Geochemical characteristics of an ophiolitic complex from Mt. Tenzan area, Saga Prefecture, northern Kyushu

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The metamorphic complex from the Mt. Tenzan area in northern Kyushu consists mainly of mafic rocks with small amounts of siliceous, calc-silicate, and ultramafic rocks. These lithofacies can be recognized as an ophiolitic complex. Metamorphosed mafic rocks are divided into two types, amphibolites I and II, which are probably derived from supracrustal and intrusive rocks, respectively. The geochemical data of both amphibolites plotted within the field between mid-ocean ridge and island arc basalts; such geochemical features resemble those of back-arc basin basalts. As the metamorphic complex was intruded by Cretaceous granitoids, protoliths of the complex could have been formed prior to the Cretaceous. The protolith lithofacies assemblage and geochemical constraints of the Tenzan metamorphic complex indicate the correlation with the Yakuno ophiolite rather than the Oeyama ophiolite.

Keywords: Whole-rock geochemistry, Metamorphosed mafic rocks, Ophiolite complex, Tenzan Area, Northern Kyushu

INTRODUCTION

The metamorphic rocks from northern Kyushu occur as large blocks in the Cretaceous granitoids. The protoliths of these metamorphosed rocks are composed of pelitic, calc-silicate, and mafic rocks, with small amounts of limestone and ultramafic rocks (Karakida et al., 1969). The zircon uranium–lead (U–Pb) dating of the metamorphosed clastic rocks from northern Kyushu indicates that the ages of the youngest peaks obtained from detrital zircon cores are approximately 400 and 250 Ma, which correspond to the Renge and Suo high-pressure metamorphic belts, respectively (Fig. 1; Tsutsumi et al., 2003, 2011; Adachi et al., 2012; Miyazaki et al., 2017; Yuhara et al., 2021).

The Tenzan area in Saga Prefecture is underlain by metamorphosed mafic rocks with small amounts of siliceous, calc-silicate, and ultramafic rocks (Oshima, 1964) (Fig. 1). According to the previous study (Nishimura, 1998), the Tenzan area has been thought to be a member of the Renge belt because the regional foliations are continued to the high-pressure/low-temperature metamorphic rocks in the Sasaguri area, 40 km north-east from the Tenzan area. Based on the metamorphic pressure–temperature conditions and lithological features, Yamada et al. (2008) correlated the metamorphic rocks from the Tenzan area with those of the Higo metamorphic rocks without any whole-rock analyses. However, the lithological assemblage of the Tenzan area is equivalent to an ophiolite complex defined by Ishiwatari (2010), which indicates at least two members of the following lithofacies: 1) mantle peridotite, 2) mafic and/or ultramafic cumulate, and 3) mafic volcanic rocks. This study examined the mineralogical and whole-rock geochemical characteristics of metamor-
phosed mafic and ultramafic rocks from the Tenzan area and compared our geochemical results with those of other Paleozoic ophiolite complexes in southwest Japan. This is the first geochemical analysis of metamorphosed mafic and ultramafic rocks in this area. The data presented here provide useful information for geotectonic comparisons with Paleozoic ophiolite complexes in southwest Japan.

FIELD OCCURRENCE AND PETROLOGICAL DESCRIPTIONS

In terms of field occurrence, the metamorphosed mafic rocks are divided into two types: amphibolites I and II. Amphibolite I is fine-grained and well-foliated, with thin layers and lenses of siliceous and calc-silicate gneisses. In contrast, amphibolite II is coarse-grained with weak foliation locally accompanied by serpentinite and tremolite rocks as lenticular bodies or lenses (Fig 1c). Based on textural evidence, the mafic and ultramafic rocks in this area underwent a thermal effect at the boundary with the Cretaceous granitoids, but outside of the boundary, the rocks were barely affected by contact metamorphism (Yamada et al., 2008).

Amphibolite I has a fine-grained and nematoblastic texture. It consists mainly of hornblende and plagioclase with small amounts of quartz and includes ilmenite, titanite, and apatite as accessory minerals (Fig. 2a). The hornblende has a zonal structure with a light-green core, a greenish-brown mantle, and a light-green rim. In addition to these minerals, the calc-silicate thin layers intercalated with amphibolite I include clinopyroxene, epidote, and calcite.

Amphibolite II is coarse-grained and is composed of hornblende and plagioclase with small amounts of biotite, potassium feldspar, and quartz, containing ilmenite, titanite, and apatite as accessory minerals (Fig. 2b). Hornblende rarely developed zonal structure and frequently contains poikilitic plagioclase inclusions. Potassium feldspar appears in the plagioclase grains as an antiperthitic phase.

