Geochemistry and magmatic zircon U-Pb dating of amphibolite blocks in the Omi serpentinite mélange, north central Japan: Possible subduction of the Cambrian oceanic crust

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Early Paleozoic serpentinite melanges in Japan preserve the oldest high-P metamorphic rocks in the circum-Pacific orogenic belt. To understand the tectonic regime at the subduction initiation of the proto-Japan convergent plate boundary, whole-rock geochemistry, and zircon U-Pb geochronology were investigated for amphibolite blocks in the Omi serpentinite mélange, central Japan. The studied amphibolites from two different localities have the mineral assemblage of albite + clinozoisite + amphibole ± rutile ± titanite, which characterize epidote-amphibolite facies metamorphism. Whole-rock trace element concentrations of the amphibolites suggest that gabbroic protoliths formed possibly in an oceanic setting. The zircon U-Pb weighted mean ages obtained from two amphibolite samples indicate that the protolith was formed in the Cambrian. The protolith ages of the studied amphibolites are comparable with those of reported Early Paleozoic ophiolite and high-pressure rocks in Paleozoic serpentinite melanges in Japan. This fact implies that the young hot oceanic crust was subducting into the East Asian convergent plate margin during the Cambrian.

Keywords: Serpentinite mélange, Amphibolite, Zircon U-Pb dating, Cambrian

INTRODUCTION

The Japanese islands comprise part of the circum-Pacific orogenic belt, where the continuous accretion of oceanic materials since Early Paleozoic has created a nappe pile, where valuable information of ancient subduction dynamics is preserved. For example, Late Paleozoic high-P blueschist metamorphic rocks (350–280 Ma Renge metamorphic rocks) underthrust below an Early Paleozoic ophiolitic complex (Oeyama ophiolite), and they represent a subducted oceanic plate and a Paleozoic supra-subduction zone complex, respectively (Ishiwatari et al., 2003). In addition to the Late Paleozoic high-P metamorphic rocks, older 480–400 Ma amphibolites with high-P assemblage are closely associated with Early Paleozoic serpentinite mélanges (Tsujimori, 2010). Tsujimori (2010) referred to them as the Kitomyo-Fuko Pass metamorphic rocks and regarded them as the oldest high-P metamorphic rocks in the circum-Pacific orogenic belt. The petrology and geochronology of the oldest metamorphic rocks in Japan will provide crucial clues for understanding the subduction initiation and tectonic environment in the Early Paleozoic East Asian continental margin.

Recent zircon geochronological studies have determined the possible protolith ages of Early Paleozoic metamorphic and ophiolitic rocks in Japan (Osanai et al., 2014; Kimura and Hayasaka, 2019). Serpentinite mélange enclosing the tectonic blocks of various lithologies is distributed in the Omi area, northern central Japan (Fig. 1a). In the Omi serpentinite mélange, the U-Pb ages of hydrothermal-origin zircon in jadeitite indicate that the subduction initiation occurred in the Cambrian (Kunugiza and Goto, 2010; Kunugiza et al., 2017). However, the geochronological information of amphibolite blocks in the Omi serpentinite mélange has been poorly characterized so far. In this study, we performed whole-rock geochemical analysis and zircon U-Pb dating for the amphib-
oli"tes to infer the tectonic environment in the Paleozoic East Asian continental margins.

