Trace element compositions and petrogenetic implications of zircons in high-pressure granulites

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ABSTRACT

Trace element compositions of zircon play a pivotal role in unveiling their origins, the geological significance of zircon U–Pb ages, and tectonic settings of formation of their host rocks. Here, we present an integrated dataset of new cathodoluminescence images, U–Pb ages, and trace element compositions of zircons from pelitic, felsic and mafic high-pressure granulites in the eastern Himalayan orogen, and compare these against a compilation of previously reported metamorphic zircon trace elements data from various metamorphic rocks. Zircons in the studied granulites are predominantly subhedral, have prismatic and ovoid shapes, and mostly have an inherited core (magmatic and detrital, respectively) and a metamorphic rim. Metamorphic domains of zircons show planar, patchy and fir-tree zoning, and yielded metamorphic, anatectic and melt crystallization ages of 44–9 Ma, while the inherited magmatic cores of zircons show oscillatory zoning, and obtained Early Paleozoic protolith ages of 510–480 Ma. The metamorphic zircon domains in the granulites generally have high Hf, U and Sc contents, and low HREE, Y, Th, Ti, Nb and Ta contents. These indicate that the metamorphic zircons formed during the growth of garnet, rutile and Th-rich minerals (monazite, allanite and titanite), and the host rocks underwent a prolonged high-pressure granulite-facies metamorphism and partial melting. This study shows that the metamorphic zircons from garnet-bearing metamorphic rocks have lower Yb, Y, Nb and Ta contents than those of zircons from magmatic rocks in different tectono-magmatic settings. We reveal that the metamorphic zircons in garnet-bearing metamorphic rocks have higher U/Yb and Sc/Yb ratios than zircons in various magmatic rocks, and that the metamorphic zircons in ultrahigh temperature rocks have similar high Th/U ratios (mostly > 0.1) to magmatic zircons, whereas metamorphic zircons from lower-grade metamorphic rocks have low Th/U ratios (mostly < 0.1).

INTRODUCTION

Zircon occurs throughout the Earth's crust and upper mantle, being a common constituent in sedimentary, magmatic and metamorphic rocks (Finch and Hanchar, 2003; Hoskin and Schaltegger, 2003; Andersen, 2005; Rubatto, 2017). Zircon is widely used in petrogenetic and geochronological studies due to its physical and chemical durability, its low common lead content, and high closure temperature in the U–Th–Pb system (> 900 °C; Lee et al., 1997; Cherniak and Watson, 2000). Zircon is therefore a powerful tool for determining the timing and duration of geological events (Engi et al., 2017; Reimink, 2023). In particular, the trace element composition of zircon is highly sensitive to zircon-forming reactions that occur during the mineralogical evolution of its host rock (e.g., Belousova et al., 2002; Rubatto, 2002; Grimes et al., 2015).

With the continued development of in-situ and microanalysis techniques, it has become possible to obtain ever-more precise zircon U–Pb ages and corresponding trace element compositions, which may even be collected simultaneously from the same domains using split-stream mass spectrometry. Considerable progress has been made in recent years regarding interpreting trace element contents within the context of zircon genesis in magmatic rocks. In particular, zircon trace element compositions have been used to track magmatic sources (e.g., Wang et al., 2012; Carley et al., 2014;

Burnham and Berry, 2017), magmatic crystallization processes (e.g., Claiborne et al., 2006, 2010; Carley et al., 2022), to identify magmatic rock types (e.g., Hoskin and Ireland, 2000; Belousova et al., 2002), to distinguish tectonic settings of formation (e.g., Grimes et al., 2007, 2015), to explore mineralization (e.g., Dilles et al., 2015; Gardiner et al., 2017), and to estimate crustal thicknesses (e.g., McKenzie et al., 2018; Balica et al., 2020; Tang et al., 2021; Sundell et al., 2022). Similar studies have been performed on metamorphic zircon; in particular, linking metamorphic zircon U–Pb ages with trace element contents or ratios to define the timing and duration of metamorphism, partial melting, and melt crystallization of high-grade metamorphic and anatectic rocks (e.g., Rubatto et al., 2013; Ding et al., 2021a, b), and application of the titanium-in-zircon thermometer to calculate crystallization temperatures (e.g., Watson and Harrison, 2005; Ferry and Watson, 2007; Baldwin et al., 2007; Clark et al., 2018; Jiao et al., 2020). Although much progress has been made in the study of metamorphic zircons, the degree to which trace element compositions of zircons vary between metamorphic rocks with different whole-rock chemical compositions and metamorphic conditions, the trace element behaviors and petrogenetic significance of zircons during high-grade metamorphism and partial melting, and trace element differences between metamorphic and magmatic zircons remain to be revealed.

In this contribution, we present results of a comprehensive study involving whole-rock geochemistry, zircon cathodoluminescence (CL) images, U–Pb ages, and trace element compositions in the pelitic, felsic and mafic high pressure (HP) granulites from the eastern Himalayan orogen, compile trace element data of metamorphic zircons in various metamorphic rocks, and systematically compare trace element compositions between metamorphic and magmatic zircons. We reveal internal zoning profiles, trace element compositions and petrogenetic features of zircons in the HP granulites, the timing and duration of high-grade metamorphism, partial melting and melt crystallization of the HP granulites, and trace element differences between metamorphic and magmatic zircons. This study provides new insight into the petrogenetic implications of trace elements in metamorphic zircon and provides a framework with which they may be interpreted.

GEOLOGICAL BACKGROUND

The Himalayan orogen in the southern Tibetan Plateau was formed during the Cenozoic collisional orogeny between the India and Asian continents, and extends for more than 2500 km from the Western Himalayan Syntaxis (or Nanga Parbat Syntaxis) in Pakistan to the Eastern Himalayan Syntaxis (or Namche Barwa Syntaxis) in China (Burg et al., 1984; Yin and Harrison, 2000). The orogen consists mainly of four laterally and roughly parallel tectonostratigraphic units; from north to south, they are the Tethyan Himalayan Sequence (THS), Greater Himalayan Sequence (GHS), Lesser Himalayan Sequence (LHS), and Sub-Himalayan Sequence (Neogene Siwalik Formation) (Fig. 1A; Yin, 2006). These units are sequentially separated by the Yarlung-Tsangpo Suture Zone (YTSZ), the Southern Tibet Detachment System, the Main Central Thrust, and the Main Boundary Thrust (Fig. 1A).

