemical variation was considered to be derived from different melting mechanisms, e.g., dehydration melting or fluid-present melting of a single source (metapelite) or multiple sources (metapelite and metagraywacke or amphibolite) under different P-T-XH2O conditions (Guillot and Le Fort, 1995; Inger and Harris, 1993; Knesel and Davidson, 2002; Le Fort, 1975; Patiño Douce and Harris, 1998; Zeng et al., 2011a). For example, at high pressure (~ 1 GPa), fluid-present melting of muscovite would generate leucogranite melts with high Na/K and CaO contents (Gao et al., 2017; Patiño Douce and Harris, 1998; Zhang et al., 2004). While at low pressure (~ 6 kbar), muscovite dehydration melting would generate leucogranite melts with high Rb/Sr and low Na/K, Sr and Ba contents (Harris and Inger, 1992; Patiño Douce and Harris, 1998). More radiogenic Sr-Nd-Hf isotopes in some Himalayan intrusions suggests that melting source contains a contribution from the Lesser Himalayan sequence (Guo and Wilson, 2012; Hopkinson et al., 2019). Some leucogranites in southern Tibet with high Sr/Y ratios and relatively unradiogenic Sr-Nd isotopic compositions indicate partial melting of amphibolite under thickened crustal condition (Ji et al., 2020; Liu et al., 2014; Zeng et al., 2011a, 2011b, 2019). Thus, the Himalayan leucogranites have been used to reflect the P-T-XH2O conditions and the tectonic evolution of Himalayan orogenic belt (Aikman et al., 2008; Gao and Zeng, 2014; Hopkinson et al., 2019; Huang et al., 2017; Weinberg, 2016).
In contrast, the chemical variation of Himalayan leucogranites has been considered by some recent studies to result from late differentiation (Liu et al., 2014, 2019; Wang et al., 2017; Wu et al., 2020). Overall, most Himalayan leucogranites have strong negative Eu anomalies, low Zr/Hf and Nb-Ta ratios as well as a strong rare earth element tetrad effect that evolve with decreasing zircon saturation temperatures (Wu et al., 2017a, 2020). However these geochemical characteristics are equally consistent with varying melt fractions from sedimentary sources, an interpretation supported by petrographic evidence which precludes the fractional crystallization of feldspars in leucogranite petrogenesis (Inger and Harris, 1993). Given their highly evolved compositions and recently documented rare-metal mineralization, the leucogranites from Xiaru, Ramba and Cuonadong plutons have been termed as highly fractionated granites (Liu et al., 2014, 2016; Xie et al., 2019). Accordingly, it has been proposed that most Himalayan leucogranites were generated by extremely intensive fractional crystallization (e.g., four stages of 75% fractionation; Wu et al., 2020). As highly fractionated granites, the characteristics of primary magmas for Himalayan leucogranites are difficult to constrain (e.g., Liu et al., 2016). Accordingly, previous conclusions on the thermal and tectonic evolution of Himalayan orogenic belt based on leucogranites may need reconsideration. Therefore, it is crucial to elucidate whether Himalayan leucogranites were generated by intensive magma differentiation or directly by crustal anatexis. This dilemma, however, is difficult to resolve, since both petrogenesis models can explain the mineral assemblage and chemical composition of Himalayan leucogranites (e.g., Fan et al., 2021; Gao et al., 2016b; Wu et al., 2017a).

An iron (Fe) isotopic study could resolve this dilemma and shed new light on the petrogenesis of Himalayan leucogranites, since Fe isotopes fractionate significantly during granitic magmatism (e.g., $\delta^{56}$Fe from 0.08‰ to 0.64‰ for granites with SiO$_2$ > 71 wt.% in Du et al., 2017; Foden et
Fe isotopic fractionation during granitic magmatism is regulated by the redox and chemical composition of the melt and co-existing mineral assemblages (Dauphas et al., 2014; He et al., 2017; Sossi and O’Neill, 2017). Phases hosting Fe with higher charge and lower coordination number prefer heavier isotopes. Mafic silicate minerals (e.g., biotite and amphibole) enriched in ferrous Fe are isotopically lighter, while magnetite ($\text{Fe}^{3+}/\Sigma\text{Fe} = 0.69$) is isotopically heavier than coexisting granitic melts with low $(\text{Na+K})/(\text{Ca+Mg})$ (Heimann et al., 2008; Sossi and O’Neill, 2017; Telus et al., 2012; Wu et al., 2017b). With increasing $(\text{Na+K})/(\text{Ca+Mg})$ in the melt, its Fe isotopes can be heavier than magnetite in turn (Dauphas et al., 2014; He et al., 2017). The $\delta^{56}\text{Fe}$ of granites directly generated by crustal anatexis deviate from their source rocks by a limited extent, i.e., less than $\Delta^{56}\text{Fe}_{\text{melt-residua}}$, exemplified by migmatites from Black Hills and the Dabie orogen where the apparent Fe isotope fractionation between leucosomes and melanosomes is about 0.10‰ (Telus et al., 2012; Xu et al., 2017). On the contrary, Fe isotope fractionation can become much larger during extremely intensive fractional crystallization of granitic melts, e.g., elevating the melt $\delta^{56}\text{Fe}$ by $>0.5‰$ (Dauphas et al., 2014; Du et al., 2017). This difference is resulted from that crustal anatexis is a batch process while fractional crystallization is a Rayleigh process.

In order to better understand their petrogenesis, here we report combined elemental and Fe-Sr-Nd isotopic data for thirty Himalayan leucogranites. We also measured nine metasedimentary samples from the Greater Himalayan Sequence, which represent the possible source of Himalayan leucogranites (Inger and Harris, 1993; Patiño Douce and Harris, 1998). The mineral separates from 5 selected leucogranite samples were also measured, because tourmaline, an important Fe hosting mineral in the leucogranite, has not been investigated for Fe isotopes hitherto. Based on new
constraints from Fe isotopic data, the role of partial melting and extreme fractional crystallization has been investigated in Himalayan leucogranites.