GEOLOGICAL OUTLINE

The Omi area comprises Paleozoic accretionary complexes of a broad age range; the Mushikawa, Himekawa–Omi, and Omi serpentinite mélange complexes from east to west in younging order (Fig. 1a; Nagamori et al., 2010). These complexes are equivalent to the Maizuru (Permian), Akiyoshi (Carboniferous), and Oeyama (Early Paleozoic) + Renge (Late Paleozoic) belts in the Inner Zone of southwest Japan, respectively. These Paleozoic complexes are covered with Jurassic and Cretaceous sediments. The Omi serpentinite mélange is a tectonic mélange composed mainly of mafic and pelitic schists and amphibolite blocks with a serpentinite matrix (Fig. 1b). Basic and pelitic schists experienced high–P metamorphism (Banno, 1958; Shinji and Tsujimori, 2019; Shinji et al., 2019), and eclogite facies metabasite (e.g., Tsujimori, 2002) and jadeitite (e.g., Tsujimori and Harlow, 2017) also occur as tectonic blocks. The K–Ar ages of 339–285 Ma are recorded in phengitic muscovites from pelitic schists (Kunugiza et al., 2004). The Late Paleozoic HP schists are divided into the Eclogitic Unit with the occurrence of glaucophane–bearing eclogite and blueschist in hosted in paragonite–bearing pelitic schists and the Non-eclogitic Unit with dominant biotite–bearing pelitic schists (e.g., Tsujimori, 2002). Recent boron isotope study for the serpentinite found a difference in isotope signature among the two units (Yamada et al., 2019). The Omi serpentinites contain antigorite, tremolite, talc, carbonate, and metamorphic olivine (Yokoyama, 1985). In the Kotaki and Happo–One areas nearby the Omi area, the equivalent serpentinites locally experienced contact metamorphism (Nozaka, 2003; Machi and Ishiwatari, 2010). The protoliths of these serpentinites are harzburgite to dunite with rare chromitites (Yokoyama, 1985; Tsujimori, 2004; Machi and Ishiwatari, 2010). Machi and Ishiwatari (2010) suggested that these ultramafic rocks are similar to those reported from the Oeyama ophiolite and are derived from the residual mantle in Early Paleozoic forearc regions. The studied amphibolites occur as several blocks of 100’s of meters in dimension in the serpentinite matrix. Hornblende in the amphibolites yielded the K–Ar ages of 336 ± 13 and 370 ± 12 Ma (Shibata, 1981).

SAMPLE DESCRIPTION

In this study, we collected samples from two large am-

Figure 1. (a) Geotectonic map of the Omi area (after Nagamori et al., 2010). The rectangle indicates the area shown in (b). (b) Geological map along the Omi River (after Matsumoto et al., 2011) and sample locations of this study.
phibolite blocks from the Omi serpentinite mélange (Fig. 1b). These blocks are marked as ‘metagabbro’ in the geological maps provided by the previous studies (Banno, 1958; Nakamizu et al., 1989; Matsumoto et al., 2011; Nagamori et al., 2010). Both samples (MS20-01 and MS20-03) are coarse-grained and exhibit strong gneissic fabric, which is defined by the foliation of prismatic plagioclase and amphibole and their compositional banding (Figs. 2a and 2b). MS20-01 (from Kanayamadani) consists of green-color amphibole, clinozoisite, and albite with a minor amount of magnetite and titanite (Fig. 2c). The amphibole crystals in MS20-01 are classified as magnesiohornblende to tremolite-actinolite based on its Si = 7.30–7.79 p.f.u. (per formula unit calculated on the basis of O = 23) and Mg# \([=\frac{Mg}{(Mg + Fe^{2+})}]\) of 0.87–0.93.

MS20-03 (from Shimizukura) consists of green-color amphibole, clinozoisite, and albite with a minor amount of magnetite and rutile (Fig. 2d). Prehnite and albite veins are common in both samples. Albite is partially altered. The amphibole crystals in MS20-03 are also compositionally classified as magnesiohornblende to tremolite-actinolite but have lower Si (6.67–7.60 p.f.u.) than those of MS20-01 at a given Mg# range (0.86–0.94). These mineral assemblages indicate that they have experienced epidote-amphibolite facies metamorphism.

**ANALYTICAL METHODS**

We determined whole-rock major and trace elements of the studied amphibolites using an X-ray fluorescence spectrometer (XRF; Rigaku ZSX Prims II) at Earthquake Institute, The University of Tokyo. Glass beads made from the mixture of 1.000 g rock powder and 5.000 g flux (Li2B4O7) were prepared for the analysis. The detailed analytical procedures and methods followed Hokinishi et al. (2015).

Whole-rock trace element, including rare-earth element (REE), was measured using a laser-ablation inductively coupled plasma mass spectrometer (LA-ICP-MS) at Kanazawa University. A 193 nm ArF excimer laser (GeoLas Q+) and quadrupole mass spectrometer (Agilent7500s) were employed in this analysis. Direct-fused glasses made from rock powder were prepared for the analysis. The detailed analytical procedures and
methods followed Tamura et al. (2015). The results of the whole-rock analyses using XRF and LA–ICP–MS are listed in Table 1.

Zircon grains for U–Pb dating were recovered by hand picking after separations using heavy liquid (sodium polytungstate) and neodymium magnet. The recovered zircon grains were mounted into PFA fluorocarbon polymers sheets and were polished with diamond paste.

Zircon U–Pb dating was carried out using an LA–ICP–MS at Central Research Institute of Electric Power Industry. A 213 nm Nd–YAG laser (NewWave Research) and sector-field mass spectrometer (Thermo Fisher Scientific ELEMENT XR) were employed in this analysis.

