Possible polymetamorphism and brine infiltration recorded in the garnet–sillimanite gneiss, Skallelvikshalsen, Lützow–Holm Complex, East Antarctica

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Chlorine-rich (>0.3 wt%Cl) biotite inclusions in the core of garnet porphyroblasts in the garnet–sillimanite (Grt–Sil) gneiss from Skallelvikshalsen, Lützow–Holm Complex (LHC), East Antarctica is estimated to be stable under >1.2 GPa, 820–850 °C, coexisted with granitic melt as suggested by the nanogranite/felsite inclusions. Rare occurrence of matrix biotite, in spite of the common occurrence of biotite as inclusions in garnet, suggests almost complete consumption of pre-existed matrix biotite during the prograde to peak metamorphism. Brine infiltration during prograde to peak metamorphism in Skallelvikshalsen is supported by Cl-rich scapolite described in previous studies. Brine infiltration and progress of continuous biotite-consuming melting reactions were probably responsible for elevating the Cl content of biotite in the studied sample.

In situ electron microprobe U–Th–Pb dating of monazite and the in situ laser ablation inductively coupled plasma mass spectrometry (LA–ICPMS) U–Pb dating of zircon in the Grt–Sil gneiss revealed that both monazite and zircon has the ‘older age population’ with ~ 650–580 Ma and the ‘younger age population’ with ~ 560–500 Ma. The REE and trace element pattern of one of the P-rich patches in the garnet core is different from the P-rich garnet rim. The isotope mapping of the same patch by LA–ICPMS revealed that the patch is also observed as a domain depleted in 51V, 89Y, 165Ho, 166Er, 167Tm, 172Yb, and 175Lu. Clear difference in 51V concentration between the patch and the garnet rim suggests that this patch is not a continuous part from the garnet rim, but is likely a relic of pre-existed garnet. Kyanite included in the patch suggests that the precursor rock was presumably a medium- to high-pressure type metamorphic rock. Presence of the older age population (~ 650–580 Ma) monazites in Skallelvikshalsen and Skallen also suggest that rocks in these areas experienced polymetamorphism, and resetting by the ~ 560–500 Ma metamorphic event was incomplete in these areas. Taking into account the presence of Cl-rich biotite inclusions in garnet, infiltration of brine accompanied by partial melting is one probable event that took place at ~ 560–500 Ma in the Skallelvikshalsen area, and part of the monazite possibly recrystallized by this brine infiltration.

Detailed microstructural observation using trace element mapping combined with detailed petrography especially focusing on the Cl-bearing minerals as a tracer of brines would become a powerful tool for better interpreting the results of monazite and zircon dating and for investigating the fluid-related crustal processes.

Keywords: Monazite, Zircon, Partial melting, Brine, Polymetamorphism

INTRODUCTION

Estimating the timing and duration of high-grade metamorphism through zircon and monazite dating can potentially place a very important constraint on the tectonic framework of lower crustal processes during continental collision events (e.g., Johnson and Harley, 2012; Rubatto et al., 2013; Korhonen et al., 2013). Recent advance in understanding the growth/recrystallization timing of zircon and monazite during metamorphic cycle, especially the roles of partial melting and fluid infiltration events in formation/dissolution/recrystallization of these minerals enable to reconstruct the lower crustal evolution in more detail (e.g., Rubatto et al. 2001; Rubatto, 2002; Harley and Kelly, 2007; Rubatto et al., 2013; Kawakami et al., 2013; 2014; Korhonen et al., 2013; Harley and Nandan kumar, 2014).

Increasing number of natural examples of chloride brines show that its generation and migration are important geologic processes in the lower crust, in addition to the CO2-rich fluid (Newton and Manning, 2010; Touret...

Mineralssuchasbiotite,hornblende,apatiteandscapolitecanincorporateClintheircrystalstructure.Amongtheseminerals,biotiteandapatiteareoftenusedtoestimatethe\(f_{H_2O}/f_{HCl}\)ratioofthecoexistingaqueousandbrinefluid(Munozandswenson,1981;SelbyandNesbitt,2000;PiccolithemandCandela,1994),althoughapatiteissometimesconsideredinertagainstfluid-relatedprocesses(PiccolithemandCandela,1994).ThemeritinusingtheCl-bearingmineralsasanindicatorofthechloridebrineactivityisthatthe\(P-T\)pathofthelowesthermalmassivemassconvergesandmeltsrockscanbecorrelatedwiththetimingsofthebrineexistence,utilizingthemicrostructureoftheCl-bearingminerals(e.g.,Satish–Kumaretal.,2006;Higashinoetal.,2013).

