Research Paper

From source to emplacement: The origin of leucogranites from the Sikkim-Darjeeling Himalayas, India

Tanya Srivastava a,⇑, Nigel Harris b, Catherine Mottram c, Kumar Batuk Joshi d, Nishchal Wanjaria a

aDepartment of Geology, Sikkim University, Gangtok, India
bSchool of Environment, Earth & Ecosystem Sciences, Open University, Milton, Keynes, UK
cSchool of the Environment, Geography and Geosciences, University of Portsmouth, UK
dNational Centre for Earth Science Studies, Thiruvananthapuram, Kerala, India

1. Introduction

The Himalayan orogenic belt is a type example of intercontinental collision that has experienced crustal thickening, multi-phase metamorphism, deformation, and anatexis, from the start of the Indo-Eurasia continental collision at ca. 65–50 Ma, to recent < 10 Ma events (Najman et al., 2016, 2010; Green et al., 2008). The Cenozoic Himalayan leucogranites, which have been emplaced along the entire strike of the orogen, preserve evidence of crustal melting from the Eocene (ca. 49 Ma) to Pleistocene (ca. 1 Ma) (Cao et al., 2022), with a peak in activity during the Early Miocene and a broad eastward-younging trend along the orogen (Mottram et al., 2019 and references therein). Himalayan leucogranites therefore provide a window for exploring the temporal and spatial tectonic evolution of the orogen (Inger and Harris, 1993; Tartèse and Boulvais, 2010; Hopkinson et al., 2017; Lin et al., 2020; Gao et al., 2021b).

Numerous studies of Himalayan leucogranites have found that the compositional differences in leucogranites from the Himalayas are the result of variable degrees of partial melting of different source rocks (Scaillet et al., 1990; Visonà and Lombardo, 2002; Zhang et al., 2004; Yang et al., 2015; Nabelek, 2019). Trace element trends observed in many of the High Himalaya leucogranites can be theoretically modelled from either partial melting of a metasedimentary source or from fractional crystallisation. For example Rb-Sr-Ba systematics in leucogranite from the central Himalaya can be reproduced by the fractional crystallisation of alkali feldspar (Inger and Harris, 1993). However, feldspar fractionation in acidic magmas is impeded by the lack of density contrast between the solid phase and the melt and by the high viscosity of granitic liquids. Moreover petrographic evidence for many leucogranites suggests that alkali feldspar is a late phase to crystallize, thus arguing against it being a cumulative phase (Inger and Harris, 1993). It has...
also been argued that extreme fractional crystallization has resulted in rare-metal enrichments in some of the Tethyan leucogranites from the northern Himalaya of southern Tibet (Liu et al., 2019; Wu et al., 2020). Whilst this interpretation may be valid for such mineralized granites, since their parental magmas are characterized by high fluid activities lowering the melt viscosity (Wu et al., 2017), this is unlikely to be the case for the many unmineralized leucogranite bodies, such as are described in this study. Moreover, recent Fe-isotope studies of similar Himalayan leucogranites preclude significant fractional crystallization during their formation (Shi et al., 2021).

Potential heat sources that have been suggested for such partial melting include: (i) internal heating by radioactive decay of isotopes, (ii) shear heating (England et al., 1992; Searle and Szulc, 2005; Nabelek et al., 2010; Hopkinson et al., 2017), (iii) decompression during extension of the crust (Harris and Massey, 1994), or (iv) mantle advection (Liu et al., 2014; Zheng et al., 2016; Gao et al., 2021a). Trace element modeling of Himalayan crustal melts has identified pelitic sediments as their source (Harris et al., 1995) although other studies have cited metagreywacke (Guillot and Le Fort, 1995), amphibolite (Zeng et al., 2011), felsic and pelitic high pressure (HP) granulites (Zhang et al., 2017) as contributory sources for crustal melting.

Sr-Nd isotopes, zircon U-Pb ages and εNd(t) isotopic signatures have been used to suggest that the metasedimentary rocks and granitic gneiss of the High Himalayan Sequence (HHS) provide the principal source lithologies for the generation of leucogranite magma (Gou et al., 2016; Xie et al., 2018) In contrast, other studies have concluded that leucogranite melts originated from the melting of both the HHS and the Lesser Himalayan Sequence (LHS) rocks (Guo and Wilson, 2012; Hopkinson et al., 2020). O-Hf isotopic signatures from zircon have been interpreted to preclude a mantle source for melting in the leucogranites of the eastern Himalayas (Hopkinson et al., 2017).

U-(Th)-Pb ages from the zircons and monazites from leucogranites across the Himalayan orogen have been reported by many researchers ranging from 31 Ma to 9 Ma (Harrison et al., 1995, 1997; Searle and Godin, 2003; Cottle et al., 2009, 2015; Kellett et al., 2010, 2013; Guo and Wilson, 2012; Lederer et al., 2013; Greenwood et al., 2016; Hopkinson, 2016). Despite this wealth of research focusing on Himalayan leucogranites, only one previous study has analysed deformed leucogranites from Sikkim (Catalos et al., 2004). Hence the Sikkim Himalayan leucogranite bodies remain an understudied gap in the record of Himalayan melt generation. Moreover the abundance of previous geochronological constraints from the surrounding Tethyan Himalayan Sequence (THS), HHS and LHS rocks (Catalos et al., 2004; Rubatto et al., 2013; Anczkiewicz et al., 2014; Kellett et al., 2014; Mottram et al., 2014a,b) provided the ideal context for new geological investigations into the source and emplacement mechanisms of Himalayan leucogranites. This paper aims to provide a new petrological, geochemical and geochronological dataset of leucogranites from the Sikkim Himalayas.

In this contribution, we provide new whole-rock geochemical, U-Pb zircon and monazite isotopic data from Higher Himalayan leucogranite samples that provide constraints for the timing and conditions of melt emplacement. U-Pb data from an HHS gneiss from the Sikkim-Darjeeling Himalayas provides constraints on a potential source of leucogranite magma. These results together with previous studies, identify the geochemistry, source and melting mechanism and the tectonic environment responsible for the formation of leucogranite bodies in the Sikkim-Darjeeling region of the eastern Himalayas.