Serpentinite is composed of mesh-structured serpentine, talc, tremolite, clinopyroxene, and phlogopite (Fig. 2c). We performed a chemical analysis of the olivine and chromian spinel in serpentinite using a JEOL JXA-8230 electron probe micro-analyzer (EPMA) at the Center for Instrumental Analyses, Yamaguchi University. Analytical procedures of mineral chemistry were described as described by Eshima (2021). Olivine (F092-00) appears locally as a relict mineral and includes chromian spinel with a dark brownish color. Its chrome number \([\text{Cr}\# = \text{Cr}/(\text{Cr} + \text{Al})]\) ranges from 0.85 to 0.89. The tremolite displays columnar shapes with a maximum length of 2.0 cm. The phlogopite exhibits flake crystals surrounding the olivine crystals. Mineral data for olivine and chromian spinel are available upon request from the corresponding authors.

The tremolite rock primarily consists of tremolite and clinopyroxene with small amounts of serpentine, chlorite, and opaque minerals (Fig. 2d). The tremolite is weakly foliated.

WHOLE-ROCK GEOCHEMISTRY

The whole-rock chemical compositions of amphibolites I and II, serpentinite, and tremolite rocks were determined by X-ray fluorescence (XRF) analysis. The analyzed samples were free from alteration and veins, and the amounts of 300–600 g were crushed by hand with a W-mortar. Crushed samples were powdered using an automatic W-mortar. Detailed analytical procedures are described in Eshima and Owada (2018). After ignition in a furnace at 950 °C for 2 h, the samples were measured for loss on ignition; the ignited samples (1.0 g) were mixed five times the amount of Li2B4O7 as flux. The mixed samples were melted to make glass beads using a bead sampler. The glass beads were analyzed using an XRF analyzer (Rigaku ZSX primus-II) installed at the Center of Instrumental Analysis, Yamaguchi University. The analytical conditions included an electric voltage of 50 kV and an electric current of 60 mA, using a Rh anode X-ray tube. The analyzed elements were SiO2, TiO2, Al2O3, Fe2O3, MnO, MgO, CaO, Na2O, K2O, and P2O5 as the major elements and were Ba, Cr, Nb, Ni, Rb, Sr, V, Y, Zn, and Zr as trace elements. To validate the quantitative da-

Figure 2. Thin section photographs. Plane-polarized light image of (a) amphibolite I, (b) amphibolite II, and (c) serpentinite. Cross-polarized light image of (d) tremolite rock. Hbl, hornblende; Pl, plagioclase; Ol, olivine; Cpx, clinopyroxene; Tlc, talc; Srp, serpentinite; Tr, tremolite; Spl, spinel.
we measured the standard rock samples, JB–2, JB–3, and JGb–2, provided by the Geological Survey of Japan.

The results are presented in Table 1. The measured values of the standard samples were identical to the recommended values. In addition to XRF analyses, trace elements, including rare earth elements (REEs), for amphibolites I and II were determined by inductively coupled plasma mass spectrometry (ICP–MS) at Activation Laboratory Ltd., Canada.

A total of 34 samples (15 of amphibolite I, 13 of amphibolite II, 4 of serpentinites, and 2 of tremolite rocks) were analyzed by XRF, and 9 samples of amphibolites I and II were analyzed using ICP–MS. The representative results are presented in Table 2. All of the whole-rock data used in this study can be requested from the corresponding authors.