The Eastern Himalayan Syntaxis (EHS) include three tectonic units, from north to south; the Lhasa terrane, the YTSZ and the Himalayan sequences (Fig. 1B). The Lhasa terrane represents the southern segment of the Asian continent, and consists of Precambrian basement, Paleozoic to Mesozoic strata, and Mesozoic to Cenozoic magmatic rocks (Pan et al., 2006; Zhu et al., 2011; Dong et al., 2022). The YTSZ represents remnants of the Neo-Tethyan Ocean situated between the India and Asian continents, and mainly consists of ophiolitic mélange (Yin, 2006). The Himalayan sequences represent the northern margin of the Indian continent, and include the THS and GHS (Yin and Harrison, 2000). The THS mainly consists of Neoproterozoic to Mesozoic sedimentary strata, and underwent greenschist to epidote-amphibolite facies metamorphism (Yin, 2006). The GHS include the upper and lower units (Fig. 1B), the former consists mainly of orthogneiss, paragneiss, schist, mafic granulite, amphibolite, marble and calc-silicate rock, and underwent amphibolite- to granulite-facies metamorphism and partial melting, and the latter consists of amphibolites and gneisses, and underwent amphibolite-facies metamorphism and partial melting (e.g., Xu et al., 2012; Zhang et al., 2012).

The upper GHS unit in the Eastern Himalayan Syntaxis contains widely exposed HP granulites, which are dominated by felsic granulites, with minor pelitic and mafic granulites, and occur as interlayers and lenses within amphibolites, gneisses and calc-silicate rocks (e.g., Zhang et al., 2022). Although the modal abundances of minerals in the pelitic and felsic granulites are different, these two types of HP granulites have similar peak mineral assemblage of garnet + kyanite + plagioclase + K-feldspar + biotite + quartz + rutile (Liu and Zhong, 1997; Ding and Zhong, 1999; Guilmette et al., 2011; Xiang et al., 2013; Zhang et al., 2015). The mafic HP granulites have a peak mineral assemblage of garnet + clinopyroxene + plagioclase + quartz + rutile + titanite (Zhong and Ding, 1996; Liu and Zhang, 2014; Zhang et al., 2022). The peak metamorphic P–T conditions of the pelitic, felsic and mafic granulites were respectively estimated to be 1.3–1.8 GPa and 800–900°C (Liu and Zhong, 1997; Ding and Zhong, 1999; Xiang et al., 2013; Zhang et al., 2015; Tian et al., 2020), 1.5–1.6 GPa and 800–850°C (Guilmette et al., 2011; Tian et al., 2016), and 1.4–1.9 GPa and 800–900°C (Ding et al., 2001; Liu and Zhang, 2014; Zhang et al., 2018, 2022; Kang et al., 2020). These HP granulites were often overprinted by medium pressure (MP) granulite- to amphibolite-facies retrograde metamorphism, which resulted in the HP granulites transforming into garnet-bearing or garnet-free amphibolites, gneisses and schists (e.g., Guilmette et al., 2011; Zhang et al., 2015, 2022; Kang et al., 2020).

Recent studies indicated that the HP granulites in the EHS have undergone extensive partial melting during HP and high-temperature (HT) metamorphism (Guilmette et al., 2011; Xiang et al., 2013; Zhang et al., 2015, 2018, 2022; Tian et al., 2016, 2019; Kang et al., 2020). The mafic granulites underwent dehydration melting of amphibole, with the melting reaction of Amp + Qz ± Pl = Grt ± Cpx + melt producing ~15–20 vol. % of melt per unit rock volume (Zhang et al., 2018, 2022; Kang et al., 2020). The pelitic and felsic granulites underwent dehydration melting of muscovite and biotite, with the melting reaction of Ms ± Bt + Qz + Pl = Grt + Ky + Kfs + melt producing 20–30 vol. % and 10–20 vol. % of melt per unit rock volume, respectively (Guilmette et al., 2011; Xiang et al., 2013; Zhang et al., 2015; Tian et al., 2019). Many in-situ zircon U–Pb geochronological analyses show that the HP granulites have a wide range of ages from 50 to 7 Ma, which represent the duration of high-grade metamorphism, partial melting and melt crystallization (Ding et al., 2001; Booth et al., 2004; Liu et al., 2007; Xu et al., 2010; Zhang et al., 2010, 2012, 2015; Su et al., 2012; Zeng et al., 2012; Tian et al., 2020; Kang et al., 2020).

SAMPLES AND ANALYTICAL METHODS

Sample Description

The studied HP granulite samples were collected from the EHS in the Jiala, Zhibai, Pai, Danniang and Baga regions (Fig. 1B), including eight pelitic granulites, six felsic granulites and six mafic granulites (Table 1). All the samples have a clearly migmatitic structure, with alternating melanosome and leucosome bands that are oriented parallel to gneissose banding in the granulites (Figs. 2A, 2C and 2E). The pelitic HP granulites consist of garnet, kyanite, plagioclase, K-feldspar, quartz, biotite, and sillimanite, and accessory minerals rutile, ilmenite, apatite, monazite and zircon (Figs. 2A and 2B; Table 1). Coarse-grained garnets occur as porphyroblasts and mostly have an inclusion-rich core and inclusion-poor or -free rim (Fig. 2B). The included minerals are quartz, plagioclase and biotite (Fig. 2B). The garnet rims are partly replaced by symplectitic corona of biotite, quartz and plagioclase (Fig. 2B). Kyanite occurs as elongate laths, although is occasionally replaced by sillimanite or biotite (Fig. 2B). Oriented mica laths, prismatic kyanite, and ribbons of K-feldspar, plagioclase and quartz define the mineralogical banding (Fig. 2B). The felsic HP granulites consist of garnet, plagioclase, K-feldspar, quartz and biotite, with minor kyanite and/or sillimanite, and accessory minerals of apatite, allanite, rutile, ilmenite, monazite and zircon (Figs. 2C and 2D; Table 1). The coarse-grained garnet and plagioclase occur as porphyroblasts, whereas medium- to fine-grained plagioclase, K-feldspar, biotite and quartz occur throughout the matrix (Fig. 2D). Some garnets are replaced by symplectitic corona of quartz + plagioclase + biotite along their rims (Fig.