The detailed analytical procedures and methods followed those of Ito (2014). Note that we employed a laser spot size of 20 µm with ∼ 7 J/cm² fluence, and the zircon standard 91500 (Wiedenbeck et al., 1995) was adopted as a primary reference zircon instead of the Fish Canyon Tuff (Ito, 2014). We performed duplicate analysis (with 30 µm spot size) for the zircon grains from MS20–03 to ensure the reliability of our measurement. The average reproducibility for 206Pb/238U age was 96%. The data obtained in the second analysis, which show lower discordance% {= [(207Pb/235U age)/(206Pb/238U age) − 1] × 100} than those of the first one, are preferentially used for discussion. A calculation software, Isoplot 4.0 (Ludwig, 2003), was used for processing data and preparing diagrams. The U–Pb ages of the Plešovice zircon was measured as a secondary standard during the ICP–MS analyses. The weighted mean 206Pb/238U ages of the Plešovice zircon obtained through the two analyses were 336.6 ± 6.3 (MSWD = 2.1) and 338.6 ± 2.8 (MSWD = 0.9), which were in concordance with the recommended value (337.13 ± 0.37 Ma; Sláma et al., 2008). The analytical results of the samples and Plešovice zircon are listed in Supplementary Tables S1 and S2 (available online from https://doi.org/10.2465/jmps.191205), respectively.

The cathodoluminescence (CL) images of the polished zircon grains were obtained using a CL detector (Oxford MiniCL) equipped with a scanning electron microscope (JEOL SEM JSM–5600) at Chiba University.

RESULTS

Whole-rock geochemistry

There is no significant difference in major element composition between the two amphibolite samples. SiO₂ and MgO contents recalculated to anhydrous are slightly less than 50 and 11 wt%, respectively. FeO*/MgO ratios are low (<1.0). Chondrite–normalized REE patterns show depletion in light REE and positive anomalies in Eu (Fig. 3a). Mid-ocean ridge basalt (MORB)–normalized trace element patterns display distinct positive anomalies in Rb, Ba, Pb, and Sr (Fig. 3b), which are highly mobile during secondary chemical modification. All MORB-normalized values, except for these anomalously enriched elements are lower than 1. Negative anomalies in Zr and Hf are present in both samples. The pattern of MS20–1 also exhibits slight negative anomalies in Nb and Ti.

Zircon CL image and U–Pb dating

We performed the U–Pb dating and CL observation of 18 and 14 grains for MS20–01 and MS20–03, respectively.

Table 1. Results of the whole-rock analyses using XRF and ICP–MS

|          | XRF analysis | ICP–MS analysis |
|----------|--------------|-----------------|
|          | MS20–1       | MS20–3          | MS20–1       | MS20–3         |
| SiO₂     | 49.13        | 47.68           | Li           | 0.83           | 3.54           |
| TiO₂     | 0.24         | 0.42            | B            | 6.11           | 6.46           |
| Al₂O₃    | 13.75        | 15.71           | Sc           | 40.71          | 39.93          |
| FeO*     | 4.86         | 5.92            | Ti           | 1637           | 3333           |
| MnO      | 0.11         | 0.12            | V             | 170.0          | 208.2          |
| MgO      | 11.28        | 10.80           | Cr            | 835.8          | 274.6          |
| CaO      | 14.73        | 12.22           | Co            | 39.70          | 39.42          |
| Na₂O     | 1.81         | 2.25            | Ni            | 211.4          | 127.1          |
| K₂O      | 0.12         | 0.23            | Rb            | 1.79           | 1.38           |
| P₂O₅     | 0.03         | 0.01            | Sr            | 192.5          | 127.7          |
| Total    | 96.06        | 95.36           | Y             | 10.19          | 12.28          |
| (ppm)    |              |                 | Zr            | 9.35           | 18.85          |
| Sc       | 36.3         | 32.7            | Nb            | 0.24           | 0.58           |
| V        | 167.8        | 197.4           | Cs            | 0.51           | 0.14           |
| Cr       | 1004.6       | 433.4           | Ba             | 28.41          | 48.26          |
| Co       | 34.8         | 34.6            | La             | 0.43           | 0.57           |
| Ni       | 240.4        | 137.0           | Ce             | 1.56           | 2.04           |
| Cu       | 39.1         | 76.1            | Pr             | 0.30           | 0.38           |
| Zn       | 25.0         | 31.7            | Nd             | 1.89           | 2.32           |
| Ga       | 10.0         | 12.5            | Sm             | 0.84           | 0.99           |
| Rb       | 0.2          | bdl             | Eu             | 0.41           | 0.53           |
| Y        | 10.7         | 12.9            | Gd             | 1.31           | 1.53           |
| Sr       | 181.2        | 121.6           | Tb             | 0.25           | 0.29           |
| Zr       | 5.2          | 9.0             | Dy             | 1.81           | 2.11           |
| Nb       | bdl          | bdl             | Ho             | 0.38           | 0.44           |
| Ba       | 31.1         | 46.0            | Er             | 1.15           | 1.37           |
| Pb       | 2.4          | 0.6             | Tb             | 0.16           | 0.19           |
| Th       | bdl          | bdl             | Yb             | 1.07           | 1.32           |
| FeO*, total iron calculated as FeO. bdl, below detection limit.
The observed zircon grains generally show stubby and anhedral shape, and some of them have fluid inclusions and irregular cracks. The zircon grains from MS20–01 do not show any distinct zoning patterns under CL observation. The CL images of the zircon grains from MS20–03 exhibit oscillatory and sector zonings, and some of them have thin overgrowth rims (Fig. 4).

