Geochronology and REE geochemistry of zircon and garnet in pelitic gneisses from the Higo metamorphic terrane, Kyushu, Japan: Constraints on the timing of high-temperature metamorphism

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The Higo metamorphic terrane in west-central Kyushu, southwest Japan, consists of a sequence of greenschist- to granulite-facies metamorphic rocks. This study reports the U-Pb Zrn ages and rare earth elements (REE) characteristics of Zrns and Grts from pelitic gneisses in the Higo metamorphic terrane to constrain its timing of high-temperature metamorphism. Based on the morphology, internal textures, occurrences, Th/U ratios, REE patterns and ages of Zrn, and the heavy REE (HREE) partition coefficient between Zrn and Grt, the results indicate that the prograde metamorphism occurred at ~ 120 Ma immediately after precursor deposition at 120–130 Ma and peak metamorphic conditions were attained during the period between 110 and 120 Ma. The ~ 110 Ma age might be indicative of the onset of cooling and exhumation after the peak metamorphic stage. Retrograde metamorphism continued at least to 105 Ma as obtained from Zrns in the granitic intrusion.

Keywords: Higo metamorphic terrane, U-Pb ages, Zircon, Garnet, Rare earth elements

INTRODUCTION

In recent geological and petrological investigations throughout the world, Zrn U-Pb chronology has been the most convincing age determination method for high- and ultrahigh-temperature (HT-UHT) metamorphic rocks. Because of the durability of Zrns against chemical and physical processes, it can record the ages of several geological events such as magmatism and metamorphism. Generally, to interpret Zrn ages, Zrn crystal morphology, internal zonal structures and Th/U ratios have been applied (e.g., Hoskin and Schaltegger, 2003; Wu and Zheng, 2004). Zrns with the euhedral shape, well-developed growth zoning (e.g., oscillatory), and the high-Th/U ratio (>0.10) are normally recognized as those of magmatic origin, while those of metamorphic origin are characterized by a rounded or ovoid shape showing a structureless internal texture, sector or fir-tree zoning, and the low-Th/U ratio (<0.10) (e.g., Hoskin and Schaltegger, 2003). How-

ever, these criteria are not always supported, particularly in the case of metamorphic Zrn in HT metamorphic rocks because the growth or replacement of the Zrn can occur during several stages of HT metamorphism including crystallization from melts during crustal anataxis (e.g., Rubatto, 2002), solid-state mineral reactions via metamorphic breakdown reactions (e.g., Fraser et al., 1997), and recrystallization of protolith Zrns (e.g., Hoskin and Black, 2000). Thus, to understand the meaning of Zrn ages, it is necessary to consider the behaviors of metamorphic Zrn during HT metamorphism. Recently, Zrn trace element geochemistry has been utilized to investigate the processes or conditions of Zrn growth/recrystallization (e.g., Whitehouse and Platt, 2003; Hokada and Harley, 2004; Rubatto, 2017). In addition, REE partitioning between Zrn and Grt ($D_{Zrn/Grt}^{HREE}$) has been applied to connect Zrn age with metamorphic condition through the assessment of their equilibrium (e.g., Harley, 2002; Rubatto, 2002; Whitehouse and Platt, 2003). Both Zrn and Grt prefer HREE relative over light to middle REE (L-MREE), resulting in equilibrium $D_{Zrn/Grt}^{HREE}$ values that are near unity (Harley, 2002; Whitehouse and Platt, 2003). There-

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fore, the combination of external, internal, and geochemical characteristics as well as Zrn behavior during HT metamorphism is crucial to link Zrn U-Pb ages to the geological events.

The Higo metamorphic terrane has been recognized as one of the HT metamorphic terranes in Japan. It consists of high-grade metamorphic rocks up to UHT granulite-facies that are characterized by a clockwise *P-T* evolution and peak metamorphic conditions of 7.8–9.0 kbar and 900–960 °C (Osanai et al., 1998). Previously, this terrane was proposed to have metamorphosed at ~260–230 Ma (Osanai et al., 1998; Hamamoto et al., 1999; Osanai et al., 2006); however, the recently conducted Zrn geochronology has suggested that the timing of HT-UHT metamorphism is approximately 110–120 Ma because of clustered age data (Sakashima et al., 2003; Takagi and Arai, 2003; Dunkley et al., 2008; Maki et al., 2014; Osanai et al., 2017; Suga et al., 2017). Though its metamorphic evolution in terms of petrography has been well understood, of which the prograde and retrograde metamorphic processes for each metamorphic zone were well reported on the basis of reaction textures, chemical zoning of minerals, and mineral inclusions in porphyroblasts (Hamamoto et al., 1999; Maki et al., 2004; Osanai et al., 2006), it is still obscured in term of *P-T-t* evolution because of the lack of information regarding the timing of the prograde