2D). The oriented arrangement of matrix minerals quartz, plagioclase, K-feldspar and biotite defines the gneissic foliation (Fig. 2D). The mafic HP granulites contain garnet, clinopyroxene, amphibole, plagioclase, quartz and minor biotite, with the accessory mineral rutile, ilmenite, titanite and zircon (Figs. 2E and 2F; Table 1). The porphyroblastic garnet cores contain quartz, plagioclase, clinopyroxene, biotite and titanite inclusions, and are often replaced by symplectitic corona of amphibole + plagioclase + quartz along their margins (Fig. 2F). The studied zircons occur both as matrix minerals and as inclusions within garnet and other minerals in all granulites. The mineral abbreviations used in this paper follow the guidelines of Whitney and Evans (2010).

Analytical Methods

Whole-rock major element analyses were determined by X-ray fluorescence spectrometry (XRF) at the Wuhan Sample Solution Analytical Technology Co., Ltd., Wuhan, China. Analytical precision was generally better than 2% for all elements. Whole-rock trace element compositions were analyzed using an Agilent 7700e inductively coupled plasma-mass spectrometer (ICP-MS) at Wuhan Sample Solution Analytical Technology Co., Ltd., which gave precisions better than 10% for most of the elements analyzed. Zircon cathodoluminescence (CL) images were obtained by a TESCAN Integrated Mineral Analyzer (TIMA) at the Institute of Geology, Chinese Academy of Geological Sciences, Beijing. Zircon U–Pb isotope and trace element analyses were performed simultaneously using laser ablation inductively coupled plasma mass spectrometry (LA–ICP–MS) at the Institute of Geology, Chinese Academy of Geological Sciences, Beijing. The laser-ablation spot diameter was 25 μm. Details of these analytical methods are provided in Supplemental Text S1, and the analytical data are listed in Tables S1 and S2.

RESULTS

Whole-Rock Major and Trace Elements

The studied HP granulites have variable major and trace element compositions (Table S1); the pelitic granulites have higher Al_2O_3 (17.36–22.90 wt.%), lower CaO (0.57–1.99 wt.%) and Na_2O (0.51–2.13 wt.%) contents compared to the other two types of granulites; the felsic granulites show higher contents of SiO_2 (65.02–73.37 wt.%) and K_2O (2.35–5.80 wt.%) but lower TFe_2O_3 (2.69–6.57 wt.%); and the mafic granulites have higher CaO (7.30–11.08 wt.%), MgO (3.56–6.70 wt.%), P_2O_5 (0.15–0.73 wt.%), TFe_2O_3 (10.21–18.07 wt.%), and lower K_2O (0.47–1.73 wt.%) (Fig. S1; Table S1).

The HP granulites contain 29–1165 ppm of Ba, 19–577 ppm of Sr, 10–292 ppm of Rb, 8.1–41 ppm of Sc, 5.5–48 ppm of Nb, 2.8–55 ppm of Th, 2.0–10 ppm of Hf, 0.74–9.3 ppm of U, and 0.50–2.9 ppm of Ta (Table S1). In the primitive mantle-normalized trace element spider diagram, the granulites are enriched in Rb, Th, U and K, but have negative Ba, Nb, Sr and Ti anomalies (Fig. S2A). These granulites have high light rare earth element (LREE) contents (56–356 ppm), and low heavy rare earth element (HREE) contents (5.2–21 ppm) (Table S1), and therefore are relatively enriched in LREE, and depleted in HREE; Eu anomalies are weak in the mafic granulite samples (Eu/Eu* = 0.53–1.08) but strong in the pelitic and felsic granulite samples (Eu/Eu* = 0.48–0.70 and Eu/Eu* = 0.26–0.82, respectively) (Fig. S2B; Table S1).

Zircon Morphology and Internal Structures

Zircon grains separated from the pelitic HP granulites are euhedral to subhedral, and prismatic shape, with lengths of 50–250 μm and widths of 20–200 μm (Figs. 3A and S3). The zircons mostly show a well-preserved core–mantle–rim structure (Figs. 3A and S3). The cores are characterized by bright luminescence in CL images, and have variable shapes, sizes, and zoning patterns; the wide mantles generally have dark luminescence, prismatic or stubby habits and weak patchy zoning or unzoning; the narrow to wide rims have moderate luminescence and weak planar zoning or are unzoned (Figs. 3A and S3). Some zircons occur as single grains that lack internal zoning (Fig. S3).

Zircon grains in the felsic HP granulites are euhedral to subhedral, prismatic in shape, and have lengths of 50–300 μm and widths of 30–150 μm (Figs. 3B and S4). The zircon grains mostly possess core–rim structures (Figs. 3B and S4). Most cores have bright luminescence and clear oscillatory zoning, while a small number of cores have dark luminescence and blurry zoning (Figs. 3B and S4). Some core features are truncated and surrounded by overgrowth rims (Figs. 3B and S4). These rims have moderate luminescence in CL images, and show planar and weak patchy zoning (Figs. 3B and S4).

Zircon grains in the mafic HP granulites are subhedral, prismatic or ovoid in shape, and 30–250 μm in length (Figs. 3C, 3D and S5). Most rounded and subrounded zircons show patchy or fir-tree zoning and have variable luminescence (Figs. 3C and S5). Zircons in one mafic granulite sample have a core–rim structure (Figs. 3D and S5). Most cores show bright luminescence and oscillatory zoning, while a few chaotic cores show convoluted zoning, while the rims have dark luminescence and planar zoning patterns (Figs. 3D and S5).