2. Geological background

The Himalayan orogen (Fig. 1) is demarcated by the Indus Tsangpo Suture Zone (ITSZ) and the Main Front Thrust (MFT), in the north and south respectively (Yin, 2006). The orogen (Fig. 1) is divided into four geologic domains which are sub-parallel and laterally continuous tectono-stratigraphic units and consist of the Tethyan Himalayan Sequence (THS), Higher Himalayan sequence (HHS), also known as Greater Himalayan Sequence, Lesser Himalayan sequence (LHS) and Neogene Siwalik Formation, from north to south (Godin et al., 2001; Yin, 2006). The THS (Fig. 1), bounded by the ITSZ in the north, and the South Tibetan Detachment System (STD), in the south, consists of Proterozoic to Eocene low-grade metamorphic to unmetamorphosed siliciclastic, carbonate sediments and volcanic rocks. The HHS (Fig. 1), bounded by the STD in the north and the Main Central Thrust zone (MCT) in the south, consists of Precambrian to Paleozoic high-grade (upper amphibolite facies) metamorphic rocks. The LHS (Fig. 1), bounded by the MCT in the north and the Main Boundary Thrust (MBT) in the south, is composed of Proterozoic metasedimentary sequence, typically greenschist to amphibolite facies (Richards et al., 2005; Kohn, 2014). The Neogene Siwalik formation (Fig. 1), bounded by the MBT and MFT in the north and south respectively, is composed of Cenozoic sediments. Three northward dipping faults, the MCT, MBT and MFT, are construed to merge at depth into the low angle
Main Himalayan Thrust (MHT) (Schelling and Arita, 1991; Nelson et al., 1996).

The leucogranites of the Himalayas include two-mica leucogranites, tourmaline leucogranites (Guillot and Le Fort, 1995), garnet leucogranites (Hopkinson et al., 2017) and cordierite leucogranites (Gou et al., 2019). Their emplacement define two sub-parallel belts (i) the northern belt known as the Tethyan Himalayan Leucogranites (THL), that intrude the THS and (ii) the southern belt referred to as Higher Himalayan Leucogranites (HHL) which intrudes into the high-grade metasedimentary rocks (King et al., 2011; Weinberg, 2016).

Initial geological mapping in the Sikkim Himalayas was carried out by Mallet (1875), that was later modified by (Auden, 1935; Heim and Gansser, 1939; Wager, 1939; Ghosh, 1952; Gansser, 1964). Since then, numerous studies have used structural analysis, geological mapping, geochronological, geothermobarometric and petrological study to understand the tectono-metamorphic evolution of this region (Catlos et al., 2004; Harris et al., 2004; Kellett et al., 2014; Mottram et al., 2014a, 2015a; Sorcar et al., 2014; Bhattacharyya et al., 2015; Srivastava et al., 2022). The present study has been carried out in North Sikkim (Fig. 2) and the Darjeeling Hills to the south (Fig. 2). The leucogranites in the study area intrude the country rocks of the HHS forming either as dykes or sills. A total of twenty-seven leucogranite samples were collected from the road sections in North Sikkim, near Thangu, Yumthang, Yumesamdong, and adjoining areas, for petrographic and geochemical studies. The sillimanite-bearing leucocratic gneiss was collected from Mirik, Darjeeling, for geochronological study (Fig. 2).

In the Sikkim Himalayas, protracted melting, documented by zircon and monazite crystallization, occurred in the HHS in North Sikkim from ca. 37 Ma to 14.6 Ma (Fig. 3) (Rubatto et al., 2013; Mottram et al., 2014b), between ca. 37–16 Ma in West Sikkim and ca. 17–14.3 Ma in south east Sikkim (Mottram et al., 2014b). Sm-Nd garnet ages of migmatites in the hanging wall of MCT span of ca. 23–16 Ma (Harris et al., 2004), Lu-Hf ages obtained from garnets within the HHS yield ages ca. 28–26 Ma and ca. 22 Ma, and from ca. 16–10 Ma within the MCT zone (Anczkiewicz et al., 2014). U-Th-Pb monazite ages of ca. 21–12 Ma in the structurally higher parts, and ca. 16–9 Ma in the structurally lower parts of the MCT zone, indicate progressive incorporation of material into the thrust zone (Mottram et al., 2014b). Folding of the MCT zone by the later Lesser Himalayan Duplex has led to differential exposure of the thrust, with windows into the younger history exposed northwards (Mottram et al., 2015a). The Sikkim leucogranites yielded Th-Pb ages of ca. 17 Ma and ca. 15–14 Ma (Catlos et al., 2004). The thermochronological data indicated that the STDS in Sikkim was active at ca. 24–13 Ma (Kellett et al., 2013).

### 3. Field description and petrography

Field work in North Sikkim was conducted along two transects: (i) the Chungthang-Lachen-Thangu-Gurudongmar section and (ii) the Chungthang-Lachung-Yumthang-Yumesamdong section (Fig. 2). The leucogranites in the study area intrude the country rocks of the HHS forming either as dykes or sills. A total of twenty-seven leucogranite samples were collected from the road sections in North Sikkim, near Thangu, Yumthang, Yumesamdong, and adjoining areas, for petrographic and geochemical studies. The sillimanite-bearing leucocratic gneiss was collected from Mirik, Darjeeling, for geochronological study (Fig. 2). The field and petro-
The graphic characteristics of rock types are provided in Table 1, and all leucogranite sampled present a massive, undeformed texture. The sample locations and the details of samples are given in Supplementary Data Table S1. Based on the macroscopic observations, the studied leucogranites have been divided into two-mica leucogranites (2mg) and tourmaline leucogranites (Tg). A similar classification has been assigned to HHL by some previous studies (Scaillet et al., 1990; Inger and Harris, 1993; Guillot and Le Fort, 1995; Searle et al., 1997; Visonà and Lombardo, 2002). The 2mg which intrude the HHS gneiss and migmatites (Fig. 4a), are fine to medium-grained. Quartz-tourmaline orbicules, encircled by leucocratic halos (Fig. 4b,c), and tourmaline veins (Fig. 4d) characterize some 2mg samples. The Tg also intrude the HHS gneiss and migmatites (Fig. 4e) and are coarser-grained than the 2mg. The tourmaline grains in Tg occur as disseminated specks and are of few cm (≤1 cm) in length (Fig. 4f).