and retrograde stages. To solve this problem, investigations of geochemical features and the behavior of metamorphic Zrn related to Grt can be useful. Thus, in this study, we applied laser ablation inductively couple plasma mass spectroscopy (LA-ICP-MS) U-Pb isotopic dating to Zrns as well as REE analyses of Zrns and Grts from pelitic gneisses in the Higo metamorphic terrane. Showing the behaviors of the Zrn and Grt, this study constrains the timing of HT metamorphism. Minerals abbreviation used in this paper is that of Whitney and Evans (2010).

GEOLOGICAL SETTING AND SAMPLE DESCRIPTION

The Higo metamorphic terrane is in the central-western part of Kyushu and is a sequence of metamorphic rocks with increasing metamorphic grade southward from greenschist- to granulite-facies (Obata et al., 1994; Osanai et al., 1998) (Fig. 1). The five metamorphic zones are defined by metapelite mineral assemblages as follows: zone A by Chl + Ms, zone B by Bt + Ms + And, zone C by Kfs + Sil + Bt, zone D by Grt + Crd + Bt, zone E by Opx-bearing granulite (Obata et al., 1994), and zone F by hyper aluminous Spr-bearing granulite only found in association with the meta-peridotite intrusion in zone C (Osanai et al., 2006) (Fig. 1). The metamorphism was

Figure 1. Metamorphic zonation of the Higo metamorphic terrane defined by the mineral assemblages of the metapelites (Obata et al., 1994; Osanai et al., 2006). Zone A by Chl + Ms, zone B by Bt + Ms + And, zone C by Kfs + Sil + Bt, zone D by Grt + Crd + Bt, zone E by Opx-bearing granulite, and zone F by Spr-bearing granulite. The gray points indicate the sample localities in this study.

also recorded for each zone corresponding to their mineral assemblage as follows: greenschist-facies in zone A, amphibolite-facies in zones B and C, and granulite-facies in zones D and E (Obata et al., 1994; Osanai et al., 1998). In addition, the *P-T* conditions have been estimated for each zone as follows: 3.5–4.2 kbar and 640–720 °C for zone C, 4.8–6.0 kbar and 740–820 °C for zone D, 6.0–7.2 kbar and 800–870 °C for zone E, and 7.8–9.0 kbar and 900–960 °C for zone F (Osanai et al., 1998).

In this study, we analyzed Grt-Crd-Bt gneiss (43007B), Grt-Bt gneiss (430T01B), and discordant Bt granite (42901G) collected from zone D (Fig. 1). These gneisses show the migmatitic textures (Fig. 2a) and were intruded by later discordant granite (Fig. 2b). The Grt-Crd-Bt gneiss consists of Grt, Crd, Bt, Pl, Kfs, and Qz (Fig. 2c) with accessory fibrolite, Ap, Zrn, Mnz, and Gr. Fibrolite only occurs as an inclusion in Crd and Grt (Figs. 2d and 2e). Porphyroblastic Grt contains numerous fine-grained inclusions of fibrolite that occur only in the rim (Fig. 2e), Bt, Qz, Pl, Ap, Zrn, and Mnz. The Grt resorption texture is observed, which is surrounded by the secondary Bt and/or Crd, Qz, and Pl (Fig. 2c). Crd also occurs as a subhedral porphyroblast with inclusions such as Bt and fibrolite (Fig. 2d). The Grt-Bt gneiss also shares quite similar petrographical features to those of Grt-Crd-Bt gneiss, without the presence of fibrolite and Crd (Figs. 2f and 2g). The Grt porphyroblasts in this gneiss are partly surrounded by Bt and Pl (Fig. 2f) or maintain a preserved euhedral shape (Fig. 2g). In addition, some Pl shows the subhedral to euhedral shape (Figs. 2f and 2g). Zrn occurs as inclusions in Grt (only in the Grt-Bt gneiss: Fig. 2g), Bt, Qz, and Pl with/without Crd and in the matrix, particularly near the Grt resorption area (Fig. 2c). The discordant Bt granite contains abundant coarse-grained Qz and Pl (Fig. 2h) along with subordinate amounts of Bt and minor accessory minerals including Ap, Zrn, and Mnz. Secondary Prh is occasionally found with Bt (Fig. 2h). Zrn crystals in this rock are notably quite euhedral, occurring in the matrix or included in the Pl, Qz, and Bt.