InordertolinkthepresenceofCl-richhydrousmineralstothefluid-relatedeventsandpartialmeltinginthelowercontinentalcrust,however,itisimportanttounderstandtheformationprocessesoftheseseminalminerals,especiallyinthepresenceofmelt.However,limitednumberofexperimentalstudyonClpartitioningisavailableonlyatlowpressureconditions(e.g.,IcenhowerandLondon,1997;Chevychelovetal.,2008).IcenhowerandLondon(1997)determinedthedistributioncoefficientofClbetweenbiotiteandgraniticmelttobe1–6at0.20GPaand640–680\(^\circ\)C.Ontheotherhand,experimentsat0.20GPaand–850\(^\circ\)CshowedthatorderoftheClpartitioningsoisfluid\(\gg\)phonolitemelt>biotite(Chevychelovetal.,2008).Sincexperimentalstudyandnaturalexamplesarestillscarcunderlowercrustalconditions,moreinformationonthemodeofoccurrenceandchemistryofCl-richhydrosmineralsinanaturalexamplesthatexperiencedpartialmeltingisneededinordertounderstandthepartitioningbehaviorofClbetweenhydrousmineralsandmelt,andtheformationconditionsofCl-richhydrosmineralsintheuppercrust.DetailedstudyofsuchnaturalexamplehashanadvantagebecausethebehaviorofzirconandmonazitecanbeexaminedandcorrelatedwiththepossiblyCl-bearingfluid(brine)infiltrationandmeltdownformation/crystallization.


**GEOLOGY OF SKALLEVIKSHALSEN, LÜTZOW–HOLM COMPLEX, EAST ANTARCTICA**


\[
\text{reaction staurolite} = \text{garnet} + \text{Al}_{2}\text{SiO}_5 + \text{spinel} + \text{H}_2\text{O}
\]


presenceof‘nanogranite’(Cesareetal.,2009)or‘felsite’
Inclusions in garnet porphyroblasts (Hiroi et al., 2014) and peak metamorphic condition of Skallevikshalsen were previously estimated to be 770–940 °C, 0.65–1.2 GPa from the garnet-biotite gneiss and 780–960 °C, 0.60–1.1 GPa from the garnet-two pyroxene gneiss (Yoshimura et al., 2004). The UHT condition exceeding 1000 °C was recently proposed, based on the occurrence of possible armalcolite pseudo-morph (Kawasaki et al., 2013).

Previous SHRIMP ages from zircon separates concentrate around 550–530 Ma, and were interpreted as timing of peak metamorphism in the LHC (e.g., Shiraishi et al., 1994). However, from the in situ SHRIMP analyses of zircon inclusions in garnet porphyroblasts, Dunkley (2007) revealed a stage of U–rich zircon growth at ~600 Ma, with flat HREE–MREE profiles indicating prograde growth in the presence of garnet (Dunkley, 2007). Therefore, ~600–570 Ma is considered to be the timing of prograde to peak metamorphism, and 550–530 Ma to be the retrograde zircon growth stage (Dunkley, 2007). Kato (2013) dated zircon from syenite dyke and associated charnockite that postdate the peak metamorphism at Skallevikshalsen, and obtained the age of 550–530 Ma. This is also consistent with the peak metamorphism and penetrative deformation in the Skallevikshalsen area to be earlier than 550–530 Ma (Kato, 2013).

**ANALYTICAL METHODS**

The X-ray mapping of the garnet grains were obtained by a JEOL JXA8800M at the National Institute of Polar Research (NIPR) and by a JEOL superprobe JXA–8105 at Kyoto University. Analytical conditions for X-ray mapping were 15.0 kV acceleration voltage, 400–800 nA probe current with focused beam to defocused beam up to 5 µm diameter. Minerals were quantitatively analyzed by the JEOL superprobe JXA–8105 at Kyoto University. Analytical conditions were 15.0 kV acceleration voltage, 10 nA probe current with probe diameter of 3 µm. The counting time for the peak and backgrounds were 30 s and 15 s for Cl, 60 s and 30 s for F, and 10 s and 5 s for other elements, respectively. Natural and synthetic minerals were used for standards, and ZAF correction was applied. Analytical condition of rutile for Zr–rutile geothermometry followed that recommended by Zack et al. (2004). Counting error was about ±2% for Zr, which yielded ignorable small (±2°C) variation in the result of Zr–in-rutile geothermometry. Al₂SiO₅ minerals and fluid inclusions were identified by Raman spectroscopy at NIPR (JASCO NRS 1000) and Kyoto University (JASCO NRS 3100).