2. Geological Background and sample description

The Himalayan orogen rises as a consequence of the ongoing continental collision between India and Asia since the Paleogene (Fig. 1) (Ding et al., 2005; Hodges, 2000; Hu et al., 2016; Yin and Harrison, 2000; Zhu et al., 2015a). It is divided into four lithotectonic units: (i) the Tethyan Himalayan Sequence, bounded by the Indus-Tsangpo suture to the north and the Southern Tibetan Detachment System to the south and mainly composed of very low to low grade Neoproterozoic to Eocene marine sediments; (ii) the Greater Himalayan Sequence, lying tectonically below the Tethyan Himalayan Sequence and mainly comprised of medium to high grade metasedimentary and meta-igneous rocks; (iii) the Lesser Himalayan Sequence, bounded by the Main Central Thrust at the top and the Main Boundary Thrust at the bottom and made up of very-low grade to lower amphibolite facies metamorphic rocks with an age range of 1870 - 850 Ma; (iv) the Sub-Himalayan Sequence, as the lowermost tectonic unit, mainly composed of Neogene sediments deposited in the active Himalayan foreland basin (Carosi et al., 2018; Kohn, 2014; Yin, 2006). Two roughly parallel granite belts are identified due to their temporal and spatial differences in the Himalayan orogen (Yin and Harrison, 2000). The southern one is the High Himalayan granite belt and consists of Oligocene – Miocene granite dikes, sills and plutons intruded into the Greater Himalayan Sequence along with the Southern Tibetan Detachment System (Weinberg, 2016; Wu et al., 2015). The northern one is the Tethyan Himalayan granite or North Himalayan granite belt which intrudes into the Tethyan Himalayan Sequence as the North Himalayan Gneiss Dome (Zeng et al., 2011a).
Thirty Himalayan leucogranites and nine metasedimentary rocks were collected in this study (Fig.1, Tables S1 and S2), and the latter represent the exposed counterparts of source rocks for the former. The leucogranite samples can be divided into three types based on their petrology: two-mica, tourmaline and garnet leucogranites, which represent the major magmatic stage. The aplite, pegmatite and hydrothermal alteration granite which represent the magmatic-hydrothermal and hydrothermal stages, were not considered here, given the potential complex modification on whole rock $\delta^{56}$Fe during hydrothermal processes (e.g., Markl et al., 2006; Wang et al., 2011; Zhu et al., 2016). The two-mica leucogranites are composed of biotite, muscovite, feldspars and quartz. The other two types of leucogranites contain characteristic tourmaline and garnet instead of biotite respectively, except for sample YD1306. This sample, as a tourmaline leucogranite, also contains trace biotite. A detailed petrological description is given in Table S1. All leucogranite samples were collected distal from the pluton boundary without xenoliths and are recovered from both the Tethyan and High Himalayan granite belts (Fig.1). Metasedimentary samples were collected from the kyanite zone to sillimanite zone of Greater Himalayan Sequence in central Nepal and Bhutan. They range from schist to gneiss with the key pelitic mineral assemblages of muscovite ± biotite ± garnet ± kyanite ± sillimanite (Table S2). The muscovite-garnet schist (Sample N13) has been used as the starting material of melting experiments that succeeded in generating melts of Himalayan leucogranite compositions (Patiño Douce and Harris, 1998).

3. Analytical methods

All the samples were thoroughly cleaned, and the weathered surface layers were removed before crushing in a corundum jaw crusher to 60 mesh. Subsequently, the samples were powdered in an agate ring mill into 200 mesh. Whole-rock major and trace elements were measured at the
State Key Laboratory of Geological Processes and Mineral Resources (GPMR), Wuhan. Sample powders were mixed with Li$_2$B$_4$O$_7$, LiBr, and NH$_4$NO$_3$, and fused in a Pt crucible. Major elemental data were obtained on the prepared glasses using X-ray fluorescence spectrometry (Shimadzu XRF-1800). Loss on ignition (LOI) was measured gravimetrically (Table S3). Accuracy is better than 3% (Ma et al., 2012). For trace elements analysis, 50 mg whole-rock powders were dissolved by HF + HNO$_3$ in Teflon bombs, and then sealed at 190 °C for 48 h. The samples were dried, dissolved in 100 g 2% HNO$_3$, and then measured by Agilent 7500a (Table S3). The detailed analytical procedures have been previously reported (Liu et al., 2008). The accuracy is routinely better than 5% for the reported trace elements, which is confirmed by analyses of international rock standards (Table S4). The FeO contents were measured by redox titration using K$_2$Cr$_2$O$_7$ solution at China University of Geosciences, Beijing, and Fe$^{3+}$/ΣFe was calculated accordingly (i.e., (FeOt - FeO) / FeOt). The accuracy is <10% for the obtained Fe$^{3+}$/ΣFe (He et al., 2017). Mineral separation was handpicked under a binocular from coarsely crushed samples (80 ~ 120 mesh).

Whole rock powders for Sr-Nd isotopes were dissolved in the clean Teflon bombs with HF + HNO$_3$ + HClO$_4$, and then chemical separations were performed using conventional ion exchange procedures. Sr and Nd isotopes were analyzed using a Thermo-Finnigan TRITON thermal ionization mass spectrometer (TIMS) at the Tianjin Institute of Geology and Mineral Resources following the procedure by Liu et al. (2017). Sr and Nd isotopic ratios were corrected for instrumental fractionation by normalization against $^{86}$Sr/$^{88}$Sr = 0.1194 and $^{146}$Nd/$^{144}$Nd = 0.7219, respectively (Table S5). Repeated analyses of Sr standards BCR-2 and NBS987 yielded average $^{87}$Sr/$^{86}$Sr ratios of 0.704975 ± 0.000021 (2σ, n = 6) and 0.710218 ± 0.000005 (2σ, n = 7), respectively. The average $^{143}$Nd/$^{144}$Nd values of BCR-2 and a laboratory internal standard solution
(LRIG-Nd) are $0.512635 \pm 0.000004$ (2σ, n = 7) and $0.512201 \pm 0.000003$ (2σ, n = 8), which agrees well with the recommended values (Li et al., 2007).

Iron isotope analyses were conducted at the Isotope Geochemistry Laboratory, China University of Geosciences, Beijing, following the procedures previously established (Dauphas et al., 2009b; He et al., 2015; Zhu et al., 2018). Approximately 3 - 35 mg whole rock powders were dissolved in a 3:1 mixture of concentrated HF-HNO$_3$ in Teflon beakers on a hotplate at ~ 160 °C until the solutions became transparent. After evaporation to dryness at 140 °C, the residues were refluxed subsequently with aqua regia (HCl : HNO$_3$ = 7:3) and excess HNO$_3$ aqua regia (HCl : HNO$_3$ = 2:1) at 130 °C for two times. The samples were dissolved in 6 N HCl prior to chromatographic purification. Iron was separated from matrix elements and potential isobars with 1 ml AG1-X8 pre-clean resin in a HCl medium. Whole procedure blank is less than 10 ng for Fe and thus can be considered negligible compared to >50 μg sample Fe processed. Fe isotopic ratios were analyzed on a Neptune Plus multiple collector inductively coupled plasma mass spectrometer (MC-ICP-MS) on high-resolution modes, with mass bias corrected by the sample-standard bracketing method. The Fe isotopic data are reported in δ values relative to IRMM-014 ($\delta$$_{\text{Fe}}$(‰) = [($^{56}$Fe/$^{54}$Fe)$_{\text{sample}}$/($^{56}$Fe/$^{54}$Fe)$_{\text{IRMM-014}}$ − 1] × 1000, where i can be 56 or 57). The isotopic measurement sequence for each sample Fe solution was repeated four times, and the mean values are reported. The internal uncertainties are given as 2SE after Dauphas et al. (2009b) and He et al. (2015), considering the errors arising from both the chemical procedures and the MC-ICP-MS measurement, which are typically ≤0.05‰ for $\delta$$_{\text{Fe}}$ (Table S6). Both the long-term reproducibility and accuracy are better than 0.05‰ for $\delta$$_{\text{Fe}}$, which comes from the duplicate measurements on 24 geological standards over 7 years (He et al., 2015; Zhu et al., 2018). The USGS standard AGV-2 and GSP-2 were processed with unknown samples, yielding values ($\delta$$_{\text{Fe}}$ = 0.09 ± 0.04‰) and

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0.15 ± 0.04‰ respectively) consistent with the previously published values within quoted uncertainties (e.g., Craddock and Dauphas, 2011; Dauphas et al., 2009a; He et al., 2015; Sossi et al., 2012). Duplicate analyses of samples, independent from sample dissolution to instrument analysis, also show consistent results within quoted errors (Table S6).