U-Pb ZRN DATING AND REE CHARACTERISTICS

U-Pb dating for Zrn separated from pelitic gneisses and discordant granite and REE analyses for the separated Zrn and Grt in thin section (for the pelitic gneisses) were performed using an Agilent 7500cx quadrupole ICP-MS with a New Wave Research UP-213 Nd-YAG UV (213 nm) laser ablation system at Kyushu University, Japan. These analytical procedures are described in detail by Adachi et al. (2012) for the dating and Nakano et al.

(2010) for the REE analyses. The error of all analyses is 2 sigma intervals. The analytical results for the age and REE composition of the Zrn grains are listed in Supplementary Tables S1 and S2, respectively; major element and REE compositions of the Grt are presented in Supplementary Tables S3 and S4, respectively (Supplementary Tables S1–S4 are available online from <https://doi.org/10.2465/jmps.181115>).

Grt-Crd-Bt gneiss (43007B) contains transparent to translucent Zrns with ovoid, stubby, and minor elongated shapes, 40–100 µm in length. The cathodoluminescence (CL) images of these Zrns exhibit a faintly oscillatory to unzoned dark-CL core overgrown by a bright-CL structureless rim or sector zoned or structureless grains without any rim (Figs. 3a and 3b). The core and sector zoned grains mostly show high-Th/U ratios (>0.10) and scattered ages of ~ 150–280, 350, 450–500, 650, 900, 1000, 1650, and 1850 Ma (Figs. 3a, 3b, and 3h). However, five data points from the structureless overgrown rims and grains provide clustered data with a weighted mean age of 114.4 ± 2.4 Ma ($n = 5$, MSWD = 0.51) and low-Th/U ratios (<0.10) (Figs. 3b and 3g).

In the Grt-Bt gneiss (430T01B), the Zrn grains are 80–200 µm in length, have long prismatic subhedral to euhedral with a lesser amount of ovoid to equant shapes, colorless, and transparent. Their internal texture shows banded oscillatory zoning, blurred zoning, and core-rim textures with an oscillatory zoned core overgrown or truncated by the unzoned or faintly oscillatory zoned rim (Figs. 3c and 3d). The blurred zoning grain and the truncating rim show Th/U ratios of approximately 0.10 and an age range of 121–125 Ma, which defines a weighted mean age of 123.2 ± 2.5 Ma ($n = 4$, MSWD = 0.65) (Figs. 3d and 3g). The unzoned or faintly oscillatory rims overgrowing the core show low-Th/U ratios (<0.10) and concordant data of 105–115 Ma (concordia age of 111.2 ± 2.0 Ma; $n = 7$, MSWD = 0.14) (Figs. 3d and 3g). Similar ages of 107–118 Ma (111.0 ± 1.1 Ma, $n = 29$, MSWD = 0.92) were also obtained from sub- to euhedral grains with banded oscillatory zoning and scattered Th/U ratios of 0.06–1.21 (Figs. 3d and 3g). The relic core indicates an age population of ~ 130–180, 210–300, 350–400, 800, 1800, and 2100 Ma with high-Th/U ratios (Figs. 3c, 3d, and 3h).

Discordant Bt granite (42901G) contains transparent to translucent Zrns with sub- to euhedral or ovoid shapes, 30–70 µm in length. The CL images of these Zrns show mainly structureless zoning and a small number of faintly oscillatory zoning or core-rim textures with an unzoned core overgrown by a structureless thin rim (Figs. 3e and 3f). The core and weak oscillatory zoned grains show high-Th/U ratios (>0.10) and scattered ages of ~ 150–