Zircon U–Pb Dating

Two hundred and seventy six spot analyses located at mantle and rim domains of zircons from eight pelitic granulite samples yielded concordant and variable $^{206}Pb/^{238}U$ ages ranging from 44.1 ± 0.8 to 14.0 ± 0.7 Ma (Fig. 4; Table S2). On the whole, the zircon mantles of most samples yielded relatively old $^{206}Pb/^{238}U$ ages of ~44 to 30 Ma, while the rims yielded relatively young $^{206}Pb/^{238}U$ ages of ~30 to 14 Ma (Fig. 4). The zircon mantles in sample T19-26-15 have $^{206}Pb/^{238}U$ ages of 30.0 ± 1.4 to 26.1 ± 0.7 Ma, and the rims give $^{206}Pb/^{238}U$ ages of 25.6 ± 0.7 to 14.0 ± 0.7 Ma (Fig. 4). Sixty dating sites located at the rims of zircons from six felsic granulite samples yielded concordant and variable $^{206}Pb/^{238}U$ ages ranging from 31.1 ± 1.1 to 9.3 ± 0.2 Ma (Fig. 5; Table S2). One hundred and three dating sites located at zircon rims and single grains from six mafic granulite samples obtained concordant $^{206}Pb/^{238}U$ ages ranging from 34.4 ± 1.1 to 14.8 ± 1.1 Ma (Fig. 6; Table S2).

A wide age range spanning ~44 to 7 Ma is obtained when these new data are combined with previously reported U–Pb ages of zircons in HP granulites from the study area (Fig. 7; Table 1). Specifically, the age spectrum of zircon mantle and rims from the pelitic granulites show two peaks at ~37 Ma and ~23 Ma (Fig. 7A); zircon rims and single zircon domains in the mafic granulites have a single age cluster at ~25 Ma (Fig. 7B); the ages of zircon rims in the felsic granulites show a main peak centered at ~22 Ma (Fig. 7C).

One hundred and five dating sites located at the cores of zircon from six felsic granulites and a mafic granulite yielded concordant $^{206}Pb/^{238}U$ ages, ranging from 507–470 Ma, 527–462 Ma, 517–444 Ma, 510–473 Ma, 501–468 Ma, 518–460 Ma and 557–468 Ma, with similar weighted mean $^{206}Pb/^{238}U$ ages of 487 ± 4.5 Ma (mean square of weighted deviates [MSWD] = 2.7, n = 12/12), 487 ± 3.8 Ma (MSWD = 2.9, n = 16/17), 502 ± 4.2 Ma (MSWD = 1.1, n = 15/17), 489 ± 7.2 Ma (MSWD = 0.43, n = 17/17), 478 ± 4.6 Ma (MSWD = 1.4, n = 12/12), 481 ± 6.1 Ma (MSWD = 1.5, n = 21/21) and 513 ± 9 Ma (MSWD = 0.76, n = 5/9), respectively (Figs. 5A–F and 6D).

Zircon Trace Element Compositions

Zircon mantle and rim domains, and homogenous single grains in the studied granulites have high Hf (8883–17,910 ppm), U (19–6683 ppm) and Sc (218–709 ppm) contents, and low HREE (3.35–206 ppm), Yb (1.20–125 ppm), Th (0.32–89 ppm), Ti (0.50–9.97), Nb (0.013–4.97 ppm), and Ta (0.005–3.85 ppm) contents, and variable Th/U ratios (0.002–0.62; Fig. 8; Table S2). The single zircon grains and zircon rims in the mafic granulites have lower Hf (8883–13,710 ppm) and U (19–983 ppm) contents, and higher Eu/Eu* values (0.10–2.13) compared to zircon mantles and/or rims in the pelitic and felsic granulites (Hf = 11,021–17,910 ppm and 11,900–17,656 ppm, U = 187–6143 ppm and 157–6683 ppm, Eu/Eu* = 0.06–0.98 and 0.02–1.10, respectively) (Figs. 8A, 8B and 8F; Table S2).

Zircon cores in the felsic and mafic granulites have lower Hf (8960–15352 ppm), and higher Ti (0.68–13.0), and similar Sc (297–411 ppm) contents compared to the rims in the same sample (Fig. 8; Table S2). However, compared to single grains and zircon mantles and/or rims, zircon cores have higher REE (426–1667 ppm), Th (96–988 ppm), HREE (327–1327 ppm), Y (561–2469 ppm), P (158–1509 ppm), Nb (0.91–6.30 ppm) and Ta (0.69–4.81 ppm) contents and Th/U ratios (0.18–1.2) (Fig. 8; Table S2). The single zircon grains, and mantles and rims of zircons in the HP granulites show flat, or even depleted HREE patterns and with weakly negative Eu anomalies, while zircon cores exhibit steep HREE patterns with significant negative Eu anomalies (Fig. 9).

DISCUSSION

Zircon Genesis and U–Pb Age Significance

Zircons are chemically robust, can be precisely dated, and can survive a range of magmatic, hydrothermal, and metamorphic geological processes, and typically exhibit a variety of morphologies and internal zoning patterns (Vavra et al., 1996, 1999; Corfu et al., 2003; Wu and Zheng, 2004; Kohn and Kelly, 2017; Rubatto, 2017). Magmatic zircons generally have euhedral to subhedral crystal habits, and show prismatic shapes, oscillatory and/or sector zoning, while metamorphic zircons commonly have more complex shapes and zonation profiles (e.g., Hoskin and Schaltegger, 2003). In general, metamorphic and anatectic zircons that form under granulite-facies conditions exhibit rounded-ovoid or prismatic shapes, and planar, patchy and fir-tree zoning (e.g., Corfu et al., 2003; Rubatto, 2017). The term anatectic zircon refers to zircon that has grown during prograde metamorphism and associated partial melting (Yakymchuk, 2023).