The electron microprobe (EMP) dating and REE analyses of monazite were carried out using a JEOL JXA–8200 at NIPR. Detailed analytical technique is given in Hokada and Motoyoshi (2006).

In situ laser ablation inductively coupled plasma mass spectrometry (LA–ICPMS) analyses of rare earth elements (REE) and trace element concentrations of garnet were performed using an iCAP-Qc quadrupole ICPMS coupled to a NWR–193 laser-ablation system at Kyoto University. Detailed analytical conditions are reported in Higashino et al. (2015). In situ isotope mapping on a thin section sample was performed using the same system. Data processing was done using a software iQuant2. Analytical conditions are given in Table S1 (Tables S1 is available online from http://doi.org/10.2465/jmps.150812).

In situ zircon U–Pb dating on thin section samples by LA–ICPMS was carried out using a Nu Plasma II HR–
MC–ICPMS coupled with a NWR–193 laser-ablation system at Kyoto University. Analytical conditions are given in Table S2 (Tables S2 is available online from http://doi.org/10.2465/jmps.150812). Backscattered electron (BSE) and cathodoluminescence (CL) images were obtained prior to the analyses to identify spot positions, overlapping multiple growth zones, grain edges, cracks or damaged zircon grains. Data were processed and plotted using Isoplot 4.15 (Ludwig, 2012). The CL images were obtained using a JEOL JXA-8105 superprobe equipped with Hamamatsu Photonics high voltage power supply C9525 and photon counting unit C9744 at Kyoto University. Analytical condition for the CL mapping of zircon was 15.0 kV acceleration voltage, 1 nA beam current, focused beam, and dwell time of 1 milliseconds.

**SAMPLE DESCRIPTION**

The sample used in this study is a khondalitic garnet-sillimanite gneiss (sample TK2002011804) collected during the summer season of the 44th Japan Antarctic Research Expedition (JARE44; 2002–2003). The sample mainly consists of garnet (~ 5 mm in diameter), sillimanite, K-feldspar, plagioclase and quartz with minor biotite, rutile, ilmenite, zircon, monazite and pyrite (Kawakami and Motoyoshi, 2004; Kawakami et al., 2006; Kawakami and Hokada, 2010). It is noteworthy that the rock matrix consists of garnet (~ 5 mm in diameter), sillimanite, biotite needles, plagioclase and quartz with minor biotite, rutile, ilmenite, zircon, monazite and pyrite (Kawakami and Motoyoshi, 2004; Kawakami et al., 2006; Kawakami and Hokada, 2010). Another example of a large nanogranite/felsite inclusion that consists of intergrowth of plagioclase (An26–28) + K-feldspar + quartz + muscovite + minor biotite (Fig. 6a) is newly found in this study. Muscovite and chlorite are formed between the intergrowth and host garnet, and cuspat or spear-shaped replacement by muscovite is developed from the edges of the nanogranite/felsite inclusion into the host garnet (e.g., Hiroi et al., 2014). Post entrapment chemical re-equilibrium between host garnet and nanogranite/felsite inclusion is inferred from the halo of Fe and Mg developed around the inclusion (Fig. 6a). Chlorine content of biotite in the intergrowth is lower than that included in garnet as a single mineral. Finer-grained nanogranite/felsite inclusions with the cuspat or spear-shaped replacement microstructure are abundantly included in the garnet core without any internal alignment (Figs. 6b–6c). Apatite included in the garnet core is mostly Cl-bearing fluorapatite (up to 1.88 wt% Cl, 0.26 a.p.f.u. under O = 12.5, n=18) (Table 1).

On the other hand, the garnet rim includes sillimanite needles, plagioclase, K-feldspar, quartz, zircon and coarse-grained monazite (<147 µm). K-feldspar is included in the rim or at the boundary between the core and the rim (Kawakami and Hokada, 2010).

Matrix biotite shows lower Cl contents (0.06–0.13 wt% Cl) than the inclusion biotite. TiO₂ content varies from 2.0 to 4.9 wt% with one exception of 6.0 wt%, the average being 3.8 wt% (n = 14) (Table 1). Rare matrix apatite is fluorapatite, and its Cl content is below detection limit (n = 1) (Table 1).