Figure 2. Mode of occurrence (a) and (b). Photomicrographs of the Grt-Crd-Bt gneiss (c), (d), and (e). Grt-Bt gneiss (f) and (g). Bt granite (h). (a) Migmatitic texture with anatectic melt observed in the Grt-Crd-Bt gneiss. (b) Grt-Bt gneiss and amphibolite intruded by later granite. (c) Grt porphyroblast partly replaced by Crd, Bt, and Qz in the Grt-Crd-Bt gneiss. (d) Subhedral Crd porphyroblast occurring with inclusions of Sil and Bt and pinitized rim in the Grt-Crd-Bt gneiss. (e) Sil occurs as fibrolite in the Grt rim in the Grt-Crd-Bt gneiss. (f) Grt porphyroblast partly replaced by Bt, Qz, and Pl in the Grt-Bt gneiss; the euhedral shape of Pl is also apparent. (g) Euhedral Grt porphyroblast including euhedral Pl and Zrn inclusions in the Grt-Bt gneiss. (h) Coarse-grained Qz and Pl as well as Prh lenses in the discordant Bt granite. Color version is available online from <https://doi.org/10.2465/jmps.181115>.

210, 280, 330, 750, and 1700 Ma (Figs. 3e and 3f). However, seven data points from structureless grains provide clustered data (concordant age = 105.6 ± 1.3 Ma, $n = 7$, MSWD = 1.8) with low-Th/U ratios (<0.10) (Fig. 3f).

The REE patterns normalized to chondrite values

(McDonough and Sun, 1995) of some representative analyzed Zrns in both pelitic gneisses were characterized by steep patterns enriched in HREE relative to LREE with a negative Eu anomaly and a weak positive Ce anomaly (Fig. 4). However, in the Grt-Bt gneiss, the ~ 120 Ma

Figure 3. Concordia diagrams for the results of LA-ICP-MS Zr U-Pb dating and the representative CL images of the analyzed Zrns with the analytical spots, $^{206}\text{Pb}/^{238}\text{U}$ ages, and Th/U ratios shown for the Grt-Crd-Bt gneiss (a) and (b). Grt-Bt gneiss (c) and (d). Bt granite (e) and (f). In the concordia diagrams, concordant and discordant data are shown with solid and dash ellipses, respectively. Th/U ratio-age plots are shown for the metamorphic domains (g) and detrital domains (h).

Figure 4. REE patterns normalized by chondrite value for the Zr from the Grt-Bt gneiss (a). Grt-Crd-Bt gneiss (b). The gray shadow indicates the inherited core domain, the dashed black line the ~ 120 Ma domain, solid black line for the newly crystallized domain (~ 110 Ma), and gray line for the overgrowth domain (~ 110 Ma). See the text for the details. Chondrite values are after McDonough and Sun (1995).

and other older Zrns show mostly similar REE concentrations and patterns, while most of the ~ 110 Ma Zr grains are relatively enriched in LREE compared to the others (Fig. 4b).

MAJOR ELEMENTS AND REE COMPOSITION OF GRT

Prior to Grt analysis, X-ray elemental maps were developed for major elements (Fe, Mn, Mg, and Ca) and Y for both pelitic gneisses. Grt in the Grt-Crd-Bt gneiss (43007B) has relatively wide homogeneous core zones which have a lower Mn and higher Mg core than that of the thin rim (Fig. 5a). $X_{Mg}[\text{Mg}/(\text{Mg} + \text{Fe}^{2+})]$ values increase from the core ($X_{Mg} = 0.24$), reaching a maximum value in the mantle ($X_{Mg} = 0.25$) and decreasing again to the rim ($X_{Mg} = 0.22\text{--}0.23$) (Table S3). Ca is near-

ly constant throughout the grain with Ca (pfu) ranging from 0.17 to 0.18 (Table S3). In contrast, the Grt in the Grt-Bt gneiss (430T01B) shows normal chemical zoning with an increase in Mg and Ca, a decrease in Mn toward the rim and a subsequent decrease in Mg and Ca, and an increase in Mn at the outer rim (Fig. 5b). X_{Mg} values gradually increase from the core ($X_{Mg} = 0.20$) to the rim ($X_{Mg} = 0.28$) (Table S3). The Grt for both samples is rich in almandine and poor in grossular ($X_{alm} = 0.65\text{--}0.72$; $X_{grs} = 0.03\text{--}0.07$, Figs. 5c and 5d). However, the yttrium zoning patterns of Grt in both gneisses are similar and gradually decreases from the core toward the rim (Figs. 5a and 5b). HREE zoning of Grt is considered to be correlated with mapped Y zoning defining the core-mantle-rim texture. All of the Grt core shows the highest Y concentration, decreasing toward the mantle and rim. The REE patterns of the Grt from both samples are similar with following features: strong LREE depletion, a negative Eu anomaly, and an MREE to HREE with a positive slope for the Grt core, a slightly positive to flat slope for the Grt mantle, and a flat to slightly negative slope for the Grt rim (Figs. 6a and 6b).