Zircons in the studied HP granulites can be categorized into two groups according to their textural features. One type of zircon has a core–mantle–rim or core–rim structure, and another type is a single homogenous grain without any internal structure (Figs. 3 and S3–S5). Zircon cores in the pelitic granulites have variable shapes, sizes and zoning patterns (Figs. 3 and S3), typical of inherited detrital zircons in meta-sedimentary rocks (e.g., Vavra et al., 1996; Rubatto et al., 2001). The zircon cores from all felsic granulites and one mafic granulite mostly show euhedral to subhedral prismatic shape, and oscillatory zoning (Figs. 3, S4 and S5), and have high REE (426–1667 ppm) contents and Th/U ratios (0.18–1.2), steep REE patterns with significantly negative Eu anomalies (Figs. 8 and 9), typical of a magmatic origin (e.g., Hoskin and Schaltegger, 2003). In this case, the zircon cores are considered to be inherited magmatic zircons sourced from the protoliths of the felsic and mafic granulites; as such, the Early Paleozoic ages of 513–478 Ma obtained from these grains represent the age of formation of the protoliths to these granulites.

The zircon mantle and rim domains, and single zircon grains in the studied granulites mostly show subhedral prismatic or ovoid shape, and planar, patchy, or fir-tree zoning (Figs. 3 and S3–S5), have low Th/U ratios (mostly < 0.1), and HREE contents (mostly < 100 ppm) and flat or even depleted HREE patterns (Figs. 8 and 9). These are typical features of metamorphic and anatectic zircons that grew coevally with garnet during high-grade metamorphism and partial melting (e.g., Rubatto, 2002; Rubatto et al., 2013). The depleted HREE patterns in zircon can be attributed to the hosting HP granulites containing abundant garnet (Fig. 2; Table 1), which is the main host of HREE in metamorphic rocks (e.g., Rubatto, 2002; Baxter et al., 2017). In addition, nanogranite (former melt inclusions) and metamorphic zircon occur together as inclusions within garnet in the felsic HP granulites from the study area, indicating that metamorphic and anatectic zircon (and host garnet) formed together during partial melting of the granulites (Liu et al. 2023). Some zircon rims or single zircon grains of the granulites have relatively high HREE contents (> 100 ppm) and steep HREE patterns (Figs. 8 and 9), which may be related to the breakdown of garnet during retrograde metamorphism. Our petrographic observations indicate that garnet rims are often partly replaced by symplectitic corona of quartz, plagioclase, biotite, and amphibole during retrograde metamorphism of the various HP granulites (Figs. 2B, 2D and 2F).

This work shows that metamorphic zircon domains in the pelitic, felsic and mafic granulites record wide $^{206}Pb/^{238}U$ ages of 44.1–14.0 Ma, 31.1–9.3 Ma and 34.4–14.8 Ma, respectively (Figs. 4–6; Table S2). These individual age ranges are all interpreted to have particular geological significance, rather than simply reflecting Pb loss or resetting of the U–Pb system during retrogression of the granulites. This supposition is justified as follows: (1) where zircons have core–mantle–rim or mantle–rim structures, the mantle domains always have older ages than the rim domains for each individual sample, and the mantles generally have older ages than the rims across *all* samples (Figs. 3 and 4). (2) Zircon Th/U and Eu/Eu* values in the pelitic granulite samples systematically vary with age (Fig. 10), whereby Th/U ratios gradually increase as grains become younger (Fig. 10A), and Eu/Eu* values progressively decrease from 44 Ma to 35 Ma, and from 25 Ma to14 Ma (Fig. 10B). (3) Considering that rocks within the Himalayan orogen have not been subjected to overprinting and reworking by later tectonic thermal events, that zircon has excellent physical and chemical durability, and it has a high closure temperature for the U–Th–Pb system (e.g., Cherniak et al., 1997; Lee et al., 1997), we consider it impossible that the studied metamorphic and anatectic zircon domains occurred Pb loss during a single stage of prograde and retrograde metamorphism, and partial melting and melt crystallization. (4) The current in-situ microanalysis methods employed here carry an analytical error of ~2% for zircon U–Pb dating, such that we can obtain a ≤ 1 Myr chronologic resolution for the Cenozoic (< 50 Ma) evolution of Himalayan rocks (Kohn, 2014), allowing several events to be clearly distinguished. (5) The metamorphic and anatectic zircon domains in single high-grade metamorphic rock sample from the present study area and other areas of the Himalayan orogen commonly yielded varying ages, which are similar to those of this study. These variable ages have been robustly interpreted to represent the duration of metamorphism, partial melting and melt crystallization, as opposed to simply representing varying degrees of Pb loss or overprinting (e.g., Rubatto et al., 2013; Wang et al., 2013; Zhang et al., 2015; Ding et al., 2021a).

Several studies have demonstrated that the HP granulites in the study area witnessed a long-lived metamorphic, anatectic and melt crystallization process that lasted over 20 to 30 Myr, and followed a clockwise P–T–t path with initial prograde metamorphism and partial melting, minor cooling and decompression during early post-peak retrogression, and more extensive cooling and decompression and melt crystallization associated with final retrogression and exhumation (e.g., Zhang et al., 2015, 2022; Tian et al., 2019, 2020; Kang et al., 2020). We believe that the studied granulites should have experienced the same metamorphic conditions and P–T–t path as the previously reported granulites from the studied area, as all of these rocks are closely spatially related. We interpret the age interval from 44 Ma to 35 Ma obtained by the zircon mantle domains of the pelitic granulites to represent the duration of prograde metamorphism prior to partial melting (Fig. 10) based on the following reasons: (1) The mode of zircon gradually increases through time (Fig. 7A), which is consistent with zircon growth dominantly occurring during prograde metamorphism below the solidus of metamorphic rocks (e.g., Yakymchuk et al., 2017). (2) Zircons have low Th/U ratios of < 0.02 (Fig. 10A), typical of metamorphic zircons coexisting with hydrous fluids (e.g., Rubatto, 2017). (3) Zircon Eu/Eu* values decrease with time (Fig. 10B), which be related to the growth of Eu-rich plagioclase of the pelitic granulite during prograde metamorphism below the solidus. This is consistent with the pelitic granulites in this study area having experienced prograde metamorphism crossing the following reaction Bt + Qz = Grt + Pl + H_2O (Xiang et al., 2013).