Trace element and REE analyses by LA–ICPMS revealed that garnet core is lower in P and Cr, and higher in HREE contents than the rim (Fig. 7 and Table 2: Table 2 is available online from http://doi.org/10.2465/jmps.150812). P-rich patches are present in garnet cores as revealed by EMP mapping (Figs. 2 and 5; Kawakami and Hokada, 2010). Two of these patches are analyzed quantitatively for trace elements and REE. As a result, one patch from garnet E (Fig. 5) showed similar trace element and REE composition with the garnet rim, and the other patch from garnet A (Fig. 2) showed lower Cr and higher Y and HREE concentrations than the rim (Fig. 7). In situ isotope mapping of this P–rich patch from garnet A by LA–ICPMS revealed that it is also observed as a domain depleted in ⁵¹V, ⁶⁸Y, ¹⁶⁶Ho, ¹⁶⁶Er, ¹⁶⁹Tm, ¹⁷²Yb, and ¹⁷⁵Lu (Fig. 2).
Zirconium-in-rutile geothermometry

Zr-in-rutile geothermometer (Zack et al., 2004; Ferry and Watson, 2007; Tomkins et al., 2007) was applied to rutile crystals included in the garnets A, B, C and E. Rutile inclusions are not accompanied with zircon or quartz, and these minerals are separately included in the core of garnet porphyroblasts. Rutile grains coexisting with ilmenite are not used, because activity of TiO$_2$ may be lower than unity and difficult to estimate. Zr-in-rutile geothermometer by Zack et al. (2004) gave 960–1025 °C for most grains. The result by Tomkins et al. (2007) is preferred in this study because it is experimentally calibrated and pressure effect is taken into account. Zr-in-rutile geothermometer by Ferry and Watson (2007) gave slightly lower (~ 10 °C) temperature estimate than Tomkins et al. (2007), suggesting the reliability of the estimated values.

Most of the grains gave Zr concentration of ~ 2200–3300 ppm, with the average of ~ 2560 ppm (n = 43, averaged excluding the data below 1200 ppm). Under the coexistence of kyanite, and using the Al$_2$SiO$_5$ triple point and Sil/Ky transition of Pattison (1992), these values corresponds to pressures above ~ 1.1 GPa and temperatures above ~ 840 °C. In Figures 2–5, temperature value calculated using the pressure of 1.2 GPa is shown. Some rutile grains included together with other minerals, such as plagioclase or biotite + staurolite + kyanite + spinel, gave lower temperature estimates. In the latter case, biotite coexisting with rutile showed apparently lower Cl content than other biotite grains included independently in the same garnet grain (Fig. 5b). A rutile grain included in garnet together with plagioclase (An43–45) gave 795–802 °C.

EMP monazite dating

The monazite grains dated in situ by EMP are from the rock matrix (3 grains), included in the garnet core (5 grains including one grain at the core/rim boundary),...
or included in the garnet rim (2 grains). Inclusion monazite grains dated are from garnets A, B, C and E described in Kawakami and Hokada (2010).

Back-scattered electron image of monazite grains and result of EMP monazite dating are summarized in Figure 8, and the analytical results are reported in Table S3 (Tables S3 is available online from http://doi.org/10.2465/jmps.150812). Y2O3 was accidentally not analyzed. Five grains that occur as garnet inclusion and matrix phase yielded weighted average age (±2σ error) of 564 ± 42 Ma (n = 5, MSWD = 0.42), 549 ± 11 Ma (n = 42, MSWD = 0.28), 538 ± 31 Ma (n = 9, MSWD = 0.11), 520 ± 28 Ma (n = 9, MSWD = 0.26), and 516 ± 27 Ma (n = 7, MSWD = 0.45). On the other hand, one grain included at the rim of garnet C yielded weighted mean age of 604 ± 13 Ma (n = 34, MSWD = 0.29). Other grains did not give sufficient number of analyses to calculate the weighted mean age from a single grain. Compared to the monazite with older age (604 ± 13 Ma), the grains with younger ages tend to be lower in MREE and U concentration, and higher in Th concentration and Th/U ratio (18–73) (Fig. 9; Table S3).

LA–ICPMS U–PB ZIRCON DATING

Zircon grains dated in situ by the LA–ICPMS are mostly from the matrix (n = 15, major axis = 24–104 µm), and 4 grains are included in garnet (major axis = 15–50 µm). Most of the zircon grains are zoned, and tend to have inherited cores showing oscillatory zoning with bright and dark CL zones (Figs. 10a–10d). It is commonly overgrown by gray to dark CL rims (Figs. 10a-10d).