DISCUSSION

Metamorphism characteristics for collected pelitic gneisses and retrogression features

Both pelitic gneisses were collected from zone D (Fig. 1) with the similar X_{Mg} values of Grt (Figs. 5c and 5d) possibly indicating that these samples were subjected to a similar metamorphic temperature condition. Therefore, the absence of Crd in the Grt-Bt gneiss while present only in the Grt-Crd-Bt gneiss could be a result of the difference in bulk chemistry. Petrographically, the Grt-Crd-Bt gneiss is mainly composed of Grt, Crd, Bt, Pl, Kfs, and Qz. Some Grts show resorption texture, of which Grts are partly replaced by Bt or Crd suggesting a retrograde reaction: $\text{Grt} + \text{Kfs} + \text{melt} \rightarrow \text{Bt} + \text{Crd} + \text{Qz}$ (Maki et al., 2004; Miyazaki, 2004). The Sil occurrence as fibrolite inclusions in the Grt and Crd porphyroblasts may indicate a melting reaction at a temperature for $\text{Qz} + \text{Kfs} + \text{Pl}$ bearing assemblages as follows: $\text{Bt} + \text{Sil} + \text{Qz} \rightarrow \text{Kfs} + \text{Crd} + \text{Grt} + \text{melt}$ (Maki et al., 2004; Miyazaki, 2004). The Grt-Bt gneiss also contains a similar mineral assemblage excluding Crd and Sil. Some Grt shows resorption texture, replaced by Bt, Pl, and Qz, suggesting a hydration retrograde reaction. These petrographical characteristics for the Grt-Crd-Bt gneiss are typical for those reported from zone D (Obata et al., 1994; Maki et al., 2004) which underwent peak P - T condition of 4.8–6.0 kbar and 740–820 °C (Osanai et al., 1998).

Figure 5. X-ray images for Mn, Mg, Ca and Y in the Grt of the Grt-Crd-Bt gneiss (a), the Grt-Bt gneiss (b). Their chemical composition is shown in (c) and (d). Color version is available online from <https://doi.org/10.2465/jmps.181115>.

The pelitic gneisses analyzed in this study were highly retrogressed/re-hydrated during fluid infiltration affecting the discordant granite. Given the distribution and textural-chemical features of the late-forming Bt, Crd, and Prh, at least two stages of re-hydration are recorded. The first stage of re-hydration is evidenced by

extensive secondary Bt and/or Crd after Grt porphyroblasts (Figs. 2c and 2f). A second stage of subsequent re-hydration appears to have occurred under relatively lower T conditions, as inferred from the abundant secondary Prh occurring as veins or lenses in the cleavages of the Bt in the discordant Bt granite (Fig. 2h), which could

Figure 6. REE pattern normalized by chondrite value for Grt from the Grt-Crd-Bt gneiss (a). Grt-Bt gneiss (b). See the text for the details. Chondrite values are after McDonough and Sun (1995).

have formed because of the retrograde effects of the host gneisses after granite crystallization had completed.

Interpretation of Zrn U-Pb ages

The discordant Bt granite contains sub- to euhedral or ovoid Zrns with the internal texture of structureless to core-rim structures or faintly oscillatory zone. The relic core and weak oscillatory domains obtained the scattered data between ~ 150 and 1700 Ma with relatively high-Th/U ratios (>0.1) (Figs. 3e and 3f). However, the clustered ages of ~ 105 Ma was calculated from the other unzoned euhedral Zrn grains which completely isolated from the old ones; therefore, it is constrained as timing of emplacement/crystallization of this discordant granite. The older data are similar to the detrital ages obtained from the pelitic gneisses (Figs. 3a, 3c, and 3e) which may propose that this Bt granite might be formed by the partial melting of the pelitic gneisses. However, it is still obscured and further study for bulk chemistry is required to tackle this problem.