The 2mg (Fig. 5a–e) are composed of quartz (ca. 35%), K-feldspar (ca. 25%), plagioclase (ca. 25%), biotite (ca. 10%), muscovite (ca. 3%), and tourmaline (if present ca. 2%) with zircon, monazite and apatite occurring as accessory phases. The K-feldspar grains are subhedral to anhedral, ca. 15 mm in length and show perthitic texture. The plagioclase grains are up to 10–12 mm in length, are euhedral to subhedral with a few grains of plagioclase showing zoning due to changes in composition (Fig. 5c). The biotite grains are subhedral to anhedral, ca. 10 mm in length and form along grain boundaries of quartz; some of the biotite grains contain zircon inclusions and pleochroic halos (Fig. 5d). Muscovite grains are up to 5–8 mm in length, subhedral to anhedral and occur mostly along the grain boundaries (Fig. 5b). The studied samples also show micrographic texture resulting from intergrowths of quartz and alkali feldspar (Fig. 5e).

The Tg (Fig. 5f–h) contains quartz (ca. 35%), K-feldspar (ca. 20%), plagioclase (ca. 25%), tourmaline (15%), and muscovite (ca. 5%) grains with minor or negligible biotite with zircon, monazite and apatite occurring as accessory phases. The quartz grains are anhedral while the K-feldspar shows perthitic texture (Fig. 5h) and range in form from subhedral to anhedral. The plagioclase grains are subhedral to euhedral and shows polysynthetic twinning. The tourmaline grains, ca. 15 mm in length, are euhedral to subhedral, show green to blue pleochroism and occur with quartz and feldspar. The muscovite grains are subhedral to anhedral and may show serrated contacts with quartz (Fig. 5g). Some of the muscovite grains also occur along grain boundaries (Fig. 5h).

The sillimanite-bearing felsic gneiss from the Darjeeling Hills contains quartz (ca. 65%), sillimanite (ca. 30%), feldspar, biotite and muscovite (ca. 5%). The sillimanite grains (ca. 1–2 mm) occur as fibrous aggregations alternating with quartz bands (Fig. 5i,j).

### 4. Analytical methods

To investigate the petrogenesis of the North Sikkim leucogranites twenty-seven leucogranites samples, including both two-mica and tourmaline leucogranites, were selected for whole-rock major
and trace element analysis. For geochronology, one two-mica leucogranite sample was dated using both zircons and monazites (ZY-LG-2) and a second sample of tourmaline leucogranite (ZY-LGT-1) was dated using only monazites. One HHS felsic gneiss sample (SK-10-8) was dated using zircon U-Pb geochronology.

4.1. Whole-rock major and trace elements

Whole-rock major and trace elements were analyzed at Bureau Veritas, Canning Vale, Australia, and at National Centre for Earth Science Studies (NCESS). Major element oxides were determined by X-ray fluorescence (XRF). The accuracy for the reference standards is better than 0.5% for SiO₂ and < 3% for other major oxides and the precision for measurements is better than 1%. Trace elements, including REE, were determined using Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS). The measurements were carried out on a New Wave EXCIMER 193 nm UV laser coupled to an Agilent 7900 ICP-MS with precision and accuracy better than 5% (Srivastava et al., 2022). The detailed analysis of whole-rock major and trace element data from the studied leucogranites are listed in Supplementary Data Text S1. The whole rock major and trace element data are listed in Supplementary Data Table S2.

4.2. Zircon U-Pb geochronology

The U-Pb zircon geochronology for sample ZY-LG-2 was carried out in CODES Analytical Laboratories at the University of Tasmania, Australia with an ASI RESOLUTION S-155 ablation device and a Coherent Compex Pro 110 Ar-F excimer laser with a wavelength of 193 nm and a pulse duration of 20 ns coupled to an Agilent 7900 quadrupole ICP-MS. Zircon was ablated using a spot size of 20–30 μm, a 5 Hz repetition rate and a fluence of ca. 2 J/cm². The detailed analytical conditions for the measurement can be seen in Thompson et al. (2018). The operating details are shown in Supplementary Data Text S2 and Table S3 and analytical data are presented in Supplementary Data Text S2, Table S4.

Zircons from the sample of sillimanite-bearing gneiss, SK-10-8, were separated and analysed at the NERC Isotope Geosciences Laboratory (NIGL) using a Nu Instruments, Nu Plasma HR multicollector inductively coupled plasma mass spectrometer (MC-ICP-MS) equipped with a multi-ion-counting array. The instrumental configuration and measurement procedures follow previous methods (Mottram et al., 2014a). The instrumental details are presented in Supplementary Data Text S3, Table S5 and analytical data are given in Supplementary Data Table S6. The quoted uncertainties include the long-term reproducibility of the secondary reference material.

4.3. Zircon REE

The trace element abundances in zircons from sample ZY-LG-2 were analyzed at CODES Analytical Laboratories at the University of Tasmania, Australia using LA-ICP-MS and were calibrated on the NIST610 glass from values of (Jochum et al., 2011), using secondary standard corrections based on the compositions of glasses BCR-2G and GSD-1G (GeoReM preferred values; https://georem.mpch-mainz.gwdg.de/). The abundances of trace elements measured in the 91500 zircons are within the range of values published on the GeoReM website. Quantification was performed using ⁹¹Zr as the internal standard element, normalizing all measured cations to the Zr site total of a stoichiometric zircon. The analysis procedure is detailed in Supplementary Data Text S2. The zircon trace element data are given in Supplementary Data Table S4.

4.4. Monazite U-Th-Pb geochronology

Monazite U-Th-Pb isotopic analysis was carried out in CODES Analytical Laboratories at the University of Tasmania, Australia with an ASI RESOLUTION S-155 ablation device and a Coherent Compex Pro 110 Ar-F excimer laser with a wavelength of 193 nm and a pulse duration of 20 ns coupled to an Agilent 7900 quadrupole ICP-MS. Monazites were analysed using a 13 μm laser spot, a 5 Hz repetition rate and a fluence of ca. 2 J/cm². The detailed analytical conditions for the measurement is given in Thompson et al. (2018). The instrumental data are shown in Supplementary Data Table S7 and the monazite analytical data are given in Supplementary Data Table S8.