In general, concordance between the $^{207}\text{Pb}/^{235}\text{U}$ and $^{206}\text{Pb}/^{238}\text{U}$ ratios obtained from the studied amphibolites is not sufficient enough for a meaningful discussion (Figs. 5a and 5b), and, particularly, the results from MS20–01 are lacking reliability. This comes from the high errors of $^{207}\text{Pb}/^{235}\text{U}$, possibly due to the counting statistics of measurement and also possible common Pb. Only two analyses from MS20–01 and 11 from MS20–03 show discordance of less than 10%. The weighted mean $^{206}\text{Pb}/^{238}\text{U}$ ages calculated from the concordant zircon grains are $573 \pm 29$ Ma (mean square weighted deviation (MSWD) < 0.1) and $483 \pm 17$ Ma (MSWD = 4.6) for MS20–01 and MS20–03, respectively (Figs. 5c and 5d). Instead, when the discordance is increased to 20%, the weighted mean $^{206}\text{Pb}/^{238}\text{U}$ ages result in $541 \pm 28$ Ma (7 grains; MSWD = 2.0) and $477 \pm 13$ Ma (17 grains; MSWD = 4.3) (Figs. 5c and 5d), which are consistent with the above ages within the error ranges. In this study, the $^{206}\text{Pb}/^{238}\text{U}$ age of $483 \pm 17$ Ma is considered for MS20–03 as a reliable geochronological data, and the $^{206}\text{Pb}/^{238}\text{U}$ age of $541 \pm 28$ Ma for MS20–01 is treated as reference information.

**DISCUSSION**

The whole-rock geochemistry of the studied amphibolites is characterized by the depletion in trace element content. The REE patterns are similar to that of MORB in terms of light REE depletion (Fig. 3a). The Eu positive anomalies were probably caused by plagioclase accumulation, indicating that the protolith of these amphibolites is gabbroic rocks. On the other hand, the Zr and Hf negative anomalies are detected in the trace element patterns (Fig. 3b). In particular, MS20–1 also has slight Nb and Ti negative anomalies, which imply supra-subduction zone magmatism (e.g., Pearce and Peate, 1995). However,
such Nb, Zr, and Ti negative anomalies are also observed in gabbroic rocks recovered from fracture zones in the Mid-Atlantic Ridge (Fig. 3b; e.g., Godard et al., 2009). These anomalies are attributed to plagioclase accumulation because of the high incompatibility of these elements against plagioclase (Drouin et al., 2009). In addition, zircon fractionation could have also been involved in Zr and Hf anomalies. Although it is difficult to identify which tectonic settings are suitable for the protolith of the studied amphibolites, a spreading axis in an oceanic basin or a backarc basin would be more likely.

The zircon U–Pb data of the studied amphibolites yield Cambrian ages. The CL images of the zircon grains analyzed exhibit zoning indicating crystallization in an igneous environment (Fig. 4) but not the characteristics of metamorphic and inherited zircons. The zircon Th/U ratios ranging from 0.4 to 1.2 support that they crystallized from igneous processes but not metamorphic events that are expected to be as low as <0.1 (e.g., Rubatto, 2017). Zircon in high-grade metamorphic rocks, such as granulite and eclogite, exhibit homogeneous metamorphic overgrowth (Corfu et al., 2003). The thin overgrowth rims observed in the studied zircon grains possibly formed during epidote-amphibolite facies metamorphism. The obtained Cambrian U–Pb ages indicate the ages of zircon crystallization from magma, i.e., the solidification ages of the protolith of the studied amphibolites. It is uncertain what the age gap of more than 50 m.y. between the two samples means at this moment.