Analyzed Zrns in the pelitic gneisses are mainly characterized by rounded termination morphology with

a sub- to euhedral or ovoid shape and internal features of core-rim zonation. The Zrn relic cores show obvious to faint oscillatory or sector zoning and scattered dates between ~ 130 and 2100 Ma with relatively high-Th/U ratios (mostly higher than 0.10) (Figs. 3a, 3c, and 3g). These results strongly indicate that these zoned Zrn cores preserve inherited detrital domains of magmatic origin. The youngest detrital Zrn cores shows a maximum depositional age pooled at ~ 130 Ma, approximately 50 Ma younger than previously reported by Suga et al. (2017). In contrast, the rim domains, which truncate or overgrow the relic cores, and the domains with a variety of internal zoning such as banded oscillatory and blurred zoning and unzoned to faint oscillatory zoning show concentrated ages of ~ 110 and 120 Ma, with Th/U ratios of 0.06–1.21 (Figs. 3b, 3d, and 3g). These distinct ages from detrital Zrns might indicate metamorphism timing, which is quite consistent with previous reports (Sakashima et al., 2003; Takagi and Arai, 2003; Dunkley et al., 2008; Maki et al., 2014; Osanai et al., 2017; Suga et al., 2017).

Zrn growth in relation to Grt: Constraining the high-temperature metamorphism timing

Based on internal texture and morphology, three metamorphic Zrn types can be categorized as follows: type 1: recrystallization (replacement), type 2: overgrowth, and type 3: newly crystallized Zrn following the definition of Rubatto (2017). Type 1 is characterized by an unzoned or faintly oscillatory zoned rim that truncates the inherited core (Figs. 7a and 7b) and blurred zoning (Figs. 7c and 7d) with the Th/U ratios of ~ 0.10 (Fig. 3g); its mean age is ~ 120 Ma (Fig. 3d). Type 2 is an ovoid metamorphic Zrn composed of an oscillatory zoned or homogenous core and a faint oscillatory zoned to unzoned rim (Figs. 7e and 7f) with low-Th/U ratios (<0.10) (Fig. 3g). Type 3 is sub- to euhedral Zrn with the oscillatory zoning (Figs. 7g and 7h) and more scattered Th/U ratios of 0.06–1.21 (Fig. 3g), similar to magmatic Zrn. The last two types of Zrn have a similar concordia age of ~ 110 Ma. The REE chemistries of the three types of metamorphic Zrns exhibit similar REE patterns with steep REE patterns normalized by chondrite values and HREE enrichment. However, type 1 and type 2 Zrns have lower LREE concentrations as compared to that of type 3, which are quite enriched in LREE (Fig. 4b).

As previously mentioned, the Grt-Crd-Bt gneiss (43007B) might have been subjected a similar metamorphic temperature condition to that of the Grt-Bt gneiss (430T01B); however, their Grt compositional zoning patterns show slight differences. In the Grt-Crd-Bt gneiss, the major elements (Mg, Mn) in the Grt compositional

Figure 7. Representative internal zoning and texture of three metamorphic Zrn types in the Grt-Bt gneiss (a)–(e), (g), and (h). Grt-Crd-Bt gneiss (f). Recrystallized Zrn with an oscillatory core is truncated by an unzoned to faintly oscillatory rim (a) and (b). Recrystallized Zrn with blurred zone (c) and (d). Faintly oscillatory zoned to unzoned dark-CL core overgrown by a structureless rim (e) and (f). Newly crystallized euhedral Zrn showing oscillatory zoning texture (g) and (h).

zoning pattern show a wide homogeneous core and a reverse-zoned rim strongly suggesting the effect of diffusion during HT metamorphism and retrograde cooling. Y and Ca showed truncation as well as embayments along the lower right side of the Grt grain (Fig. 5a) that might have resulted from Grt resorption (Spear et al., 1995). The peak metamorphic condition was possibly preserved in the mantle (lower left side), corresponding to the highest Mg and lowest Mn. However, the Grt-Bt gneiss still preserved the prograde pattern in association with the diffusion pattern at the margin. The core to rim increase in Ca and Mg and decrease in Mn and Fe follows a similar pattern to that of the Y zoning (Fig. 5b). All of these elements show truncation along the lower right side of the Grt grain which is interpreted as Grt resorption (Spear et al., 1995) or pinning of Grt grain boundaries adjacent to Qz (Pyle and Spear, 1999). The highest temperature condition was possibly recorded in the highest-Mg and lowest-Y rim of the Grt grain (Fig. 5b). Although the major element zoning pattern of the Grt is slightly different, the Grt in both samples show the same Y zoning pattern. The Y content decreases from the core to rim (Figs. 5a and 5b), typical prograde zoning for Grt (Pyle and Spear, 1999). Because the chemical behavior of the REE is similar to that of Y, the same Y zoning pattern of the Grt in the Grt-Crd-Bt gneiss and Grt-Bt gneiss strongly

suggests that the REE in both rock specimens preserve prograde chemical zoning.