4. Results

The Himalayan leucogranites show large geochemical variation, especially for two-mica leucogranites which can be divided into high Sr/Y and low Sr/Y groups, based on their Sr and Y contents and Sr-Nd isotopic distinction (Fig. 2a and 3). The low Sr/Y two-mica leucogranites have high SiO$_2$ (70.80 – 76.26 wt.%) and Al$_2$O$_3$ (12.49 – 15.69 wt.%) and low MgO (0.16 – 0.94 wt.%), TiO$_2$ (0.09 – 0.33 wt.%) and (Na + K)/(Ca + Mg) (2.94 – 15.23 wt.%) values (Fig. 2). They show high Rb (181 – 398 ppm) and varied Sr (35 – 262 ppm), Y (5.52 – 45.18 ppm) and Ba (52 – 615 ppm) contents, with Rb/Sr and Sr/Y ratios ranging from 0.80 to 7.32 and 0.77 to 22.39 respectively.

Compared to the low Sr/Y two-mica leucogranites, the high Sr/Y two-mica leucogranites show lower Rb (84 – 277 ppm) and Y (2.19 – 8.30 ppm) and higher Sr (157 – 801 ppm) and Ba (202 – 968 ppm) contents, along with lower Rb/Sr (0.11 – 1.76) and higher Sr/Y (26.47 – 111.17) ratios (Fig. 2). The tourmaline and garnet leucogranites both have TiO$_2$ (0.02 – 0.07 wt.%), Sr (13 – 72 ppm) and Ba (6 – 187 ppm) contents systematically lower than the low Sr/Y two-mica leucogranite. Garnet leucogranites have the lowest Mg$^+$ (14.84 – 27.63), Zr/Hf (11.15 – 19.31) and Eu/Eu* (0.10 – 0.29) among Himalayan leucogranites (Fig. 2). The Fe$^{3+}$/ΣFe ratios of these leucogranite samples range from 0.00 to 0.55, with an average of 0.17 and a median of 0.15 (Fig. 2f), similar to that of S-type granites (~ 0.15) from the Lachlan Fold Belt (Chappell and White, 1992).
Sr-Nd isotope data of these Himalayan leucogranites show large variations (Table S5). The $^{87}\text{Sr}/^{86}\text{Sr}(i)$ and $\varepsilon_{\text{Nd}}(t)$ of low Sr/Y two-mica, tourmaline and garnet leucogranites range from 0.7216 to 0.7674 and -16.0 to -12.4 respectively, showing affinity to the Himalayan metasedimentary rocks. While the Sr-Nd isotopic composition of high Sr/Y leucogranite are relatively unradiogenic ($^{87}\text{Sr}/^{86}\text{Sr}(i) = 0.7088$ to 0.7175; $\varepsilon_{\text{Nd}}(t) = -11.1$ to -10.2) and comparable to the metabasites in THS (Fig. 3 and Table S5).

Nine kyanite-zone to sillimanite-zone schists and gneisses from Bhutan and Nepal show relatively homogenous $\delta^{56}\text{Fe}$ ranging from 0.05‰ to 0.15‰ with an average of 0.10 ± 0.08‰ (2SD), similar to values from global clastic sediments and the mean upper continental crust ($\delta^{56}\text{Fe} \sim 0.10‰; \text{Beard et al., 2003; Foden et al., 2015}$) (Table S6 and Fig 4). The tourmaline and two-mica leucogranites yield slightly isotopically heavier $\delta^{56}\text{Fe}$, but with a limited range from 0.13‰ to 0.24 ‰ despite their large variations in petrology, elemental and Sr-Nd isotopic compositions (Fig. 4). Garnet leucogranites have $\delta^{56}\text{Fe}$ (-0.04‰ to 0.06‰) lower than the other two types of leucogranites. Compared to the literature data, garnet leucogranites represent the lowest $\delta^{56}\text{Fe}$ end-member of granites at a given SiO$_2$.

Biotite (0.12 – 0.24‰) and tourmaline (ca. 0.15‰) have relatively consistent values among different samples, while muscovite has variable $\delta^{56}\text{Fe}$ sample by sample, ranging from 0.20‰ to 0.45‰. In YD1306 where biotite co-exists with tourmaline, $\delta^{56}\text{Fe}_{\text{biotite}}$ is marginally higher than $\delta^{56}\text{Fe}_{\text{tourmaline}}$, with a difference of 0.08 ± 0.04‰. Garnet has the lightest Fe isotopic composition with a $\delta^{56}\text{Fe}$ of -0.11‰ (Fig. 5a). The similarity in $\delta^{56}\text{Fe}$ of the whole rock compared to biotite and tourmaline suggests that these minerals are the dominant Fe carrier in two-mica and tourmaline leucogranites respectively. This view is consistent with the observed mineral assemblages and EPMA analyses.
5. Discussion