4.5. Monazite REE

The trace element abundances in monazite from samples ZY-LG-2 and ZY-LGT-1 were analysed by LA-ICP-MS and calibrated on the NIST610 glass from values of (Jochum et al., 2011), using secondary standard corrections based on the compositions of glasses BCR-2G and GSD-1G (GeoReM preferred values; http://georem.mpch-mainz.gwdg.de/). Quantification was performed using ¹⁴⁰Ce as the internal standard element, normalizing all measured cations to the Ce site total of a stoichiometric monazite. The 14,971 monazite and the NIST610, BCR-2G and GSD-1G glasses were analysed in duplicate at the beginning, end and every 60 min throughout the analytical session.

The calibration of the U-Pb ages was checked on analyses of the RLG4b monazite (Rubatto et al., 2001) and the Banaeira monazite (Gonçalves et al., 2016) analysed throughout the analytical session and treated as unknowns. The detailed analytical conditions are presented in Supplementary Data Text S4 and the monazite trace element data are given in Supplementary Data Table S8.

5. Results

5.1. Whole rock major and trace element compositions

The North Sikkim 2mg have SiO₂ of 70.91–74.61 wt.% (avg = 72.71 wt.%), Al₂O₃ of 13.69–15.82 wt.% (avg = 14.80 wt.%), CaO of 0.56–1.67 wt.% (avg = 1.24 wt.%), TiO₂ of 0.12–0.29 wt.% (avg =
0.20 wt.%), \(\text{Fe}_2\text{O}_3\) of 1.23–2.42 wt.% (avg = 1.80 wt.%), MgO of 0.21–0.74 wt.% (avg = 0.45 wt.%) and \(\text{P}_2\text{O}_5\) of 0.06–0.15 wt.% (avg = 0.10 wt.%). (Supplementary Data Table S2; Fig. 6) with \(\text{K}_2\text{O}/\text{Na}_2\text{O}\) ratios ranging from 1.08 to 2.42. A/\(\text{CNK}\) ratios for 2mg range of 1.03–1.37.

As compared to 2mg, the Tg have elevated \(\text{SiO}_2\) (74.38–74.9 wt.%; avg = 74.62 wt.%), \(\text{P}_2\text{O}_5\) (0.15–0.17 wt.%; avg = 0.16 wt.%), lower concentration of \(\text{TiO}_2\) (0.06–0.09 wt.%; avg = 0.07 wt.%), \(\text{Fe}_2\text{O}_3\) (0.73–0.98 wt.%; avg = 0.85 wt.%), MgO (0.13–0.20 wt.%; avg = 0.17 wt.%), \(\text{Al}_2\text{O}_3\) (13.78–14.8 wt.%; avg = 14.18 wt.%), and CaO (0.45–1.13 wt.%; avg = 0.71 wt.%). (Supplementary Data Table S2; Fig. 6). The Tg have \(\text{K}_2\text{O}/\text{Na}_2\text{O}\) ratios of 1.13–2.31 and, A/\(\text{CNK}\) ratios ranging from 1.11–1.27.

The chondrite-normalized REE and primitive mantle normalized spider diagrams for 2mg and Tg are shown in Fig. 7. The 2mg leucogranites are enriched in LREEs ([\(\text{REE}\) = 72.57–203.52 ppm; avg = 129.79 ppm]) and depleted in HREEs ([\(\text{REE}\) = 8.42–18.2 ppm; avg = 14.27 ppm]) showing fractionated REE patterns with (\(\text{La}/\text{Yb}\))\(_\text{N}\) ratios of 7.33–37.35 (avg = 13.30). Most of the 2mg show negative Eu anomalies with \(\text{Eu}/\text{Eu}^*\) = 0.48–0.75 (avg = 0.56) with an exception of Y-LG-1 which shows slight positive Eu anomaly (\(\text{Eu}/\text{Eu}^*\) = 1.03). In comparison to 2mg, Tg have flatter REE patterns, are less fractionated with (\(\text{La}/\text{Yb}\))\(_\text{N}\) ratios ranging from 3.25 to 4.80 (avg = 3.84) and have strong negative Eu anomalies \(\text{Eu}/\text{Eu}^*\) = 0.20–0.57 (avg = 0.42). In the chondrite-normalized REE plot (Fig. 7a, c) both 2mg and Tg show enrichment of LREEs and depletion in HREEs, with pronounced negative Eu anomalies due to retention of feldspar in the source. The lower LREE enrichment of the Tg may be related to residual accessories (allanite, monazite) in the source region. In the multi-element spider diagram (Fig. 7b, d) the North Sikkim leucogranites are enriched in Rb, K, Th, U, Pb and depleted in Ba, Nb, Sr, Zr, and Ti. The enrichment of Rb, Th, U, K, Pb and depletion of Nb are consistent with a crustal source for the studied leucogranites.

In the primitive mantle normalized spider diagrams (Fig. 7b, d) all the studied leucogranites are enriched in large ion lithophile elements (LILEs: e.g., Rb, U, K, and Pb) and depleted in high field strength elements (HFSE: e.g., Nb, Ta, and Ti). The 2mg exhibits U
Fig. 5. Photomicrographs from North Sikkim leucogranites. (a,b) Mineral assemblages of two mica leucogranite; (c) zoning in plagioclase representing compositional change; (d) zircon inclusion in biotite; (e) Micrographic texture representing intergrowth of quartz and alkali feldspar; (f,g) tourmaline leucogranites; (h) perthitic texture representing subsolidus exsolution; (i,j) sillimanite-bearing gneiss. Mineral Abbreviations after Whitney and Evans (2010) Bt: biotite; Kfs: K-feldspar; Ms: muscovite; Pl: plagioclase; Qz: quartz; Tur: tourmaline; Sil: sillimanite; Zrn: Zircon.
contents of 4.4–28.7 ppm (avg = 11.88 ppm), Th contents of 8.89–25 ppm (avg = 16.83 ppm), Hf contents of 2.29–4.98 ppm (avg = 3.53 ppm) and Zr contents of 60.5–158 ppm (107.73 ppm). In comparison to 2mg, the Tg have a limited range of U (14.9–18.9 ppm; avg = 16.4 ppm), Th (4.38–7.6 ppm; avg = 5.71 ppm), Hf (91.26–1.68 ppm; avg = 1.50 ppm) and Zr (25–39.5 ppm; avg = 33.67 ppm). The studied 2mg show Rb concentration ranging from 176 to 327 ppm (avg = 244 ppm), with wide range of Ba (295–1190 ppm; avg = 598 ppm), Sr (86–277 ppm; avg = 182 ppm) and Zr/Hf (25.64–33.82; avg = 30.12). The Tg have slightly elevated Rb contents (258–301 ppm; avg. = 280 ppm) as compared to the 2mg and show lower Sr (23.7–79.4 ppm; avg = 56.1 ppm), Ba (65–342 ppm; avg = 200 ppm) and Zr/Hf (19.84–23.51; avg = 22.20) ratios.