To interpret the meaning of the two metamorphic ages of ~ 110 and ~ 120 Ma, the HREE distribution coefficients between the metamorphic Zrn and Grt were calculated for both the Grt-Bt gneiss and Grt-Crd-Bt gneiss. Though this method has been shown by the empirical and experimental works of Rubatto and Hermann (2007) and Taylor et al. (2015) as well as applied in many studies (e.g., Whitehouse and Platt, 2003), there is some uncertainty regarding the data required for use and it should be used with caution. Partitioning coefficients calculated by Rubatto and Hermann (2007) for Zrn and Grt from HP granulite-facies metapelites (800–1000 °C, 20 kbar, Ca-rich Grt) showed greater than unity values and a steep pattern from $^{Eu-Lu}D_{Zrn/Grt}$. This differs from the inferred partitioning of Whitehouse and Platt (2003) for granulite-facies metapelite (750–800 °C, 4–8.5 kbar; Ca-poor Grt) and Taylor et al. (2015) for UHT granulites (900–1000 °C at 7 kbar; Ca-free), which predict HREE partition coefficients that favor Grt over Zrn. However, all of them supposed that the HREE more favored in Grt with the temperature increasing. Their results also suggested that the chemistry of Grt, temperature, and pressure gave a great control on the partition coefficient, except the $^{Eu}D_{Zrn/Grt}$ which is approximately 1 in both data sets.

Figure 8. REE pattern of Zrn/Grt partitioning for metamorphic Zrn domain and Grt from the Grt-Crd-Bt gneiss and Grt-Bt gneiss.

Considering the low-Ca composition of the Grt in the current samples and published P - T conditions in zone D (6 kbar and 820 °C; Osanai et al., 1998), the partition coefficient values reported by Whitehouse and Platt (2003) and Taylor et al. (2015) are more suitable to apply to the current samples. For both samples, the $^{HREE}D_{Zrn/Grt}$ values between the ~ 110 Ma Zrns and Grt (core, mantle, and rim) in the Grt-Bt gneiss as well as the Grt (mantle and rim) in the Grt-Crd-Bt gneiss are significantly higher than 1 (Fig. 8), not typical equilibrium distribution values (Whitehouse and Platt, 2003; Taylor et al., 2015). Although the $D_{Zrn/Grt}$ shows nearly unity for Zrn-Grt_{core} in the Grt-Crd-Bt gneiss, the $D_{Zrn/Grt}$ for Eu is significantly greater than unity (Fig. 8); therefore, the equilibrium state is also not approached. Nevertheless, the equilibrium state seems to be reached for the ~ 120 Ma Zrn and Grt core (Grt-Bt gneiss) which has a nearly similar $D_{Zrn/Grt}$ value to those equilibrium ones reported by Whitehouse and Platt (2003) and Taylor et al. (2015), while the $D_{Zrn/Grt}$ steepens for the Grt mantle-rim corresponding to the decreased HREE toward the Grt rim (Fig. 8). As previously mentioned, Grt has been proposed to grow during prograde metamorphism; therefore, the equilibrium between the ~ 120 Ma Zrn and Grt core may indicate that the prograde metamorphic stage occurred at ~ 120 Ma when the Grt began to grow.