Inherited cores commonly yield discordant data (Figs. 10a–10c and 10e). On the other hand, zircon rims commonly yield concordant data (Figs. 10a, 10c–10d, and 10f). Gray to dark CL rims of the matrix zircon tend to show 206Pb/238U ages of ~ 530–510 Ma (Figs. 10a and 10c; Table 3: Table 3 is available online from http://doi.org/10.2465/jmps.150812). Rim of one zircon inclusion in garnet and some of the matrix zircon rims show 206Pb/238U ages of ~ 595–580 Ma (Fig. 10d). Zircon domain with ~ 530–510 Ma is not found as garnet inclusions in the studied sample. Zircon domains with ~ 530–510 Ma age tend to show higher Th, lower U contents and slightly higher Th/U ratio (0.02–0.30) than those with ~ 595–580 Ma age (0.01–0.10). A concordia diagram of all analyses and the probability density plot constructed for the concordant data are shown in Figures 10c–10g. Age unmixing based on the algorithm of Sambridge and Compston (1994) assuming two components for concordant zircon rim ages yielded 595 ± 11 Ma and 524 ± 7 Ma (Fig. 10g).

DISCUSSION

Pressure–temperature condition of Cl-rich biotite formation

Since the homogenization of rutile grain in terms of Zr content during high-temperature metamorphism cannot be ruled out, the highest temperature estimate obtained from the Zr-in-rutile thermometer should be considered as representing the minimum temperature attained. Therefore, the temperature estimate of 820–850 °C obtained...
Table 1. Representative electron microprobe analyses of biotite and apatite

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### Analytical Conditions

- **Analysis no.**
- **Inclusion in**
  - GrtB
  - GrtC
  - GrtE

### Mode of occurrence

- **SiO₂**: 37.33 ± 1.31
- **TiO₂**: 4.08 ± 0.40
- **Al₂O₃**: 22.72 ± 2.27
- **Fe₂O₃**: 9.93 ± 1.14
- **ZnO**: not determined
- **MnO**: 0.00 ± 0.00
- **MgO**: 15.54 ± 1.56
- **CaO**: 0.00 ± 0.00
- **BaO**: 0.25 ± 0.03
- **Na₂O**: 0.20 ± 0.02
- **K₂O**: 9.04 ± 0.93
- **P₂O₅**: n.d.
- **F**: 0.09 ± 0.01
- **Cl**: 0.31 ± 0.03
- **O-E**: 0.04 ± 0.01
- **O-Cl**: 0.07 ± 0.01

### Total Cation

- **Mg/(Fe₃⁺+Mg)**: 0.74 ± 0.06
- **log(fCO²/fH²O)** of fluid: 3.78 ± 0.56
- **log(fCO²/fH₂)** of fluid: 4.93 ± 0.46
- **Temperature (°C)**: 850 ± 100

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*1 Based on Munoz (1992) for biotite, and Piccoli and Candela (1994) for apatite.
*2 Temperature used in calculating log(fCO²/fH₂O) of the fluid coexisted with biotite and/or apatite. Temperature estimate is based on Zr-in rutile thermometer. See text for details.
*3 Analyzed in NIPR. See Kawakami and Motoyoshi (2004) for analytical conditions.
*4 In nanogranite-like inclusion in GrtE.

n.d., not determined.
from the rutile included in the garnet core is regarded as minimum temperature for the garnet core formation. Thus, Cl–rich biotite and kyanite included in the garnet core were stable under pressures >1.2 GPa and temperatures >850 °C, and coexisted with granitic melt as suggested by the presence of nanogranite/felsite inclusion in the garnet core (Fig. 6). Peak metamorphic temperature of 848 ± 55 °C reported from Skallen (Satish–Kumar and Wada, 2000) adjacent to Skallevikshalsen is quite similar to our temperature estimate. Kawasaki et al. (2013) also reported the temperature estimate by Zr–rutile thermometer, which gave mostly consistent results with this study. The pressure condition under the kyanite stability field is previously estimated to be slightly lower than 1.2 GPa (e.g., Yoshimura et al., 2004; Kawakami and Motoyoshi, 2004), because of the lower-temperature estimate than the present study.