5.2. Zircon U-Pb isotopes and trace elements in leucogranites

The zircons from the 2mg (ZY-LG-2) are euhedral to subhedral and range in size between 50 μm and 150 μm (Fig. 8a). Most of the zircon grains exhibit oscillatory zoning in Cathodoluminescence (CL) images suggesting a magmatic origin (Corfu, 2003). The analysed spots have U contents ranging from 141 ppm to 8268 ppm, Th contents from 19.5 ppm to 1274 ppm, Hf contents of 9075–19,762 ppm and Th/U ratios varying of 0.01–1.34. The analysed zircons show oscillatory zoning with rims containing high U and Th contents, but low Th/U ratios which has been suggested to be indicative of anatexis (Hoskin and Schaltegger, 2003; Liu et al., 2014). A total of 60 analyses were performed on 60 zircon grains which were dominantly discordant and range between ca. 2400 Ma and ca. 14 Ma. Eleven grains yield U/Pb Concordia ages of 1718 ± 16 Ma (n = 2, MSWD = 0.58); 953 ± 28 Ma (n = 1); 775 ± 17 Ma (n = 2, MSWD = 3.2); 444 ± 7 Ma (n = 2, MSWD = 1.4); 18.9 ± 0.6 Ma (n = 1) and of 15.29 ± 0.20 Ma (MSWD = 1.3; n = 3) (Fig. 8b–d). The Kernel density estimate (KDE) shows younger Miocene ages ranging of ca. 19–15 Ma; the cut-off for the concordance was > 95% (Fig. 8 e).

A total of 60 spots in the studied zircons were measured for trace element concentrations (including REEs). The chondrite-normalized REE patterns (Fig. 8f) of zircons from 2mg have typical heavy REE enriched patterns with marked negative Eu (Eu/Eu* = 0.01–0.43) and positive Ce (Ce/Ce* of 1.22–142) anomalies. The average melt temperature calculated for zircon saturation are 754 °C (2mg) and 675 °C (Tg) using the calibration of Watson and Harrison (1983).

Fig. 6. Harker variation plots of major elements from North Sikkim leucogranites.
5.3. Zircon U-Pb isotopes in sillimanite-bearing gneiss

The zircons from the sillimanite-bearing gneiss sample SK-10-8 are euhedral to subhedral and range in length from 80 μm to 150 μm long (Fig. 9g). Zircons display a range of cathodoluminescence (CL) zoning textures (Fig. 9g) which have been categorized into 6 main categories: (i) large (>100 μm) euhedral dark grains that lack CL zoning; (ii–iv) euhedral to subhedral grains with (ii) metamict cores, (iii) concentrically zoned mantles and (iv) bright rims; (v) small (<100 μm) concentrically zoned; (vi) elongate grains. A total of 129 U-Pb analyses were performed on 70 zircon grains which span a range of ages from ca. 2640 Ma to ca. 14 Ma (Fig. 9a–e). Large (>100 μm) dark CL grains typically yield discordant ages but yield some concordant analyses ranging to the oldest 207Pb/206Pb spot age of 2643 ± 191 Ma (Fig. 9c). Metamict cores typically yield discordant ages that lie on a tie line between Miocene ages to a concordant 206Pb/238U spot age of 503 ± 14 Ma (Fig. 9d). Zircon mantles yield ages along the ca. 500 Ma – Miocene tie line to a youngest age of ca. 30 Ma (Fig. 9d–e). Bright CL rims yield ages that range between 206Pb/238U spot ages of 33.4 ± 0.5 Ma to 15.15 ± 0.5 Ma (Fig. 9b). Elongate and concentrically zoned zircons yield ages of ca. 15–14 Ma and yield an individual U-Pb spot age of 14.2 ± 0.4 Ma (Fig. 9b). A KDE shows the range of young concordant Miocene-Oligocene ages that span at ca. 33–14 Ma (Fig. 9f).

5.4. Monazite U-Th-Pb isotopes and trace elements in leucogranites

A total of 70 (35 each for 2mg and Tg) spots were analysed in monazites for U-Th-Pb isotopes and trace elements. The monazite grains in both 2mg and Tg are subhedral to anhedral and range of 80–200 μm in size (S5). The Th-U plots of monazite data for sample ZY-LGT-1 with variation in Eu* and sample ZY-LG-2 with Y (ppm) concentration is represented in (Fig. 11a, b).

In 2mg sample (ZY-LG-2), the 35 monazite grains yielded 206Pb/238U intercept ages ranging between two end members of 15.44 ± 0.33 (MSWD = 0.5, n = 4) and 14.24 ± 0.23 (MSWD =1.6, n =14) (Fig. 10 b). The chondrite-normalized REE patterns (Fig. 12 b) of monazites for 2mg are enriched in LREE relative to HREE with marked negative Eu anomaly (Eu/Eu* of 0.014–0.079), with little variation between the different aged parts of the grains. The analyzed spots have high Th contents of 41,760.6 ppm to 78,715.3 ppm and low U contents of 2440.17 ppm to 17,780.02 ppm.
In Tg sample (ZY-LGT-1) the 35 monazite grains yield
\(^{206}\text{Pb}/^{238}\text{U}\) intercept date ranging between 14.63 ± 0.23 (MSWD = 1.3, n =13) to 13.51 ± 0.21 (MSWD =1.5, n=12) (Fig. 10 a). The chondrite-normalized REE patterns (Fig. 12 a) in monazite for Tg are strongly enriched in light REEs relative to the heavy REEs with marked negative Eu (Eu/Eu* of 0.005–0.05) and positive Ce anomalies (Ce/Ce* of 1.02–1.13). In comparison to 2mg, Tg has a wide range of Th (12,175–126,255 ppm) and comparatively high U (2842–82,528 ppm) contents. In (Fig. 12a, b) the younger grains are shown in red and are interpreted to represent the age of crystallization of the melt. The average melt temperature calculated for monazite saturation are 767°C (2mg) and 689°C (Tg) using the calibration of Montel (1993).