5.1 Fe isotopic fractionation factors among minerals and melt

Iron isotope fractionation factors for a range of minerals and melt ($\Delta^{56}\text{Fe}_{\text{mineral-melt}}$) can be deduced from one certain mineral-melt fractionation and the inter-mineral fractionation using the equation $\Delta^{56}\text{Fe}_{\text{B-melt}} = \Delta^{56}\text{Fe}_{A\text{-melt}} + \Delta^{56}\text{Fe}_{B\text{-A}}$ (A and B represent different minerals). $\Delta^{56}\text{Fe}_{\text{biotite-melt}}$ (~0.1‰) was calculated through the fractionation between the leucosomes and melanosomes of Black Hills migmatites where biotite is the dominant Fe carrier in the residua (Telus et al., 2012). This $\Delta^{56}\text{Fe}_{\text{biotite-melt}}$ can be influenced by the redox state of Fe in the melt and biotite, because phases with higher $\text{Fe}^{3+}/\Sigma\text{Fe}$ prefer heavier isotopes (Dauphas et al., 2014; Sossi and O’Neill, 2017). For a granitic melt, biotite tends to have similar $\text{Fe}^{3+}/\Sigma\text{Fe}$ to the melt under reductive conditions (Baker and Rutherford, 1996; Cesare et al., 2005). The Himalayan leucogranites have relatively low $\text{Fe}^{3+}/\Sigma\text{Fe}$ similar to the S-type granites in the Lachlan Fold Belt (Fig. 2 and Chappell and White, 1992), and also to the Black Hills migmatites inferred to have formed under reducing conditions from the presence of graphite (Nabelek, 1999). Coordination of Fe in the melt tends to be reduced leading to a larger $\Delta^{56}\text{Fe}_{\text{biotite-melt}}$ with increasing $\text{SiO}_2$ and / or $(\text{Na+K})/(\text{Ca+Mg})$ (Dauphas et al., 2014; Foden et al., 2015; He et al., 2017; Sossi et al., 2012). Himalayan leucogranites have comparable $\text{SiO}_2$ contents (70.80 – 78.36 wt.%) and $(\text{Na+K})/(\text{Ca+Mg})$ values (2.94 – 15.50) to the leucosomes of Black Hills migmatites ($\text{SiO}_2$: 72.46 – 79.45 wt.%; $(\text{Na+K})/(\text{Ca+Mg})$: 0.08 – 18.37; Nabelek, 1999). Accordingly, it is reasonable to apply the $\Delta^{56}\text{Fe}_{\text{biotite-melt}}$ estimated on a basis of Black Hills migmatites to the fractionation between biotite and Himalayan leucogranite melts.
The Fe isotopic fractionation factor between biotite and tourmaline has not been well-calibrated yet. Since Fe$^{2+}$ occupies the octahedral site in both biotite and tourmaline (Hawthorne and Dirlam, 2011), they should have comparable Fe isotopic composition at the same Fe$^{3+}$/ΣFe ratio which is supported by the ab initio calculations (Nie et al., 2021). Tourmaline in the Himalayan leucogranite yields δ$^{56}$Fe slightly lower than biotite, with a Δ$^{56}$Fe$_{\text{biotite-tourmaline}}$ of 0.08 ± 0.04‰. While the Δ$^{56}$Fe$_{\text{biotite-tourmaline}}$ constrained from Moosilauke metapelite is about -0.06 ± 0.03‰ at 700 °C (Nie et al., 2021). This discrepancy may result from their different Fe$^{3+}$/ΣFe ratios and/or compositional variations in biotite and tourmaline (Nie et al., 2021; Wu et al., 2017b).

Muscovite, like biotite, belongs to the mica group, and Fe in these minerals both occupies the same coordination sites (e.g., Fe$^{2+}$ in the octahedral site), which suggests a Δ$^{56}$Fe$_{\text{biotite-muscovite}}$ near 0 if these two minerals have a similar Fe$^{3+}$/ΣFe. Δ$^{56}$Fe$_{\text{biotite-muscovite}}$ in the Himalayan leucogranites, however, ranges from -0.31‰ to 0.04‰ (Fig. 5). This indicates either isotope disequilibrium or mineral compositional control on isotope fractionation. Fe isotope equilibrium may have not been reached between muscovite and the other minerals, possibly due to the timing of their crystallization. For example, biotite crystallizes early near the liquidus while muscovite is a near-solidus phase (Scaillet et al., 1995). Thus some muscovite may occur within Fe-free domains of quartz and feldspar representing the last formed intergranular melt (Fig. S2), hence impeding Fe isotope exchange with early crystallized biotite. Muscovite may record the variably heavy Fe isotopic compositions of the intergranular melts under near solidus conditions, which suggests that the δ$^{56}$Fe of melt could be significantly elevated after substantial crystallization of the leucogranites.

Given its euhedral shape with few inclusions, the garnet investigated here is magmatic (Gao et al., 2012; Zeng et al., 2019), which is also consistent with its higher Mn contents (spessartine =
21 - 29%, Grossular <1.92%; Table S7) than the xenocrystic garnets (spessartine < 5%; Harris et al., 1992) and lower Ca content than the peritectic garnet (Grossular = 11 – 41%; King et al., 2011). Due to the lack of biotite in the garnet leucogranites, our data cannot provide direct constraints on the Fe isotopic fractionation between biotite and garnet. While $\Delta^{56}\text{Fe}_{\text{biotite}-\text{garnet}}$ has been previously well calibrated by measurements of biotite-garnet pairs from metamorphosed iron formations, yielding a positive $\Delta^{56}\text{Fe}_{\text{biotite}-\text{garnet}} \sim 0.09 \pm 0.05 \times 10^6/T^2$ (Ye et al., 2020). This translates to a $\Delta^{56}\text{Fe}_{\text{garnet}-\text{melt}} \sim -0.20‰$ at 700°C, a crystallization temperature typical for Himalayan leucogranites.

Despite their low Fe contents, feldspars become important Fe carrier in high-Si granites. The Fe isotopic fractionation factors between biotite and feldspars have been calibrated from a suite of I-type granitoids in the Dabie orogen (Wu et al., 2017b). The $\Delta^{56}\text{Fe}_{\text{biotite-plagioclase}}$ and $\Delta^{56}\text{Fe}_{\text{biotite-alkali-feldspar}}$ show linear relationships with the albite and orthoclase contents in plagioclase and alkali-feldspar respectively ($\Delta^{56}\text{Fe}_{\text{biotite-plagioclase}} = 0.015 \times \text{Ab}$% - 0.48; $\Delta^{56}\text{Fe}_{\text{biotite-alkali-feldspar}} = 0.026 \times \text{Or}$% - 1.35), reflecting the compositional control on Fe isotope fractionation. From the typical compositions of plagioclase and K-feldspar in the Himalayan leucogranites (Liu et al., 2019), the Ab% (in plagioclase) and Or% (in K-feldspar) were both assumed to be 90%, which indicates a $\Delta^{56}\text{Fe}_{\text{biotite-plagioclase}}$ of 0.77‰ and $\Delta^{56}\text{Fe}_{\text{biotite-K-feldspar}}$ of 0.89‰.

Accordingly, biotite, tourmaline, muscovite and garnet are all predicted to be Fe isotopically lighter, while feldspars are isotopically heavier than the co-existing leucogranitic melt (summarized in Table 1).

5.2 Iron isotope fractionation of Himalayan leucogranites
δ^{56}Fe of high-silica granites may be significantly elevated by the exclusion of isotopically light fluids, evidenced by negative correlations between δ^{56}Fe and Zr/Hf (Heimann et al., 2008; Poitrasson and Freydier, 2005). Note that melt Zr/Hf ratios may be also influenced by the dissolution / crystallization of zircon (Du et al., 2017; Gao et al., 2017), and thus not be applied to tracing fluid exsolution for Himalayan leucogranites. Despite this, δ^{56}Fe does not increase with decreasing Zr/Hf from 33.59 to 11.15, nor is it correlated with either TE_{1,3} (the degree of the lanthanide tetrad effect) or Y/Ho (Figs. 4c and 6), which together argues against a significant role of fluid exsolution in fractionating melt δ^{56}Fe. The result here supports the previous conclusion that change in δ^{56}Fe during fluid exsolution may not be quantitatively important for high-silica granitic melts (Du et al., 2017). Our leucogranite samples were collected far away from the pluton boundary and without xenoliths, and are spread across both the Tethyan and High Himalayan leucogranite belts, greatly exceeding the length-scale that diffusion-driven isotope fractionation may influence natural samples (Teng et al., 2011; Wu et al., 2018; Zambardi et al., 2014) (Fig. 1). Therefore, neither chemical nor thermal diffusion will be considered here.