**Formation mechanism of Cl-enriched biotite included in garnet**

Chlorine-rich biotite is exclusively preserved as inclusions in garnet (Figs. 2–5), suggesting prograde to peak metamorphic origin. Therefore, the process such as retrograde formation of biotite to preferentially consume H2O in the fluid phase, resulting in the increase of the Cl concentration in the remaining fluid (e.g., Markl and Bucher, 1998) is not likely in the present case. Instead, partial melting process is clearly related to the present case, because the nanogranite/felsite inclusions do not represent the melt that infiltrated after the garnet rim growth. Progress of continuous dehydration melting of biotite to form peritectic garnet would increase the Cl concentration in biotite by consuming originally Cl-poorer biotite, if Cl is preferentially incorporated into biotite. Scarcity of the matrix biotite, in spite of the common occurrence of bio-

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**Figure 6.** (a) X-ray elemental maps of a nanogranite/felsite inclusion in the garnet C (Fig. 4), in terms of Ca, Si, Cl, Mg, K, Al, Na and Fe. Biotite (Bt), muscovite (Ms), K-feldspar (Kfs), quartz (Qtz), plagioclase (Pl) and chlorite (Chl) are shown. (b) Back scattered electron (BSE) image of the garnet core containing the nanogranite/felsite inclusion shown in (a). Fine-grained nanogranite/felsite inclusions are also included in the garnet core. (c) Enlargement of fine-grained nanogranite/felsite inclusions in the garnet core. BSE image. Color version of Figure 6 is available online from http://doi.org/10.2465/jmps.150812.
biotite as inclusions in garnet, suggests almost complete consumption of pre-existing biotite during the prograde to peak metamorphism in the studied sample.

Experimental studies showed that partial melting triggered by the infiltration of H$_2$O fluid can also produce peritectic garnet (Weinberg and Hasalová, 2015). Therefore, the processes such as selective consumption of H$_2$O in the fluid into the melt to increase the Cl concentration in the remaining fluid, or the infiltration of already Cl-rich fluid, such as brines, to trigger partial melting (Aranovich et al., 2013), may also account for the presence of Cl-rich biotite coexisted with melt. High TiO$_2$ contents of the Cl-rich biotite inclusions in the garnet core support their formation during the prograde to peak metamorphism.

However, experimental study on the partitioning of Cl between biotite and melt is only available at low-pressure condition at 0.2 GPa (Icenhower and London, 1997; Chevychelov et al., 2008) and thus partitioning behavior of Cl in high-pressure condition is not well understood. On the other hand, partitioning of Cl between melt and coexisting H$_2$O fluid is better constrained at higher-pressure up to 0.6 GPa (Shinohara et al., 1989; Piccoli and Candela, 1994), and a remarkable negative pressure dependence of D$_{Cl}$(melt/fluid) [= (Cl concentration in melt) / (Cl concentration in H$_2$O fluid)] is reported (Shinohara et al., 1989). Chlorine is significantly partitioned into fluid, if present, than the coexisting melt at higher pressures.

If we assume D$_{Cl}$(Bt/melt) of ~ 1 to 6 as observed in 0.2 GPa for a granitic melt (Icenhower and London, 1997) to be applicable to high-pressure conditions observed in the Skallevikshalsen sample, the melt coexisted with Cl-rich biotite (~ 0.40 wt% Cl; Table 1) should have 0.07 wt% < Cl < 0.40 wt%. On the other hand,apatite included in the garnet shows X$_{Cl}$(apatite) [= Cl/(Cl + F + OH)] of 0.12–0.25 (Table 1), corresponding to ~ 0.45–0.55 wt% Cl in the rhyolitic melt at 0.2 GPa (Webster et al., 2009). These are not unrealistic values for natural rhyolite melts. Therefore, keeping in mind that high-pressure behavior of Cl may be different from low-pressure as 0.2 GPa, we can tentatively consider that Cl-rich biotite and apatite in the studied sample.
sample would have coexist with melt, and were possibly produced through partial melting consuming biotite.

Whether external input of Cl is required to attain ~0.40 wt% Cl in biotite or not depends on initial Cl content in biotite and degree of partial melting that consumes biotite. However, in Skallen and Skallevikshalsen, Cl-rich scapolite in marbles has been considered as a result of hypersaline fluid that was present during scapolite formation (Satish-Kumar et al., 2006; 2008b). Since Cl-rich scapolite is stable under the peak P-T condition (>1.2 GPa, 820–850 °C) of Skallevikshalsen (e.g., Filiberto et al., 2014), infiltration of brine during prograde to peak metamorphic stage is highly likely (e.g., Satish-Kumar et al., 2008b). Therefore, brine infiltration into the pelitic lithology, followed by the partial melting process discussed above, is one of the possible processes to have elevated the Cl content in biotite in the studied sample. Source of such brine is still not understood, and should be solved in the future studies.