6. Petrochronology interpretation

6.1. Zircon petrochronology

In the sillimanite-bearing gneiss sample (SK-10–8), elongate and concentrically zoned and zircon rims yield a large range of zircon ages between ca. 2640 and ca. 14 Ma, with significant populations at ca. 2640 Ma, 500 Ma and a spread of Cenozoic \(^{206}\text{Pb}/^{238}\text{U}\) ages that range from ca. 33–14 Ma. Zircon can experience Pb loss due to recrystallisation and radiation damage (e.g. Cherniak and Watson, 2001). Metamict zircon grains in sample SK-10–8 indicate that radiation damage has altered their internal structure, increasing the susceptibility to alteration by aqueous fluids (Fig. 9c). Discordance in the U–Pb system is due to Pb loss during subsequent metamorphic events (Hoskin and Schaltegger, 2003). The spread of Cenozoic zircon ages could also represent Pb loss, potentially representing a mixing of two or more melting events that form a tie line between ca. 30 Ma and 14 Ma. However, there are several pieces of evidence that point to prolonged crystallisation during crustal melting rather than Pb loss. (i) The Cenozoic zircons preserve textural complexity including oscillatory zoned zircon rims, suggesting multiple periods of melt crystallization. The youngest ages are yielded from elongate euhedral zircon crystals, suggesting new zircon growth at ca. 14 Ma. (ii) The yielded Cenozoic zircon ages sit within the range of previously published HH monazite data from the Sikkim Himalaya, which show prolonged recrystallisation of monazite during metamorphism and partial melting at ca. 31–14.5 Ma (Rubatto et al., 2013; Mottram et al. 2014b). This points to an interpretation of prolonged zircon and monazite growth during partial melting and metamorphism during the main stages of the Himalayan orogeny.

Zircons in the leucogranite samples show well-developed oscillatory zoning, particularly in the rims of grains Supplementary Data Text S2. Previous studies (Pupin, 1980; Wang and Kienast, 1999; Belousova et al., 2006) have suggested that the zircon morphology is closely related to the source and evolution of the parent magma, geological setting, and the chemical composition. The internal zoning in magmatic zircon is pronounced and minor compositional differences are reflected in this zoning with variations in REE, Th, and U (Corfu, 2003; Belousova et al., 2006). The zircon crystallization ages ranging of ca. 19–15 Ma indicate that the leucogranites underwent prolonged crystallization, consistent with the findings of Hopkinson et al. (2020) who suggested a protracted period of crustal melting in the eastern Himalayas.

The magnitude of Ce and Eu anomalies in zircon have been widely used in estimating redox conditions of magmas.
Fig. 9. U-Pb zircon data from HHS sillimanite gneiss sample SK-10-8. Labelled ages are ²⁰⁶Pb/²³⁸U; individual spot ages ± 2 s uncertainties. (a) Tera Wasserburg plot of all zircon data, (b) Tera Wasserburg plot of < 33 Ma zircons in detail; (c) Wetherill plot of all zircon data; (d) Wetherill plot of zircon grains that yield < 500 Ma ages; (e) Wetherill plot of <ca. 50 Ma zircon data in detail, (f) kernel density plot of all ²⁰⁶Pb/²³⁸U zircon spot ages; (g) Cathodoluminescence (CL) images of typical zircon textures colour-coded in concordia plots by texture.

Fig. 10. Terra-Wasserberg plots of monazite data. (a) Sample ZY-LGT-1; (b) sample ZY-LG-2. Monazite populations are shown in different colors based on Eu* and Y (ppm) for samples ZY-LGT-1 and ZY-LG-2 from Sikkim HHS respectively; see text for discussion of significance.
Experimental results, as well as studies on natural samples, suggest that Ce anomalies in zircon increase with increasing oxygen fugacity and/or decreasing temperature (Pettke et al., 2005; Taylor et al., 2009; Claiborne et al., 2010; Trail et al., 2012). Positive Ce anomalies in the studied zircons point towards their formation in a high oxygen fugacity environment. Eu/Eu* ratios are less effective for understanding the redox conditions because Eu/Eu* ratios are also sensitive to fractional crystallization of feldspar or equilibration with restitic feldspar (Trail et al., 2012). Magmatic zircons are generally LREE depleted and enriched in HREEs with a clear negative Eu anomaly and positive Ce and Sm anomalies (Hoskin and Schaltegger, 2003). This suggests that the zircons from the study are magmatic in origin.

6.2. Monazite petrochronology

Monazite grains in both the two-mica (ZY-LG-2) and tourmaline (ZY-LGT-1) leucogranite yield Miocene crystallization ages around ca. 14 Ma, with some evidence of prolonged crystallization for ca. 1 Ma of ca. 15–14 Ma. The chondrite-normalized REE patterns of monazites in both sample ZY-LGT-1 (Tg) and ZY-LG-2 (2mg) are enriched in LREE relative to HREE (Fig. 12a, b) with marked Eu anomalies, which change through time in sample ZY-LGT-1. The negative Eu anomaly suggests co-crystallization of monazite with feldspar during melt crystallization (Holder et al., 2020), where Eu favorably partitions into feldspar. The monazite Eu anomaly could also reflect changes in oxygen fugacity of the system (Holder et al., 2020). Variation in the Eu anomaly suggests prolonged crystallization of monazite during feldspar growth supporting a significant period of leucogranite emplacement.