It has been widely documented that the Himalayan leucogranites were generated by anatexis of metasedimentary rocks, except for the high Sr/Y two-mica leucogranites which are likely to have been derived from partial melting of metabasite (Harris and Massey, 1994; Le Fort, 1981; Patiño Douce and Harris, 1998; Zeng et al., 2011a, 2011b). Recently, it has been suggested that the Himalayan leucogranites represent highly fractionated granites, derived from intensive fractional crystallization (FC) with or without assimilation (Ji et al., 2020; Liu et al., 2016, 2019; Wang et al., 2020; Wu et al., 2020). In the following section, we will discuss the petrogenesis of Himalayan leucogranites based on a combination of Fe isotopic data with elemental and Sr-Nd isotope indices.
5.2.1 Fe isotope evidence and a simple fractional crystallization (FC) model

In an intensive fractional crystallization origin model for Himalayan leucogranites, there are two scenarios: (1) the two-mica leucogranites represent the cumulate rocks after extracting derivative liquids which crystallized as tourmaline and garnet leucogranites (Wu et al., 2020); (2) the two-mica, tourmaline and garnet leucogranites represent a magma differentiation series (Ji et al., 2020; Liu et al., 2020; Scaillet et al., 1990; Zeng et al., 2019). Given the lighter Fe isotopic compositions at any given index of differentiation, e.g., SiO$_2$ and FeOt (Fig. 4), garnet leucogranites, could not be controlled by the same mechanism as two-mica and tourmaline leucogranites and thus will be discussed separately.

Here we consider the possible effect of fractional crystallization for two-mica and tourmaline leucogranites. For the Rayleigh fractionation model (equation 1) and mass balance (equation 2),

\[
\delta^{56}\text{Fe}_{\text{melt}} = \delta^{56}\text{Fe}_{\text{melt0}} + \Delta^{56}\text{Fe}_{\text{crystal-melt}} \times \ln(f_{\text{Fe}})
\] (1)

\[
\delta^{56}\text{Fe}_{\text{melt0}} = f_{\text{Fe}} \times \delta^{56}\text{Fe}_{\text{melt}} + (1-f_{\text{Fe}}) \times \delta^{56}\text{Fe}_{\text{average cumulate}}
\] (2)

we can obtain:

\[
\Delta^{56}\text{Fe}_{\text{crystal-melt}} = \frac{(f_{\text{Fe}}-1)}{\ln(f_{\text{Fe}})} \times (\delta^{56}\text{Fe}_{\text{average cumulate}} - \delta^{56}\text{Fe}_{\text{melt}})
\] (3)

where $f_{\text{Fe}}$ is the residual Fe fraction in the crystallizing melt and ranges from 1.0 to 0. For the first scenario, the isotopic difference ~0 between the average cumulate ($\delta^{56}\text{Fe}_{\text{two-mica leucogranite}} = 0.17 \pm 0.06\%$, 2SD) and residual melts ($\delta^{56}\text{Fe}_{\text{tourmaline leucogranite}} = 0.18 \pm 0.05\%$, 2SD) should be the maximum of $\Delta^{56}\text{Fe}_{\text{crystal-melt}}$ (note that $\frac{(f_{\text{Fe}}-1)}{\ln(f_{\text{Fe}})}$ is always <1 and << 1 for extremely intensive fractional crystallization with $f_{\text{Fe}} \rightarrow 0$). However, this estimated $\Delta^{56}\text{Fe}_{\text{crystal-melt}}$ is inconsistent with the
mineral assemblage (dominated by mica, feldspar and quartz; Table S1) of the assumed cumulates (i.e., two-mica leucogranites), which yields a $\Delta^{56}\text{Fe}_{\text{crystal-melt}}$ between -0.05‰ to -0.10‰ (Fig. 7b, see Table 1 for partition coefficients and fractionation factors which refer to discussions in section 5.1). This scenario is therefore not applicable to the Himalayan leucogranites studied here. This view is also supported by the observation that some of the two-mica leucogranites show more evolved chemical compositions than the tourmaline leucogranites, e.g., higher SiO$_2$ and lower FeOt.

If the two-mica and tourmaline leucogranites represent a cogenetic differentiation magma series, $\Delta^{56}\text{Fe}_{\text{crystal-melt}}$, required by a simple fractional crystallization model to predict the whole rock $\delta^{56}\text{Fe}$, can be constrained by substituting $f_{\text{Fe}} = F_{\text{melt}} \times C_{\text{melt}} / C_{\text{melt0}}$ into the Rayleigh fractionation equation (1), where $F_{\text{melt}}$ is the mass fraction of residual melt. Given the difficulty of constraining $F_{\text{melt}}$ for evolved granitic magmas, here we relate $\delta^{56}\text{Fe}_{\text{melt}}$ to $C_{\text{melt}} / C_{\text{melt0}}$ through:

$$C_{\text{melt}} / C_{\text{melt0}} = F_{\text{melt}}^{(D-1)}$$

where $D$ is the bulk partition coefficient of the crystallizing assemblage. Then we obtain:

$$\delta^{56}\text{Fe}_{\text{melt}} = \delta^{56}\text{Fe}_{\text{melt0}} + \Delta^{56}\text{Fe}_{\text{crystal-melt}} \times \frac{D}{D-1} \times \ln\left(\frac{C_{\text{melt}}}{C_{\text{melt0}}}\right) (D \neq 1)$$

Note that in the case $D = 1$, $\delta^{56}\text{Fe}_{\text{melt}}$ will change with no variation in $C_{\text{melt}}$ (i.e., constant as $C_{\text{melt0}}$).

A linear regression between $\delta^{56}\text{Fe}_{\text{melt}}$ and $\ln\left(\frac{C_{\text{melt}}}{C_{\text{melt0}}}\right)$ indicates a slope (i.e., $\Delta^{56}\text{Fe}_{\text{crystal-melt}} \times \frac{D}{D-1}$) of -0.02 (Fig. 7a). As FeOt of two-mica and tourmaline leucogranites decreases with increasing SiO$_2$, $D$ should be higher than unity, which suggests a $\Delta^{56}\text{Fe}_{\text{crystal-melt}}$ between 0 and -0.02‰ (Fig. 7b).
Again, this estimate cannot be easily explained by the mineral assemblages observed in our samples (see discussions above).