Moreover, the combination of internal textures, REE chemistry, and Zrn ages can also provide insight to infer the period of prograde, peak, and retrograde stages of the HT metamorphism. The ~ 120 Ma ages were obtained from the recrystallized Zrn rim only in the case of the Grt-Bt gneiss. Generally, this type of Zrn can form during the prograde stage because of the consumption of the protolith Zrn or during the later retrograde stage with the presence of fluid (Rubatto, 2017). The recrystallized

Zrn in this study showed the texture of blurred zones or truncated detrital cores without any resorption texture (Figs. 7a-7d) and the similarity in REE concentrations and patterns to those of the detrital cores (Fig. 4b). Thus, this may suggest that the ~ 120 Ma Zrns were recrystallized during the prograde stage, possibly occurring mainly at the expense of the precursor detrital Zrns. However, two types of Zrn provided a similar age result of ~ 110 Ma but differences in morphology, texture, and Th/U ratios. The ovoid Zrn with an unzoned overgrowth rim surrounding the oscillatory zoned or unzoned core, with low-Th/U ratios (<0.10), was observed in both pelitic gneisses (Figs. 7e and 7f). These unzoned Zrns are considered to have formed via the Zr-liberating reaction relating to the breakdown of Grt (e.g., Fraser et al., 1997; Whitehouse and Platt, 2003; Rubatto, 2017). Some ovoid Zrn occurrences in this study were observed near the Grt resorption part in both the Grt-Crd-Bt gneiss and Grt-Bt gneiss. Nevertheless, the newly crystallized Zrn has similar external and internal textures and Th/U ratios to magmatic Zrn as previously mentioned. Most of the REE patterns represent slight LREE enrichment compared to the recrystallized and overgrowth Zrns with strong HREE enrichment (Fig. 4b). Therefore, this type of Zrn might be regarded as crystallized from the melt during cooling. The presence of melt in pelitic gneisses can be inferred by the occurrence of migmatitic texture (Fig. 2a) and subhedral Crd (Fig. 2d), euhedral Pl (Figs. 2c and 2f) and Bt (Figs. 2c-2e).

Interestingly, the metamorphic Zrn age of ~ 120 Ma was obtained from only the Crd-free sample (430T01B) but it was absent from the Crd-bearing sample (43007B). Under thin section, Zrn is mainly found as an inclusion in secondary Crd after Grt in the Grt-Crd-Bt gneiss (Fig. 2c) while it is often included in Grt and exists in the

Figure 9. Summary diagram outlining the HT metamorphic history recorded by Zrn–Grt geochemistry and the Zrn U–Pb isotopic system in pelitic gneisses and a later granitic intrusion in the Higo metamorphic terrane (modified after Harley et al. 2007) (a). Corresponding to the *P*–*T* path suggested by Osanai et al. (2006) (b). Color version is available online from <https://doi.org/10.2465/jmps.181115>.

matrix in the Grt–Bt gneiss (Fig. 2g). Generally, Zrn only crystallizes when Zr saturation is reached or Zr solubility decreases; it is assumed that the Zr saturation point is controlled by the temperature and bulk chemistry (Kohn et al., 2015; Rubatto, 2017). The absence of prograde Zrn in the Crd-bearing sample might be interpreted as a result of the different Zr saturation points between the two samples. Because both pelitic gneisses underwent a similar metamorphic condition, it might have been caused by the different bulk chemistry. Under the metamorphic condi-

tion at 120 Ma, Zr saturation was reached in the Grt–Bt gneiss but not in the Grt–Crd–Bt gneiss.

In summary, these results suggest the following view of the HT metamorphism in the Higo metamorphic terrane (Fig. 9). The pelitic sedimentary protoliths were deposited until ~ 130 Ma as recorded by the youngest detrital Zrn age. The HT metamorphism occurred immediately after the deposition and the protolith Zrns recrystallized at ~ 120 Ma during the prograde stage. The pelitic gneisses might have undergone a peak metamorphic condition

between ~ 120 and ~ 110 Ma. The subsequent retrograde stage promoted new Zrn crystallization from melts and Grt breakdown at ~ 110 Ma, finally involving granitic intrusion into pelitic gneisses at ~ 105 Ma. Although the timing of the HT metamorphism in the Higo metamorphic terrane (~ 110–120 Ma) is consistent with that of previous studies (e.g., Sakashima et al., 2003; Takagi and Arai, 2003; Dunkley et al., 2008; Maki et al., 2014; Osanai et al., 2017; Suga et al., 2017), this study reconfirmed the rapid HT metamorphism framework from detailed observation of Zrn internal textures, REE chemistry, and U–Pb ages, providing critical information to understand the tectonic evolution of this terrane.

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SUPPLEMENTARY MATERIALS

Color version of Figures 2, 5, and 9 and Supplementary Tables S1–S4 are available online from <https://doi.org/10.2465/jmps.181115>.

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