Table 1. Chemical compositions of GSJ standard samples, JB–2, JB–3, and JGb–2

| Sample | JB–2 | JB–3 | JGb–2 |
|--------|------|------|-------|
|        | r.v. | m.v. | r.v. | m.v. | r.v. | m.v. |
| (wt%)  |      |      |      |      |      |      |
| SiO2   | 52.96| 52.52| 50.77| 51.27| 47.13| 46.73|
| TiO2   | 1.18 | 1.18 | 1.43 | 1.42 | 0.57 | 0.55 |
| Al2O3  | 14.56| 14.45| 17.13| 17.09| 23.81| 23.51|
| Fe2O3  | 14.17| 14.46| 11.78| 11.99| 6.78 | 6.83 |
| MnO    | 0.22 | 0.22 | 0.18 | 0.18 | 0.13 | 0.13 |
| MgO    | 4.59 | 4.57 | 5.17 | 5.37 | 6.27 | 6.18 |
| CaO    | 9.77 | 9.86 | 9.75 | 9.76 | 14.30| 14.33|
| Na2O   | 2.03 | 1.91 | 2.72 | 2.68 | 0.93 | 0.92 |
| K2O    | 0.42 | 0.43 | 0.78 | 0.78 | 0.06 | 0.06 |
| P2O5   | 0.10 | 0.10 | 0.29 | 0.31 | 0.02 | 0.03 |
| Total  | 100.00| 99.70| 100.00| 100.85| 100.00| 99.27|
| (ppm)  |      |      |      |      |      |      |
| Ba     | 222  | 213  | 245  | 273  | 37   | 60  |
| Cr     | 28   | 25   | 58   | 63   | 125  | 115 |
| Nb     | 2    | 1    | 3    | 3    | 2    | 1   |
| Ni     | 17   | 2    | 36   | 31   | 14   | 10  |
| Rb     | 7    | 5    | 15   | 11   | 3    | 3   |
| Sr     | 178  | 180  | 403  | 407  | 438  | 438 |
| V      | 575  | 576  | 372  | 382  | 174  | 174 |
| Y      | 25   | 26   | 27   | 24   | 5    | 7   |
| Zn     | 108  | 110  | 100  | 103  | 49   | 47  |
| Zr     | 51   | 51   | 98   | 100  | 12   | 13  |

r.v., recommended value; m.v., measured value; GSJ, Geological Survey of Japan.

Table 2. Representative whole-rock chemical composition of metamorphosed mafic rocks from the Tenzan area

| Type   | Amp–I | Amp–I | Amp–I | Amp–I | Amp–II | Amp–II |
|--------|-------|-------|-------|-------|--------|--------|
| Sample | no.   | 1903  | 1903  | 1903  | 1905   | 1905   |
|        | 0202  | 1401  | 2601  | 2603  | 0301   | 0302B  |
| (wt%)  |       |       |       |       |        |        |
| SiO2   | 52.96 | 52.52 | 50.77 | 51.27 | 47.13  | 46.73  |
| TiO2   | 1.18  | 1.18  | 1.43  | 1.42  | 0.57   | 0.55   |
| Al2O3  | 14.56 | 14.45 | 17.13 | 17.09 | 23.81  | 23.51  |
| Fe2O3  | 14.17 | 14.46 | 11.78 | 11.99 | 6.78   | 6.83   |
| MnO    | 0.22  | 0.22  | 0.18  | 0.18  | 0.13   | 0.13   |
| MgO    | 4.59  | 4.57  | 5.17  | 5.37  | 6.27   | 6.18   |
| CaO    | 9.77  | 9.86  | 9.75  | 9.76  | 14.30  | 14.33  |
| Na2O   | 2.03  | 1.91  | 2.72  | 2.68  | 0.93   | 0.92   |
| K2O    | 0.42  | 0.43  | 0.78  | 0.78  | 0.06   | 0.06   |
| P2O5   | 0.10  | 0.10  | 0.29  | 0.31  | 0.02   | 0.03   |
| Total  | 100.00| 99.70 | 100.00| 100.85| 100.00| 99.27  |
| (ppm)  |       |       |       |       |        |        |
| Ba     | 222  | 213  | 245  | 273  | 37   | 60   |
| Cr     | 28   | 25   | 58   | 63   | 125  | 115  |
| Nb     | 2    | 1    | 3    | 3    | 2    | 1    |
| Ni     | 17   | 2    | 36   | 31   | 14   | 10   |
| Rb     | 7    | 5    | 15   | 11   | 3    | 3    |
| Sr     | 178  | 180  | 403  | 407  | 438  | 438  |
| V      | 575  | 576  | 372  | 382  | 174  | 174  |
| Y      | 25   | 26   | 27   | 24   | 5    | 7    |
| Zn     | 108  | 110  | 100  | 103  | 49   | 47   |
| Zr     | 51   | 51   | 98   | 100  | 12   | 13   |

Type: Amp–I, amphibolite I; Amp–II, amphibolite II; LOI, loss on ignition.