It is possible that the crystallizing assemblage during magma differentiation differed from the assemblage observed in the granites. Hereafter we further test the possibility of intensive fractional crystallization considering varying crystallizing mineral assemblages. Potential major liquidus phases include plagioclase, K-feldspar, quartz, biotite, muscovite, tourmaline and garnet (Huang et al., 2017; Inger and Harris, 1993; Liu et al., 2019; Scaillet et al., 1990, 1995; Wang et al., 2020). Quartz is free of Fe and thus not considered here. Biotite is considered as the only Fe-rich mineral to provide the minimum estimate of potential isotope fractionation, for the reasons below: i) the fractionation factors of other Fe-rich minerals (i.e., muscovite, tourmaline and garnet) are either similar to or greater than $\Delta^{56}$Fe$_{\text{biotite-melt}}$ (Sossi and O’Neill, 2017; Ye et al., 2020 and this study); and ii) magnetite which is isotopically heavier (Heimann et al., 2008; Sossi and O’Neill, 2017; Telus et al., 2012; Wu et al., 2017b; Ye et al., 2020), is absent based on petrological observations and experimental results (Scaillet et al., 1995; Wu et al., 2020). Magnetite has neither been observed in thin-sections of our samples, nor has been separated by magnet from whole rock powders. Moreover, significant crystallization of magnetite will result in a decrease in Fe/Mn along with $\delta^{56}$Fe (Sossi et al., 2012), which is inconsistent with the observation (Fig. 9b). Fractional crystallization from an initial melt represented by sample LZH1130 with the maximum FeOt in this study is illustrated in Fig.7c and Fig.7d, given a crystallizing assemblage of biotite, plagioclase and K-feldspar with variable proportions and parameters listed in Table 1. Rb/Sr of the differentiating melt is dominantly controlled by mica and feldspars and thus also considered here (Harris and Inger, 1992). The calculated curves lead to two striking inferences: i) the evolved melt tends to have $\delta^{56}$Fe higher than Himalayan leucogranite at a given FeOt, especially when F$_{\text{melt}}$
is low; ii) the limited $\delta^{56}\text{Fe}$ variation of Himalayan leucogranite indicates a $\Delta^{56}\text{Fe}_{\text{crystal-melt}} \sim 0$, which implies a crystallizing assemblage of a low biotite/(biotite + feldspars) mode ratio. Intensive crystallization of such an assemblage would produce high Rb/Sr ratios that has not been observed. Overall, intensive fractional crystallization, on its own can be ruled out as the origin of tourmaline and two-mica leucogranites.

5.2.2 Assimilation and fractional crystallization (AFC) model

Simultaneous assimilation of metasedimentary rocks with $\delta^{56}\text{Fe}$ around 0.10‰ may have counteracted Fe isotope fractionation caused by fractional crystallization. An assimilation and fractional crystallization (AFC) model has been previously proposed to explain the large variation of Sr-Nd isotopic ratios of Himalayan leucogranites (Ji et al., 2020). Ji et al. (2020) considered high Sr/Y samples with unradiogenic Sr-Nd isotope compositions as the primary melt that was generated by partial melting of amphibolite, and suggested that the other leucogranites can be produced by an AFC process. Fe isotopic systematics during AFC has been illustrated in Fig. 8 (DePaolo, 1981). As discussed in Section 5.2.1, a biotite + felspars crystallizing assemblage is adopted with partitioning and fractionation parameters from Table 1. $\delta^{56}\text{Fe}$ of the initial melt and the country rock are assumed to be 0.14‰ and 0.10‰, typical of I-type granites (Foden et al., 2015) and Himalayan metasedimentary rocks (this study) respectively. Rb and Sr contents, as well as Sr isotopic systematics, are also considered, and elemental contents and $^{87}\text{Sr}/^{86}\text{Sr}(i)$ of end-members are from Ji et al. (2020). Given the high FeOt of local country rocks (e.g., 6.81 wt.% in Ji et al., 2020 and 4.68±1.18 wt.% of metasedimentary rocks here), a higher r (i.e., the ratio of assimilation rate to fractional crystallization rate) requires more biotite in the crystallizing
assemblage to cause a decrease in FeOt, which means a larger value for $\Delta^{56}\text{Fe}_{\text{crystal-melt}}$ and a higher $\delta^{56}\text{Fe}$ of the evolved melt (Fig. 8). The $\delta^{56}\text{Fe}$-FeOt trend of Himalayan leucogranites can only be reproduced by AFC when $r$ and $F_{\text{melt}}$ are both low, e.g., $r = 0.2$ and $F_{\text{melt}} < 0.4$. In this case, the calculated melt should have a Rb/Sr substantially higher than the assumed initial melt and country rock (e.g., $> 100$) and $^{87}\text{Sr}/^{86}\text{Sr}(i)$ close to that of the country rock, which, however, are not consistent with observations on Himalayan leucogranites. Accordingly, intensive fractional crystallization with simultaneous assimilation is also unable to account for the origin of tourmaline and two-mica leucogranites.

5.2.3 The role of crustal anatexis

Although some degree of fractional crystallization cannot be completely ruled out in the origin of Himalayan tourmaline and two-mica leucogranites, as discussed above, intensive fractional crystallization is not supported by the Fe isotopic data presented in this study. $\delta^{56}\text{Fe}$ and chemical compositions of these granites are unlikely to have been significantly affected after extraction from their sources. The highly variable elemental and Sr-Nd-Hf-O isotopic compositions of Himalayan leucogranites thus can be attributed to source heterogeneity and diverse melting reactions (Guillot and Le Fort, 1995; Inger and Harris, 1993; Knesel and Davidson, 2002; Le Fort, 1975; Patiño Douce and Harris, 1998; Zeng et al., 2011a). The supposed sources may range from metabasites for high Sr/Y leucogranites with relatively unradiogenic Sr-Nd isotopic compositions (Fig. 3; Hou et al., 2012; Zeng et al., 2011a) to metasedimentary rocks for low Sr/Y two-mica and tourmaline leucogranites with relatively radiogenic Sr-Nd isotopic compositions (Gao et al., 2017; Harrison et al., 1999b; Inger and Harris, 1993; Le Fort et al., 1987).
Since metabasite and metasediments both have $\delta^{56}\text{Fe} \sim 0.10\%$ (e.g., Teng et al., 2013 and this study), $\delta^{56}\text{Fe}$ of two-mica and tourmaline leucogranites are higher than their supposed sources by $0.07 \pm 0.06 \%$ (2SD, N = 27). This value is consistent with isotope fractionation that was observed in migmatites from Black Hill and the Dabie orogen, i.e., $\Delta^{56}\text{Fe}_{\text{leucosome-melanosome}}$ from 0.04% to 0.20% (Telus et al., 2012; Xu et al., 2017), and most likely reflects isotope fractionation during crustal anatexis.