We interpret the age interval from 35 Ma to 25 Ma to represent the duration of partial melting of the pelitic granulites based on the following lines of evidence: (1) Zircons of this age are relatively rare compared to other periods (Fig. 7A), as supported by previous studies having shown that zircon rarely grows *during* migmatisation (e.g., Kelsey et al., 2008; Yakymchuk and Brown, 2014; Kohn et al., 2015). (2) Zircon Th/U ratios increase with decreasing age (Fig. 10A), which may be related to an increasing degree of partial melting of the pelitic granulites, and breakdown of Th-rich monazite during partial melting. It is widely accepted that zircons crystallized from melts have higher Th/U ratios than those from metamorphic fluids (e.g., Rubatto, 2017), and monazite often undergoes decomposition during partial melting (e.g., Kelsey et al., 2008; Yakymchuk and Brown, 2014; Yakymchuk et al., 2018).

We consider the age range of 25–14 Ma to be the duration of retrograde metamorphism and melt crystallization of the pelitic granulites based on the following lines of evidence: (1) Zircons are relatively abundant (Fig. 7A), implying that this was a period of significant zircon growth. This is consistent with the growth of zircon in high-grade metamorphic and anatectic rocks dominantly occurring during retrograde metamorphism and melt crystallization (Yakymchuk et al., 2017 and references therein). (2) Zircon domains have elevated Th/U ratios (Fig. 10B), which are typical features of grains that have crystallized from melts. (3) The Eu/Eu* values of zircon decreases gradually with decreasing ages (Fig. 10B), which may be related to the growth of plagioclase during retrograde metamorphism and melt crystallization. This is in line with that the studied pelitic granulites contain significant amounts of plagioclase, and the porphyroblastic garnets are partly replaced by biotite, plagioclase and quartz along their rims, having formed via the reaction Grt + melt = Bt + Pl + Qz (Fig. 2B).

Based on the above discussion, we interpret that the studied pelitic granulites witnessed a prolonged prograde metamorphism, partial melting and retrograde metamorphism and melt crystallization up to 30 Myr. The studied mafic granulites have similar age ranges and peaks to the pelitic granulites (Fig. 7B), and the felsic granulites have relatively narrow age ranges, but similar age peaks to the pelitic and mafic granulites (Fig. 7C). Metamorphic and anatectic zircons rarely grew in the felsic granulites due to those rocks having a low proportion of H_2O-bearing minerals and limited free aqueous fluid availability during metamorphism and anataxis. Thus, as all three types of granulites are closely spatially associated and show no structural contact relationships, we speculate that these rocks experienced the same timing and duration of metamorphic, anatexis, and melt crystallization. This is consistent with the previous conclusion that different granulites in the study area record similar metamorphic histories, anatectic conditions, and durations of melting events (Zhang et al., 2015, 2018, 2022; Peng et al., 2018; Tian et al., 2019, 2020; Kang et al., 2020).

Comparison of Trace Elements of Metamorphic and Magmatic Zircons

Zircons commonly have different trace element compositions or ratios depending on the geological environment in which they form and the geological processes responsible, and due to differences in the host rock's mineralogy and whole-rock compositions (e.g., Hoskin and Schaltegger, 2003; Rubatto, 2017). Belousova et al. (2002) revealed that zircons from granitoids, syenites and mafic rocks mostly have higher Y contents than those from kimberlites and carbonatites, and zircons from granitoids mostly have higher U contents than those in other rocks (Fig. 11). Grimes et al. (2015) showed that magmatic zircons in continental arc rocks have relatively high U and Th, and low Ti contents, magmatic zircons in oceanic island rocks have relatively high Nb and Ti contents, and magmatic zircons in mid-ocean ridge rocks have relatively low U and Th, and high Ti contents (Fig. 12).

The inherited magmatic zircons with the Early Paleozoic ages in the studied felsic and mafic granulites have similar Y, U, Nb and Ta contents, and Nb/Ta ratios to the zircons in granitoids and mafic magmatic rocks, respectively (Figs. 11A–C). These inherited magmatic zircons also have similar to U, Nb, Ti, Yb and Th contents to the magmatic zircons from continental arc rocks (Fig. 12). These support the previous conclusions that some felsic and mafic metamorphic rocks in the Himalayan orogen were derived from the Early Paleozoic arc-type magmatic rocks, and the northern margin of Indian continent underwent an Early Paleozoic Andean-type orogeny (e.g., Cawood et al., 2007; Wang et al., 2012; Zhang et al., 2012; Gao et al., 2019).

This study shows that metamorphic zircons in the mafic granulites have lower Hf and U contents, and higher Eu/Eu* values than the pelitic and felsic granulites (Figs. 8 and 11A). The metamorphic zircons in the pelitic granulites mostly have higher Y contents than those in the felsic and mafic granulites (Figs. 8C and 11C). Our compiled trace element compositions show that metamorphic zircons in garnet-bearing pelitic, felsic and mafic metamorphic rocks have similar Y, U, Nb, Ta, Yb and Th contents and Nb/Ta ratios to those in the studied pelitic, felsic and mafic HP granulites, respectively (Figs. 11 and 12). Metamorphic zircons from garnet-free amphibolites have higher Y and Yb than those from garnet-bearing mafic metamorphic rocks (Figs. 11 and 12).