6.3. Comparison of zircon and monazite age systematics

In HHS gneiss sample, zircon ages range of ca. 32–14 Ma, in leucogranite samples, zircon records a range of inherited ages and a spread of Miocene ages of ca. 19–15 Ma, whereas monazite yields a cluster of ages ca. 14 Ma. Zircon growth is limited in sub-solidus conditions and dissolves less readily in the melt.
thereby preserving inherited ages (Kohn et al., 2015). Monazite does not record inherited ages in this study, instead preserving purely Miocene crystallization ages consistent with crystallization in a minimum-temperature melt (Lederer et al., 2013).

7. Discussion

7.1. Inherited zircon provenance

Zircon populations from both the felsic HHS gneiss (SK-10-8) and the two-mica leucogranite (ZY-LG-2) contain older inherited zircon grains, with ages ranging to a maximum age of ca. 2400 Ma to ca. 500 Ma. Pre-Himalayan zircons, typically lack oscillatory zoning, and record age populations at ca. 1800 Ma, ca. 800 Ma (only seen in 2mg sample) and ca. 500 Ma, consistent with a HHS source (Mottram et al., 2014a and references therein).

The oldest Paleoproterozoic ages (ca. 1823 Ma and ca. 1718 Ma) from the 2mg are consistent with populations interpreted to have formed in a continental volcanic arc setting during the formation of Columbia supercontinent (Kohn et al., 2010; Yang et al., 2021) and are consistent with gneiss ages reported from the LHS of Sikkim (Lingtsa gneiss, ca. 1834 Ma and ca. 1853 Ma; Mottram et al., 2014a), and from Himalayan gneisses elsewhere in the orogen (Wangtu orthogneiss, Jutogh Group of Himachal Pradesh (ca. 1878 Ma; Kohn et al., 2010); Bomdila Group augen gneiss from western Arunachal Pradesh (ca. 1750 Ma; Yin et al., 2010a, 2010b)). Note that although zircons of ca. 1800 Ma are also consistent with a LHS source, they do not provide strong evidence for such an origin given that HHS gneisses near to the MCT yield detrital zircons of similar age (Mottram et al., 2014a).

The late Neoproterozoic (ca. 710–953 Ma) ages from the studied 2mg are similar to ages from the HHS meta-igneous intrusions; e.g. Cuona, southern Tibet (ca. 855–809 Ma; Wang et al., 2017), Chaur, western Himalaya (ca. 823 Ma; Singh et al., 2002; ca. 929 Ma; Singh et al., 2020), Jutogh granitic gneiss, western Himalaya (ca. 861 Ma; Singh et al., 2020); Bhutan granitic gneiss (ca. 825 Ma; Richards et al., 2006), Hapoli granitic gneiss, Arunachal Himalaya (ca. 878–825 Ma; Yin et al., 2010a); Peshawar granitic gneiss (ca. 816 Ma; Ahmad et al., 2013); Black Mountain complex, Bhutan (ca. 823 Ma age; Dipietro and Isachsen, 2001). These magmatic ages have been variously related to: the breakup of the Rodinia supercontinent (Li et al., 2008; Wang et al., 2017), the Malani magmatic event (Sharma, 2005; Ahmad et al., 2013); subduction and collision during Grenvillian orogeny and formation of the Rodinia supercontinent (Singh et al., 2020), and the presence of an active continental margin at the northern margin of India (Spencer et al., 2019; Zhang et al., 2021).

A few grains of zircons from the 2mg yield ca. 481 and ca. 444 Ma ages whereas the HHS felsic gneiss yielded an age of ca. 490 Ma pointing towards Cambro–Ordovician magmatism (Myrow et al., 2016). Similar Cambro–Ordovician magmatism is evidenced by the emplacement of a series of granite plutons within the HHC (Argles et al., 1999; Gehrels et al., 2006; Cawood et al., 2007; Spencer et al., 2012; Singh, 2019). The upper Cambrian-Lower Ordovician tectonic event, also known as the Bhimphedian orogeny (Cawood et al., 2007; Palin et al., 2018) or Kurgiakh orogeny (Srikantia, 1981; Gehrels et al., 2003; Myrow et al., 2006), preceded the deposition of post-orogenic molasses and the formation of a stable shelf margin around ca. 460–440 Ma (Palin et al., 2018). Cawood et al. (2007) proposed that the Andean-type Bhimphedian orogeny, deformed the Proto-Tethys margin of Gondwana.

7.2. Leucogranite source

The North Sikkim leucogranites are characterized by high SiO2 contents (>70 wt.%), Al2O3 (13.69–15.8 wt.%) and A/CNK values (1.03–1.37) indicating a granitic and strongly peraluminous nature. These traits are identical to those found in other Miocene Himalayan leucogranites which are indicative of their crustal origin (Schräer et al., 1986; Le Fort et al., 1987; Guo and Wilson, 2012; Liu et al., 2014; Xie et al., 2018). A crustal origin for Himalayan leucogranites is confirmed by both zircon and bulk rock isotopic characteristics (Hopkinson et al., 2017).

The nature of their sedimentary protoliths can be constrained by elemental discrimination diagrams. In the discrimination diagram (Fig. 14a,b) the 2mg bodies fall in the greywacke field indicating their formation by partial melting of a plagioclase-rich metasedimentary rock, whereas the Tg leucogranites fall in the pelitic-derived field. Plagioclase and muscovite control the Rb-Ba-Sr budget in S-type granites as Sr and Ba are compatible in plagioclase, while Rb is compatible with muscovite (Sylvester, 1998). Higher Rb/Sr (Rb/Sr > 2) ratios are suggestive of fluid-absent muscovite melting while lower (Rb/Sr < 2) ratios in fluid fluxed melting.
have been attributed to the involvement of biotite (Inger and Harris, 1993; Visonà and Lombardo, 2002; Gou et al., 2016).

Sylvester (1998) suggested that granites derived from greywacke and pelites vary in their Rb/Sr ratio wherein greywacke-derived granites have lower Rb/Sr as compared to the ones derived from pelitic sediments. The studied 2mg from North Sikkim predominantly suggest a clay-poor source with a few samples falling in a clay-rich region (Fig. 14c) whilst Tg have higher Rb/Sr ratio and fall in the clay-rich region (Fig. 14c). This suggests that the studied 2mg are largely derived from sediments of greywacke composition whilst Tg have been sourced from pelitic sources.