**EMP monazite and LA-ICPMS U-Pb zircon ages**

The EMP monazite ages of 604 ± 13 Ma and 564 ± 42 Ma to 516 ± 27 Ma obtained from this study corresponds well with two age populations of ~ 650–580 Ma and ~ 560–500 Ma previously reported from Skallen (Hokada and Motoyoshi, 2006) next to Skallevikshalsen (Fig. 1). The older ~ 650–580 Ma monazite from Skallen are reported to be relatively enriched in Nd, Sm, Gd, Dy (M-HREE) and depleted in Si, Ca and Th compared with the younger ones (Hokada and Motoyoshi, 2006). These chemical characteristics observed in these older and younger monazite populations are similar to those observed in Skallevikshalsen (Fig. 9).

The younger age population of monazite corresponds well with the ~ 550–520 Ma SHRIMP zircon age observed throughout the LHC (Shiraishi et al., 1994; 2008). In situ LA-ICPMS U-Pb zircon dating by this study (Fig. 10; Table 3) yielded two Neoproterozoic to Early Cambrian age populations, 595±11 Ma and 524±7 Ma, from zircon from Skallevikshalsen (Fig. 10g). These age populations correspond well with the older and younger age populations of monazite. Therefore, the term ‘older age population’ and ‘younger age population’ are used both for monazite and zircon hereafter to represent grains or domains yielding the ages of ~ 650–580 Ma and ~ 560–500 Ma. Chemical characteristics of the younger age population monazites, such as high Th/U ratio and high Th content than the older age population monazites, are also observed in zircon as well. That is, the younger age population zircons show the higher Th/U ratio of <0.30 and higher Th content than the older age population zircons (Table 3).

Hokada and Motoyoshi (2006) interpreted the older age population monazite to have grown in M-HREE rich condition such as garnet-free condition, and thus predated peak metamorphism. On the other hand, Dunkley (2007) revealed a stage of U-rich zircon growth at ~ 600 Ma (older age population zircons), with flat M-HREE profiles indicating prograde growth in the presence of garnet (Dunkley, 2007). He interpreted 550–530 Ma zircon (younger age population zircon) to be the retrograde product (Dunkley, 2007). It is also confirmed in the pres-
ent study that older age population zircon occur both as an inclusion in garnet and as the matrix phase, whereas younger age population zircons do not occur as inclusions in garnet. However, linking zircon growth stages using REE pattern needs caution, because under open-system condition, host-rock mineralogy such as the presence of garnet can have no chemical impact on zircon chemistry (Harley and Nandakumar, 2014). In order to account for the available observation, it is also possible to assume zircon growth timings in two separate metamorphic events; zircon growth at ~ 600 Ma metamorphism (older age population zircon) accompanied garnet growth, and that during 550–530 Ma metamorphism (younger age population zircon) postdated garnet growth. Since the presence of nanogranite/felsite inclusion in the garnet (Fig. 6) is a clear evidence of garnet coexisted with granitic melt, zircon probably experienced dissolution in the melt during the prograde garnet growth stage, and crystallized during the retrograde, melt crystallization stage.

Different from zircon, the younger age population monazites are included in garnet (Fig. 8). The chemical feature of the younger age population monazite having lower MREE concentrations than the older age population monazite (Fig. 9) is consistent with coexistence with garnet. This implies that garnet growth and most of the formation/recrystallization of the younger age population monazite predated recrystallization of zircon during the 550–530 Ma metamorphism. Therefore, it is likely that the older age population monazite recrystallized to form the younger age population monazite, possibly through the dissolution–reprecipitation mechanism, during the prograde to peak stage of metamorphism at 560–500 Ma under the presence of garnet and melt as suggested by the nanogranite/felsite inclusions in the monazite–bearing garnet. This was presumably followed by the zircon growth during the melt crystallization and partial resorption of garnet. Some of the young-aged monazite may be