### 5.2.4 Origin of low $\delta^{56}\text{Fe}$ garnet leucogranites

Given all major Fe-rich minerals in the magmatic system, e.g., biotite, muscovite, tourmaline and garnet, have $\Delta^{56}\text{Fe}_{\text{crystal-melt}} < 0$, the low $\delta^{56}\text{Fe}$ of garnet leucogranites cannot be explained by isotope fractionation during either fractional crystallization or crustal anatexis. Garnet in this type of leucogranite show euhedral shape and few inclusions with higher Mn and lower Ca contents (spessartine = 21 - 29%, Grossular <1.92%; Table S7) than examples of xenocrystic garnet (spessartine < 5%; Harris et al., 1992) or peritectic garnet (Grossular = 11 – 41%; King et al., 2011), thus indicating a probable magmatic origin (Gao et al., 2012; Zeng et al., 2019). Accumulation of this mineral in garnet leucogranites is indicated by their low (Gd/Yb)$_N$ (0.29-0.77), Fe/Mn (1.80-8.26, in mole) (Fig. 9), and HREE contents higher than the other leucogranites (Fig. S1). The low $\delta^{56}\text{Fe}$ of garnet leucogranites thus can be explained by accumulation of magmatic garnet that hosts the lightest iron isotopes of the rock-forming minerals (Figs.5a and Fig.7b). A simple binary mixing calculation suggests that addition of only 2.2 wt.% garnet in equilibrium with a low FeOt melt, represented by sample DL1703, can model the $\delta^{56}\text{Fe}$ of garnet leucogranites down to -0.04%, which is consistent with the garnet content estimation by
microscope observation. Nevertheless, the mechanism for garnet crystallization remains largely unknown, possibly due to the special pristine melt composition of garnet leucogranites.

6. Conclusion

In order to elucidate the role of partial melting and fractional crystallization in generating Himalayan leucogranites, we report Fe isotope data of 30 Himalayan leucogranites and 9 metasedimentary rocks, supplemented with mineral-pair measurement as well as geochemical and Sr-Nd isotopes data. Local metasedimentary rocks yield δ⁵⁶Fe ~0.10‰, typical of global clastic sediments. Garnet leucogranites are isotopically lighter with δ⁵⁶Fe ranging from -0.04‰ to 0.06‰, possibly due to garnet accumulation. The tourmaline and two-mica leucogranites show roughly homogeneous δ⁵⁶Fe from 0.13‰ to 0.24‰ without resolvable change with variable SiO₂ (70.80 – 78.36‰), MgO (0.04 – 0.94‰), FeOt (0.13 – 2.09‰) Mg# (18.41 – 56.53) and Eu*/Eu (0.21 – 1.31). Fractional crystallization and AFC modelling suggests that they could have not experienced intensive fractional crystallization. The higher δ⁵⁶Fe relative to the supposed sources (by ~0.07‰), probably reflects isotope fractionation during crustal anatexis. This study therefore confirms that the geochemical and isotopic characteristics of Himalayan leucogranite can be used to reflect the P-T-X_H₂O conditions of partial melting, as indicators of the thermal and tectonic evolution of Himalayan crust and orogen.

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Figure and table captions

**Fig. 1** Simplified geological map of the Himalayan orogen (modified after Xu et al., 2013). Leucogranite plutons measured in this study are labelled. JS = Jinsha suture zones; BNS = Bangong Nujiang suture zone; SSZ = Shyok suture zone; ITSZ = Indus-Tsangpo suture zone; STDS = Southern Tibetan Detachment System; MCT = Main Central Thrust; MBT = Main Boundary Thrust; MFT = Main Frontal Thrust.

**Fig. 2** Diagrams of Sr/Y vs. Y (a), A/NK vs. A/CNK (b), SiO₂ vs. FeOt (c), Mg# vs. FeOt (d), Eu/Eu* vs. FeOt (e) and Fe₃⁺/ΣFe vs. FeOt (f) for Himalayan leucogranites. A/NK = Al₂O₃/(Na₂O + K₂O) in mole, A/CNK = Al₂O₃/(CaO + Na₂O + K₂O) in mole. The literature data of Himalayan leucogranites are from: Aikman et al. (2012), Aoya et al. (2005), Carosi et al. (2013), Castelli and Lombardo (1988), Gao et al. (2016a, 2017), Gao and Zeng (2014), Gou et al. (2016), Guo and Wilson (2012), Harrison et al. (1999b), Harrison and Wielicki (2016), Hopkinson (2016), Hou et al. (2012), Hu et al. (2017), Huang et al. (2017), Inger and Harris (1993), King et al. (2011), Lin et al. (2020), Liu et al. (2014, 2016), Scaillet et al (1990), Searle et al. (1997), Shi et al. (2017), Visonà and Lombardo (2002), Wang et al. (2020), Xie et al. (2019), Yang et al. (2019), Zeng et al. (2009, 2011a, 2015, 2019), Zhang et al. (2004, 2005) and Zheng et al. (2016).

Note that aplite, pegmatite and hydrothermal altered samples are not shown. Abbreviations (also for the following figures) are: TL = tourmaline leucogranite; GL = garnet leucogranite; TML = two-mica leucogranite.
**Fig. 3** (a) Sr-Nd isotopic compositions of the investigated leucogranites. Himalayan leucogranites, metabasite from THS and metasedimentary rocks previously reported from GHS and LHS are plotted for a comparison. Metasedimentary rocks in GHS and LHS and metabasite in THS are from Ahmad et al. (2000), Deniel et al. (1987), Inger and Harris (1993), Liu et al. (2014), Richards et al. (2005) and Zeng et al. (2009, 2011a). Literature data for leucogranites are from Deniel et al. (1987), Gao et al. (2015, 2017), Gao and Zeng (2014), Guo and Wilson, (2012), Harrison et al. (1999a), Hou et al. (2012), Hu et al. (2017), Huang et al. (2017), Inger and Harris (1993), Ji et al. (2020), King et al. (2011), Lin et al. (2020), Liu et al. (2014), Zeng et al. (2009, 2011a, 2019), Zhang et al. (2004) and Zheng et al. (2016). The endmembers (high Sr/Y granite dike and Himalayan paragneiss) of mixing line are from Ji et al. (2020), whose compositions are listed in Table 1.

**Fig. 4** Diagrams of $\delta^{56}$Fe vs. SiO$_2$ (a), FeOt (b), Zr/Hf (c), Fe$^{3+}$/ΣFe (d), Mg$^+$ (e) and Eu/Eu* (f). Global igneous rock data are from: Dauphas et al. (2009a), Du et al. (2017), Foden et al. (2018), Foden et al. (2015), Gleeson et al. (2020), He et al. (2017, 2019), Heimann et al. (2008), Konter et al. (2016), Li et al. (2019), Poitrasson and Freydier. (2005), Schuessler et al. (2009), Sossi et al. (2012), Telus et al. (2012), Teng et al. (2008), Williams et al. (2018), Wu et al. (2017b), Xia et al. (2017) and Zambardi et al. (2014).

**Fig. 5** $\delta^{56}$Fe of minerals separated from representative garnet, tourmaline and two-mica leucogranites.

**Fig. 6.** $\delta^{56}$Fe variations with potential indices of fluid exsolution, e.g., Y/Ho (a) and TE$_{1,3}$ (b). TE$_{1,3}$ = $((\text{Ce}_N \times \text{Pr}_N \times \text{Tb} \times \text{Dy})/(\text{La}_N \times \text{Nd}_N \times \text{Gd}_N \times \text{Ho}_N))^{0.25}$ (Irber, 1999). The chondrite normalized data are from Boynton (1984).