The presence of inherited Paleoproterozoic, Neoproterozoic (ca. 928 Ma and ca. 710 Ma), and Cambro-Ordovician zircons, which are only found in the HHS (Richards et al., 2006; Guo and Wilson, 2012; Hopkinson et al., 2017) suggest a HHS source for the studied leucogranites. Furthermore, based on variations in whole-rock Nd, zircon U-Pb and Hf isotopes from the Oligocene–Miocene leucogranites of the Bhutan Himalayas (Hopkinson et al., 2017, 2020), suggested that ca. 32–17 Ma leucogranites from the Higher Himalayas were derived from pelitic to semi-pelitic HHS lithologies, although some contributions from melting the LHS were detected in the youngest melts from at least ca. 17 Ma.

7.3. Himalayan crustal melting

There are two models for the formation of leucogranites; fluid-fluxed and fluid-absent partial melting (Harris et al., 1993, 1995; Gao and Zeng, 2014; Weinberg, 2016; Gao et al., 2017). In metasedimentary and granitic protoliths, dehydration melting involves muscovite and/or biotite breakdown (Dyk et al., 2020). Previous studies have suggested that the Himalayan leucogranites formed predominantly by muscovite-dehydration melting (Harris and Inger, 1992; Imayama and Suzuki, 2013; Xie et al., 2018), whilst other studies have suggested that crustal anatexis occurred as both dehydration melting and fluid fluxed melting of muscovite bearing metasedimentary sources (Gao et al., 2017).

In water present partial melting feldspar is consumed in a greater proportion relative to mica resulting in elevated Sr, Ba, Ca and Sr/Y ratio with low Rb and Rb/Sr ratios. During dehydration melting, more mica is consumed compared to feldspar, resulting in higher Rb and lower Sr, Ba and Rb/Sr ratio (Gao et al., 2017; He et al., 2021). In the Rb/Sr vs. Ba plot (Harris and Inger, 1992), water-present partial melting is associated with a low and uniform Rb/Sr ratio with elevated Ba contents while muscovite dehydration melting shows a negative correlation between Rb/Sr ratio and Ba content (Fig. 13). The studied leucogranites display a negative trend in the Rb/Sr vs Ba plot, indicating that they are all products of muscovite dehydration melting. This differs from the findings of Gou et al. (2016), who interpreted that 2mg and Tg from southern Tibet have been produced by biotite dehydration melting and muscovite dehydration melting respectively.

7.4. Generation of leucogranite in the Sikkim Himalayas

Zircon in sillimanite-bearing felsic gneiss sample (SK-10–8) records a prolonged period of zircon crystallization during partial melting of ca. 33–14 Ma. Despite the small size of the dataset presented in this manuscript, the data presented here are consistent with the wider literature from the Sikkim Himalayas which estimates the timing of the anatexis in the HHS of ca. 37–14 Ma across the Sikkim Himalayas (Kellett et al., 2013; Rubatto et al., 2013; Mottram et al., 2014b), and at ca. 32–17 Ma in the northern Sikkim HHS gneisses, proximal to the studied leucogranites. Based on the monazite record, the leucogranites studied here are interpreted to have crystallized at ca. 15–14 Ma, consistent with similar leucogranite emplacement across the Himalayas (as discussed in the introduction).

A schematic model can be developed for the formation of the Sikkim leucogranites based on both our new data and previous constraints from the literature (Fig. 15). In the Sikkim Himalayas petrochronological studies have demonstrated that the STD was active from ca. 17 Ma to ca. 15–14 Ma (Catlos et al., 2004) and from ca. 23.6 Ma to ca. 13 Ma (Kellett et al., 2013). The MCT propagated structurally downwards through time resulting in progressive tectonic accretion of LHS material from the footwall to the hanging wall (Bollinger et al., 2006; Mottram et al., 2015b; Hopkinson et al., 2020). In northern Sikkim, it is interpreted that partial melting of the HHS occurred of ca. 37–16 Ma, before deformation propagated structurally downwards through the MCT zone of ca. 14.5–9 Ma (Mottram et al., 2014b). Despite the relatively small number of leucogranites studied in this manuscript, the ages are comparable to the zircon ages (ca. 17–15 Ma) and monazite ages (ca. 15–14 Ma) reported from deformed leucogranites from North Sikkim (Catlos et al., 2004). These ages overlap with both the timing of incorporation of material into the MCT zone and normal faulting on the STD. The inherited zircon and geochemical data from the studied leucogranites indicate an HHS greywacke-pelitic source. This fits into a model where leucogranites were sourced from muscovite-dehydration decompression melting during exhumation of the HHS due to coeval movement along the STD and MCT zone, resulting in melting of mostly tectonically interleaved (Mottram et al., 2014a) HHS material (Hopkinson et al., 2020).

8. Conclusions

This study examines the conditions and chronology of crustal melting and leucogranite emplacement in the Darjeeling-Sikkim region of the eastern Himalayas. Prolonged crustal melting occurred in the HHS of > 33–14 Ma which provides the crustal melting sources for Himalayan leucogranites. Both two-mica and tourmaline leucogranites from the Sikkim Himalayas have high contents of SiO2, Al2O3 and low contents of MgO and TiO2 and are enriched in LILE (Rb, U, K, and Pb) and depleted in HFSE (Nb, Ta, and Ti) suggesting that they were derived from crustal melts.
The two-mica leucogranites are predominantly derived from grey-wacke sources whereas the tourmaline leucogranites have been sourced from metapelites. Melting occurred from mica, predominantly muscovite, breakdown under fluid-absent conditions. The U-Pb ages of zircon range of ca. 19–15 Ma, overlapping with crustal melting in the source HHS rocks. Monazite crystallization ages of ca. 15–14 Ma suggest leucogranite emplacement coeval with both accretion along the MCT zone and movement along the STDS and further provides an important basis for constraining the timing and mechanism of petrogenesis of leucogranites in the Siikim Himalayas.

**CRediT authorship contribution statement**

Tanya Srivastava: Conceptualization, Formal analysis, Investigation, Writing – original draft, Visualization. Nigel Harris:
Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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References