The metamorphic zircons in the studied HP mafic granulites and the garnet-bearing mafic metamorphic rocks compiled here have lower Y contents than magmatic zircons in granitoids and mafic rocks (Figs. 11A and 11D), and lower Nb and Ta contents than those of granitoids (Figs. 11B and 11E). Compared to magmatic zircons that formed in magmatic rocks from different tectonic settings, the metamorphic zircons in garnet-bearing metamorphic rocks exhibit lower Nb and Yb contents (Fig. 12). The Y, Yb, Nb and Ta in metamorphic rocks are commonly hosted in garnet, rutile and ilmenite (e.g., Zack et al., 2002; Luvizotto and Zack, 2009; Tan et al., 2018). These minerals are common in garnet-bearing metamorphic rocks, which resulted in low Y, Yb, Nb and Ta contents of metamorphic zircons. The metamorphic zircons from garnet-free amphibolites also have lower Nb contents than magmatic zircons from different tectonic settings (Fig. 12).

The U/Yb, Nb/Yb and Sc/Yb ratios can be used to distinguish magmatic zircons formed in continental arc, ocean-island and mid-ocean ridge settings (Grimes et al., 2015). Continental arc magmatic zircons have relatively high U/Yb and Sc/Yb ratios, mid-ocean ridge magmatic zircons have relatively low U/Yb and Nb/Yb ratios, and ocean-island magmatic zircons have relatively high Nb/Yb ratios (Fig. 13). The inherited magmatic zircons in the studied HP granulites have similar U/Yb, Nb/Yb, Sc/Yb ratios to continental arc magmatic zircons (Figs. 13A, 13C and 13D). The metamorphic zircons in garnet-bearing metamorphic rocks mostly have higher U/Yb ratios than those of magmatic rock zircons (Figs. 13A and 13B). A small number of metamorphic zircons in the garnet-bearing metamorphic rocks have relatively low but similar U/Yb ratios to continental arc zircons due to their low U contents (Figs. 13A and 13B; Tables S2 and S3). The metamorphic zircons in the garnet-free mafic metamorphic rocks have similar U/Yb and Nb/Yb ratios to continental arc magmatic zircons (Fig. 13B). The metamorphic zircons from the HP granulites have higher Sc/Yb ratios than magmatic zircons (Figs. 13C and 13D). Therefore, we propose that the U/Yb and Sc/Yb ratios can be used to distinguish metamorphic zircons in garnet-bearing metamorphic rocks from magmatic zircons in various tectonic environments.

Th/U ratios of Metamorphic and Magmatic Zircons

Previous studies have shown that magmatic and metamorphic zircons have similar U contents, but magmatic zircons have higher Th contents than metamorphic zircons; therefore, magmatic zircons often have Th/U ratios of >0.1, whereas metamorphic zircons mostly have Th/U ratios of <0.1 (Williams and Claesson, 1987; Rubatto and Gebauer, 2000; Belousova et al., 2002; Hoskin and Schaltegger, 2003). Our compiled data for magmatic zircons from West Australian magmatic rocks show that Th and U contents of magmatic zircons mostly vary between 20 ppm and 2000 ppm, and Th/U ratios mostly more than 0.1, with a major peak of 0.7 (Figs. 14 and 15A; Kirkland et al., 2015).

Nonetheless, the recently compiled Th and U compositions of metamorphic zircons revealed that some metamorphic zircons have similar Th contents and Th/U ratios to magmatic zircons. For example, Th and U contents of metamorphic zircons in West Australia are mostly between 0.1 ppm and 1000 ppm, and 20 ppm and 2000 ppm, and Th/U values vary dramatically from <0.001 to >10, with a prominent peak at 0.6 (Figs. 14A and 15B; Yakymchuk et al., 2018). Metamorphic zircons worldwide generally have similar Th and U contents, and Th/U ratios to those from Australia (Figs. 14A and 15C; Roberts et al., 2024). These indicate that metamorphic zircons have similar U contents, but a wider range in Th content compared to magmatic zircons. Therefore, the metamorphic zircons have variable Th/U ratios, and with Th/U ratios of a significant portion of metamorphic zircons overlapping those of magmatic zircons (Figs. 14 and 15). Consequently, recent studies have argued that Th/U ratios cannot be solely used as the criterion for distinguishing magmatic zircons from metamorphic zircons (e.g., Yakymchuk et al., 2018; Roberts et al., 2024), zircons in low and medium temperature metamorphic rocks have low Th/U ratios (mostly <0.1), while zircons in high temperature (HT) to ultrahigh temperature (UHT, >900 ℃) metamorphic rocks have high Th/U ratios (mostly >0.1; Rubatto, 2017; Yakymchuk et al., 2018).

This study shows that the HP granulitic zircons from the Eastern Himalayan Syntaxis mostly have Th of 1–50 ppm, and U of 10–2000 ppm, and Th/U of <0.1 (Figs. 14B and 15D; Table S2). The Th and U compositions compiled here for metamorphic zircons in various metamorphic rocks show that zircons in UHT metamorphic rocks mostly have Th contents of 10–300 ppm, and U contents of 10–1000 ppm, and Th/U ratios of >0.1, which are generally similar to those of the magmatic zircons (Figs. 14B and 15D; Table S4). In contrast, metamorphic zircons in other rocks, including MP, HP and ultrahigh pressure (UHP) metamorphic rocks, have similar Th and U contents and Th/U ratios (mostly <0.1) to the metamorphic zircons in the studied HP granulites (Figs. 14B and 15D; Table S4). Based on our new results, we speculate that the previously compiled metamorphic zircons with Th/U ratios of >0.1 were probably sourced from UHT metamorphic rocks – or at least formed at UHT conditions, even if the host rocks themselves have since experienced recrystallization at lower metamorphic grades. Therefore, we suggest that a Th/U ratio of 0.1 can be used to distinguish UHT metamorphic and magmatic zircons from other metamorphic zircons.

5. Conclusion

(1) Zircons in pelitic, felsic and mafic HP granulites from the study area often contain an inherited core (detrital or magmatic) surrounded by a metamorphic mantle and rim, or are single metamorphic grains. Metamorphic domains in zircon yielded variable ages of 44–9 Ma, indicating that the HP granulites underwent a long-lasting metamorphic, anatectic and melt crystallization process. The

inherited magmatic cores of zircon yielded ages of 510–480 Ma, indicating that the host felsic and mafic granulites were derived from Early Paleozoic magmatic rocks.