Figure 9. (a) REE, Th and U in monazite as a function of apparent U-Th-Pb monazite ages determined by the EMP. Note that the older monazite has higher Gd$_2$O$_3$, Sm$_2$O$_3$, and UO$_2$ contents, and lower ThO$_2$ content than the younger monazite. (b) A plot showing the correlation between Gd$_2$O$_3$ contents in monazite and apparent U-Th-Pb monazite ages. Note that the older monazite is richer in Gd$_2$O$_3$. (c) A plot showing the correlation between the Th/U ratios of monazite vs apparent U-Th-Pb monazite ages. Note that the older monazite has low Th/U ratio. (b)–(c) ‘Garnet A’ represent monazite analyses from the grains (f) and (h) in Figure 8, ‘Garnet B’ represents those from the grains (a), (b), and (e), ‘Garnet C’ represents those from the grain (g), and ‘Garnet E’ represents those from the grains (c), (d), (i), and (j). Color version of Figure 9 is available online from http://doi.org/10.2465/jmps.150812.
affected by the diffusion of Pb and partial resetting of the U-Th-Pb system under high-temperature conditions (e.g., Suzuki et al., 1994; Kawakami and Suzuki, 2011).

Possible polymetamorphism in the Skallen and Skallevikshalsen areas of the LHC

The P-rich patch in the garnet core of the garnet-sillimanite gneiss from Skallevikshalsen was previously interpreted as three-dimensionally continuous to the P-rich rim, and appears in the core because of the cutting effect (Kawakami and Hokada, 2010). The present study revealed that one of the P-rich patches shows high Cr and HREE concentrations than the P-rich garnet rim. Because the trace element concentrations of the P-rich patch are, for most elements, between that of surrounding P-poor garnet core and that of the P-rich rim (Fig. 7), it is possible to interpret that the P-rich patch is continuous to the rim as previously considered, and that the diffuse nature of the patch observed for some trivalent elements (Figs. 2d-2f) is a result of diffusion after the patch and the garnet rim growths.

However, an isotope map by $^{51}$V (Fig. 2d) conflicts with this interpretation, because the garnet rim and the patch has distinctly different $^{51}$V concentration. Absence of diffusive zoning in $^{51}$V at the core/rim boundary of garnet also suggests that the core and rim did not have significant difference in $^{51}$V concentration as observed between the $^{51}$V-poor patch and the garnet core. Therefore, there is a chance to consider that the P-rich ($^{51}$V-poor) patch is not continuous with the garnet rim, and is originally different in composition. That is, another probable interpretation is that this part represents a relic of the preexisted garnet. Since the P-rich patch includes kyanite, the precursor rock can be a medium- to high-pressure type metamorphic rock. Presence of the older age population monazites in Skallevikshalsen and Skallen (Hokada and Motoyoshi, 2006), in addition to the possible relic garnet in Skallevikshalsen, suggest that rocks in these areas experienced polymetamorphism, and resetting by the ~ 560–500 Ma metamorphic event was incomplete.

Taking into account the presence of Cl-rich biotite inclusions in garnet, infiltration of brine that triggered partial melting is one probable event that took place during ~ 560–500 Ma metamorphism in the Skallevikshalsen area. It is known from experimental study that CaCl$_2$-bearing fluid can recrystallize monazite through dissolution-precipitation mechanism and make newly grown monazite Th-richer (Seydoux-Guillaume et al., 2002). Although the effect of the partial melting to the fluid-monazite reaction is unclear in the present case, Th-richer composition of the younger age population monazite may be explained if the infiltration of brine is considered. Therefore, possibility of polymetamorphism and role of fluids including brines should be carefully considered in the future study of the LHC.
Polymetamorphism and brine infiltration in the Lützow-Holm Complex

Recognizing the timing of fluid release/infiltration during metamorphism and crystallization of plutonic bodies, and understanding the composition of fluids is of great importance especially because brines are able to recrystallize datable minerals such as monazite and rarely zircon (e.g., Seydoux-Guillaume et al., 2002; Kawakami and Suzuki, 2011; Higashino et al., 2013), which makes the interpretation of geochronological data more complex (e.g., Kawakami et al., 2014). More reliable interpretation of zircon and monazite dating in high-grade metamorphic rocks is required to understand the rate and duration of crustal processes. As presented in this study, detailed microstructural observation using trace element mapping combined with detailed petrography especially focusing on the Cl-bearing minerals as a tracer of Cl-rich fluids would become a powerful tool in the future studies.

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SUPPLEMENTARY MATERIALS

Tables 2 and 3, Supplementary Tables S1–S3, and color version of Figures 6, 7, 9, and 10 are available online from http://doi.org/10.2465/jmps.150812.

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