**Fig. 7.** (a) A linear fit between the $\delta^{56}$Fe and $\ln\left(\frac{\text{FeOt}}{\text{FeOt}(i)}\right)$ for two-mica and tourmaline leucogranites. (b) Diagram of estimated $\Delta^{56}$Fe$_{\text{mineral-melt}}$ vs. D(Fe)$_{\text{mineral-melt}}$. The possible Fe isotope fractionation factor between a crystallizing assemblage of quartz + plagioclase + K-feldspar + muscovite + biotite and the melt is given as the grey field. Isotopic effect of fractional crystallization is illustrated along with changes in
FeOt and Rb/Sr in (c) and (d) respectively. Green-grey contour lines contour the evolved compositions with given $F_{\text{melt}}$.

**Fig. 8** Illustration of an AFC process with $r = 0.2$ and $0.8$. The details of primitive melt and country rock were listed in Table 1 and discussed in the text. Same as in Fig. 7, Green-grey contour lines contour the evolved compositions with given $F_{\text{melt}}$.

**Fig. 9.** $\delta^{56}\text{Fe}$ vs. $(\text{Gd/Yb})_N$ (a) and $\delta^{56}\text{Fe}$ vs. Fe/Mn (in mole) (b) diagrams where the garnet leucogranites have the relatively lower $\delta^{56}\text{Fe}$, $(\text{Gd/Yb})_N$ and Fe/Mn values.

**Table 1.** The partition coefficients and isotope fractionation factors for minerals, and the initial melt and country rock endmembers for AFC modelling.

*Note.* Fe partition coefficients of these minerals in the literature were compiled from $^a$Icenhowe and London (1995), $^b$Ewart and Griffin (1994), $^c$Sisson and Bacon (1992), $^d$Benard et al. (1985) and $^e$Wu et al. (2017b). $\Delta^{56}\text{Fe}_{\text{biotite-melt}}$ was suggested to be 0.10‰ according to the fractionation in Black Hills migmatites where Fe in the melanosome is dominated by biotite (Telus et al., 2012). $\Delta^{56}\text{Fe}_{\text{tourmaline-melt}}, \Delta^{56}\text{Fe}_{\text{garnet-melt}}$ and $\Delta^{56}\text{Fe}_{\text{tourmaline-melt}}$ were deduced based on the $\Delta^{56}\text{Fe}_{\text{tourmaline-biotite}}$ in section 5.1 and the $\Delta^{56}\text{Fe}_{\text{garnet-biotite}}$ and $\Delta^{56}\text{Fe}_{\text{magetite-biotite}}$ from Ye et al. (2020). $\Delta^{56}\text{Fe}_{\text{muscovite-melt}}$ was assumed to be the same with $\Delta^{56}\text{Fe}_{\text{biotite-melt}}$. $\Delta^{56}\text{Fe}_{\text{plagioclase-melt}}$ and $\Delta^{56}\text{Fe}_{\text{K-feldspar-melt}}$ were calculated by the relationship of $\Delta^{56}\text{Fe}_{\text{plagioclase-biotite}}$ and $\Delta^{56}\text{Fe}_{\text{K-feldspar-biotite}}$ assuming Ab% and Or% of plagioclase and K-feldspar is 90% (Liu et al., 2019; Wu et al., 2017b). The geochemical composition of endmembers for AFC modelling were from Ji et al. (2020) and their Sr isotopes were corrected at 20 Ma.

**References**


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on with K/Rb, Eu/Eu*, Sr/Eu, Y/Ho, and Zr/Hf of Malayan Metamorphism and Its Tectonic Implications, auphas, N. 2 26 Accepted Article
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References From the Supporting Information


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Metasedimentary rocks from GHS and LHS

$\varepsilon_{\text{Nd}}(t)$

$\frac{87\text{Sr}}{86\text{Sr}}(i)$

Mixing line

TL samples
GL samples
High Sr/Y TML samples
Low Sr/Y samples
Literature data

Metabasite from THS

Metasedimentary rocks from GHS and LHS

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(a) 

$\delta^{56}Fe$ (‰) vs. $(\text{Gd/Yb})_n$

- TL samples
- GL samples
- High Sr/Y TML samples
- Low Sr/Y TML samples
- Metasedimentary samples

(b) 

$\delta^{56}Fe$ (‰) vs. Fe/Mn (in mole)

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Table 1. The partition coefficients and isotope fractionation factors for minerals, and the initial melt and country rock endmembers for AFC modelling.

<table>
<thead>
<tr>
<th></th>
<th>Biotite</th>
<th>Muscovite</th>
<th>Magnetite</th>
<th>Garnet</th>
<th>Tourmaline</th>
<th>Plagioclase</th>
<th>K-feldspar</th>
<th>Initial melt</th>
<th>Country rock</th>
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<td>5.17-6.01$^{a}$</td>
<td>47.4-202$^{b}$</td>
<td>48$^{c}$</td>
<td>16-30$^{d}$</td>
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<td>0.013-0.121$^{e}$</td>
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<td>$D_{Fe}$ used in this study</td>
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<td>$D_{Fe}$</td>
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<td>-0.20</td>
<td>-0.18</td>
<td>0.77</td>
<td>0.89</td>
<td>$\delta^{56}Fe$ (%)</td>
<td>0.14</td>
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</tbody>
</table>

Note. Fe partition coefficients of these minerals in the literature were compiled from $^{a}$Icenhower and London (1995), $^{b}$Ewart and Griffin (1994), $^{c}$Sisson and Bacon (1992), $^{d}$Benard et al. (1985) and $^{e}$Wu et al. (2017b). $\Delta^{56}Fe_{biotite-melt}$ was suggested to be 0.10‰ according to the fractionation in Black Hills migmatites where Fe in the melanosome is dominated by biotite (Telus et al., 2012). $\Delta^{56}Fe_{tourmaline-melt}$, $\Delta^{56}Fe_{garnet-melt}$ and $\Delta^{56}Fe_{tourmaline-melt}$ were deduced based on the $\Delta^{56}Fe_{tourmaline-biotite}$ in section 5.1 and the $\Delta^{56}Fe_{garnet-biotite}$ and $\Delta^{56}Fe_{magnetite-biotite}$ from Ye et al. (2020). $\Delta^{56}Fe_{muscovite-melt}$ was assumed to be the same with $\Delta^{56}Fe_{biotite-melt}$. $\Delta^{56}Fe_{plagioclase-melt}$ and $\Delta^{56}Fe_{K-feldspar-melt}$ were calculated by the relationship of $\Delta^{56}Fe_{plagioclase-biotite}$ and $\Delta^{56}Fe_{K-feldspar-biotite}$ assuming Ab% and Or% of plagioclase and K-feldspar is 90% (Liu et al., 2019; Wu et al., 2017b). The geochemical composition of endmembers for AFC modelling were from Ji et al. (2020) and their Sr isotopes were corrected at 20 Ma.