Tsujimori (2010) regarded 480–400 Ma amphibolites associated with Early Paleozoic serpentinite mélanges in Japan as the oldest rocks that experienced high-P metamorphism. The amphibolites are generally characterized by epidote-amphibolite facies, but the Fuko Pass amphibolites from Kyoto Prefecture, southwest Japan, bear kyanite, paragonite, and rutile, and their protolith is interpreted as olivine and plagioclase cumulate (Tsujimori and Ishiwatari, 2002; Tsujimori and Liou, 2004). The presence of Al-rich clinopyroxene and spinel pseudomorph of corundum–magnetite symplectite in the Fuko Pass amphibolites implies that the rocks have experienced granulite facies metamorphism prior to epidote-amphibolite facies, and the subduction of the thick oceanic crust was inferred (Tsujimori and Liou, 2004). The 443–403 Ma hornblende K–Ar ages of the Fuko Pass amphibolites were interpreted as the ages of a subduction-
related metamorphic event (Tsujimori et al., 2000). The studied amphibolites lack exact high-P mineral assemblages, but the assemblage of clinozoisite and rutile is similar to that of the Fuko Pass amphibolites. The studied amphibolites have strong deformation texture and are distinct from non-subduction ophiolitic epidote-amphibolites. For instance, epidote-amphibolites in the Late Permian Yakuno ophiolite, southwest Japan, are generally lacking deformed texture and partially preserve igneous textures (Ishiwatari, 1985; Ichiyama and Ishiwatari, 2004). The studied amphibolites are possibly equivalent to the Fuko Pass amphibolites.

Comparable age data have been reported from several localities in the Paleozoic formations in Japan. Tsujimori et al. (2005) reported the zircon U-Pb ages of jadeite from the Osayama serpentinite melange and interpreted them as a hydrothermal origin. Fu et al. (2010) investigated the oxygen isotopic zircon composition of the Osayama jadeite and identified igneous cores surrounded by hydrothermal rims in single zircon crystals. The U-Pb ages of igneous cores and hydrothermal rims are not distinguishable in the range of 530–450 Ma, implying the subduction of young hot oceanic crust (Tsujimori, 2010). The wide age variation of the hydrothermal zircon shows a correlation with initial epsilon hafnium values, suggesting the timing of a zircon source differentiated from the mantle at ~ 570 Ma (Tsujimori, 2017). Kunugiza et al. (2017) also confirmed the older inherited ages of 560 ± 16 Ma than the hydrothermal stage of 519 ± 17 Ma from the zircon U-Pb geochronology of the Omi jadeite. The zircon U-Pb ages of about 490 Ma have also been reported from glaucophane-bearing metabasalts from the Kurosegawa serpentinite melange in the Outer Zone of southwest Japan (Osama et al., 2014). Kimura and Hayasaka (2019) have recently reported the zircon U-Pb ages of 545–532 Ma from gabbros interfingering with peridotites in the Oeyama ophiolite. The serpentinites around the Omi area are equivalent to the mantle section of the Oeyama ophiolite and are derived from the fragments of the forearc mantle (Machi and Ishiwatari, 2010; Tsujimori, 2004). The age similarity between the Oeyama ophiolite and high-P rocks in the serpentinite melanges assists the subduction of the young hot oceanic crust in the East Asian convergent plate margin during the Cambrian.

CONCLUSIONS

The amphibolite blocks in the Omi serpentinite melange have the protolith of oceanic gabbro, which probably formed in a mid-ocean ridge or a backarc basin. The zircon U-Pb ages indicate that the protolith has crystallized in the Cambrian. The protolith ages of the studied amphibolites are comparable with those of the Early Paleozoic Oeyama ophiolite and the protolith of high-P rocks in Paleozoic serpentinite melanges in Japan. The young hot oceanic crust would have subducted in the East Asian convergent plate margin during the Cambrian.

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SUPPLEMENTARY MATERIALS

Supplementary Tables S1–S2 are available online from https://doi.org/10.2465/jmps.191205.

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