(2) The metamorphic zircons in the HP granulites have high Hf, U and Sc contents, and low HREE, Y, Th, Ti, Nb and Ta contents, and formed in the presence of garnet, rutile and Th-rich minerals. This indicates that the host rocks experienced HP and HT granulite-facies metamorphism and partial melting.

(3) Metamorphic zircons from garnet-bearing metamorphic rocks mostly have lower Yb and Nb contents, and higher U/Yb and Sc/Yb ratios than magmatic zircons. Metamorphic zircons in UHT metamorphic rocks and various magmatic zircons have similar but high Th/U ratios (mostly >0.1), while the other metamorphic zircons generally have low Th/U ratios (<0.1).

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FIGURE CAPTIONS:

Fig. 1 (A) Sketch geological map of the Himalayan orogen (modified after Zhang et al., 2022). (B) Geological map of the Eastern Himalayan Syntaxis (modified after Zhang et al., 2023), showing the locations of major samples discussed in our work.

Fig. 2 Field photographs (A, C, E) and microphotographs (B, D, F) of the investigated HP granulites. (A and B) Pelitic granulite (T19-26-12), consisting of garnet, kyanite, biotite, plagioclase, K-feldspar and quartz with minor sillimanite, rutile and ilmenite. (C and D) Felsic granulite (T19-14-12), consisting of garnet, biotite, plagioclase, K-feldspar and quartz with minor allanite and sillimanite. (E and F) Mafic granulite (T19-27-10), consisting of garnet, clinopyroxene, amphibole, plagioclase, quartz and minor titanite and biotite. These granulites all show clear deformation fabrics (foliations) and contain felsic leucosomes. The diameters of the coins in A and C are 2.05 cm and 1.9 cm, respectively.

Fig. 3 Cathodoluminescence images of representative zircon grains in the pelitic (A), felsic (B), and mafic HP granulites (C and D), showing the analyzed spot locations and relevant ^{206}Pb/^{238}U ages (in Ma). The core, mantle, and rim domains of zircon are marked in a representative grain for each sample.

Fig. 4 U-Pb concordia diagrams of zircons in the pelitic HP granulites. (A)

Metamorphic rims. (B–H) Metamorphic mantles and rims.

Fig. 5 U-Pb concordia diagrams of zircon magmatic cores and metamorphic rims in the felsic HP granulites.

Fig. 6 U-Pb concordia diagrams of zircons in the mafic HP granulites. (A, B, C, E and F) Single metamorphic zircon grains. (D) Magmatic core and metamorphic rim of zircon.

Fig. 7 U-Pb age histograms of the metamorphic zircons from the pelitic (A), mafic (B), and felsic HP granulites (C). The data sources are as in Table 1.

Fig. 8 Trace element compositions of metamorphic mantle and rim, and inherited magmatic core of zircons from the HP granulites.

Fig. 9 Chondrite-normalized REE patterns of zircons from the HP granulites. The dashed lines represent inherited magmatic cores, and the solid lines represent metamorphic mantles and rims and single grains. Chondrite values are from Sun and McDonough (1989).

Fig. 10 Zircon ages versus Th/U ratios (A) and Eu/Eu* values (B) from the HP granulites, showing the division of metamorphic, anatectic and melt crystallization phases.

Fig. 11 Trace elements compositions of the studied HP granulitic zircons (A–C) and compiled data for metamorphic zircons from various metamorphic rocks (D–F, the data and sources are given in Table S3). The compositional fields of magmatic zircons from various magmatic rocks are after Belousova et al. (2002).

Fig. 12 Trace elements compositions of zircons from the HP granulites (A–C) and compiled data for metamorphic zircons from various metamorphic rocks (D–F, the data and their sources are listed in Table S3), and previously compiled magmatic zircons from the different tectono-magmatic settings (after Grimes et al., 2015).

Fig. 13 U/Yb versus Nb/Yb, and Sc/Yb versus U/Yb and U/Yb diagrams of magmatic and metamorphic zircons. The magmatic zircons in different tectono-magmatic settings are shown by two-dimensional kernel density estimation, with the contours of 50, 80, 90 and 95% levels (after Grimes et al., 2015). The metamorphic zircon data in (A, C and D) are from this study (Tables S2), and in (B) are from the literature (Table S3).

Fig. 14 U versus Th diagrams of metamorphic and magmatic zircons. (A) The previously complied magmatic and metamorphic zircons. (B) Metamorphic zircons reported and compiled in this study (the data sources are listed in Tables S2 and S4), and magmatic zircons compiled by Kirkland et al. (2015).

Fig. 15 Histograms of zircon Th/U ratios. (A) Magmatic zircons. (B, C) Metamorphic zircons. (D) Metamorphic zircons from this study and the literature (The data sources are listed in Tables S2 and S4).

TABLE CAPTIONS

Table 1 The major features of the studied HP granulites.

SUPPLEMENTAL MATERIALS

Text S1 Analytical Methods.

Fig. S1 Harker diagrams of major elements of the studied HP granulites.

Fig. S2 Primitive mantle-normalized trace element patterns (A) and chondrite-normalized REE patterns (B) of the HP granulites. The primitive mantle and chondrite values are from Sun and McDonough (1989).

Fig. S3 Cathodoluminescence images of zircons in the pelitic HP granulites, showing the analyzed spot locations and relevant ages (in Ma).

Fig. S4 Cathodoluminescence images of zircons in the felsic HP granulites, showing the analyzed spot locations and relevant ages (in Ma).

Fig. S5 Cathodoluminescence images of zircons in the mafic HP granulites, showing the analyzed spot locations and relevant ages (in Ma).

Table S1 Whole-rock major and trace element compositions of the HP granulites.

Table S2 Zircon U-Pb dating and trace elements data of the HP granulites.

Table S3 Compiled trace element compositions (in ppm) of metamorphic zircons in various metamorphic rocks.

Table S4 Compiled Th and U compositions (in ppm) of metamorphic zircons in various metamorphic rocks.