Geochemical characteristics of an ophiolitic complex from Tenzan area

Table 2. Representative whole-rock chemical composition of metamorphosed mafic rocks from the Tenzan area

A total of 34 samples (15 of amphibolite I, 13 of amphibolite II, 4 of serpentinites, and 2 of tremolite rocks) were analyzed by XRF, and 9 samples of amphibolites I and II were analyzed using ICP–MS. The representative results are presented in Table 2. All of the whole-rock data used in this study can be requested from the corresponding authors. Figure 3 shows the total alkali versus silica (TAS) and FeO*/MgO versus SiO2 wt% diagrams (FeO* = 0.9 × Fe2O3*). Amphibolites I and II belong to the subalkaline and tholeite series, respectively (Figs. 3a and 3b). The FeO*/MgO ratios and Cr (ppm) contents show that the geochemical features of amphibolite II are more evolved than those of amphibolite I (Fig. 3b, Table 2).
Amphibolite I intercalates with thin layers and lenses of siliceous and calc-silicate gneisses. In contrast, amphibolite II contacts with lenticular bodies or lenses of serpentinite and tremolite rocks, and they probably occur as xenoblocks (Fig. 1c). Based on the field occurrence, protoliths of amphibolites I and II would, therefore, be supracrustal and intrusive rocks, respectively. The lithological assemblage, including amphibolites I and II and serpentinite in the Tenzan area, is equivalent to an ophiolite complex as defined by Ishiwatari (2010). Because the samples from the Tenzan area were intruded by Cretaceous granitoids, they can be recognized as a pre-Cretaceous ophiolitic complex. Therefore, the metamorphic complex from the Tenzan area can be compared to geochemical features of the Oeyama or Yakuno ophiolites.

The rock types of the Oeyama ophiolite are dominated by mantle peridotite and cumulate rocks (Kurokawa, 1985). In contrast, the Yakuno ophiolite includes supra-crustal rocks in addition to cumulate rocks and mantle peridotites (Ishiwatari, 1985a). Figure 4 shows the chemical composition of chromian spinels and olivines in serpentinite from the Tenzan area. (a) Compositional relationship between coexisting olivine Fo and chromian spinel Cr#. Olivine spinel mantle array (OSMA) is after Arai (1994), (b) Compositional relationships between Mg# and Cr# of chromian spinel. The data of this study are plotted as the average value from one sample. The compositional ranges of the data are shown as bars accompanied with data plots. The mantle peridotites from the Oeyama (Arai, 1980; Tsujimori, 1998; Machi and Ishiwatari, 2010) and Yakuno ophiolites (Ishiwatari, 1985a, b) are shown for comparison.

Figure 5a shows the compositional range of primitive basaltic magmas with their differentiation trends and the accumulation directions of specific minerals (Kempston et al., 1997). Kempston et al. (1997) stressed that metamorphosed mafic rocks up to amphibolite facies grade should be adopted in this diagram to determine the geochemical compositions of their protoliths. Amphibolites I and II studied here underwent amphibolite facies metamorphism (Yamada et al., 2008). Therefore, the geochemical compositions of the studied samples can be adopted to the diagram of Kempston et al. (1997) for estimating the characteristics of the protoliths. The analyzed samples of amphibolites I and II generally show evolved compositions, with some samples of amphibolite I plotted in the primitive basaltic field (Fig. 5a); however, the samples do not show accumulation trends. Therefore, the geochemical data for amphibolite I and II reflect the liquid composition. The mafic rocks from the Yakuno ophiolite also possess the similar compositional ranges, but those from the Oeyama ophiolite are plotted outside the liquid compositions. Figure 5b depicts the Mg# versus TiO2 wt% diagram showing the data from the Tenzan area, the Yakuno and the Oeyama ophiolites (Tsujimori and Ishiwatari, 2002; Ichiyama and Ishiwatari, 2004; Suda and Hayasaka, 2009; Kimura and Hayasaka, 2019). All data were plotted within the field, from mid-ocean ridge basalt (MORB) to island arc basalt (IAB). A diagram of the N–MORB-normalized La/Y and Nb/La ratios are plotted for amphibolites I and II and the mafic rocks from the Yakuno and Oeyama ophiolites, as well as the
compositions of the IAB, E-MORB with the compositional field of back-arc basin basalts (BABB) compiled by Suda et al. (2014) (Fig. 5c). Amphibolites I and II from the Tenzan area and the mafic rocks from the Yakuno ophiolite are plotted between N-MORB and IAB compositions, and many samples exhibit BABB affinity (Fig. 5c). The spider diagram shows that amphibolites I and II have negative Nb anomalies (Fig. 5d). The two samples of amphibolite II plotted near the composition of IAB in Figure 5c display a significant negative Nb anomaly in the spider diagram (Fig. 5d) and rightward decrease in the REE patterns similar to those of the metabasalt from the Yakuno ophiolite (Fig. 5e). This implies that amphibolites I and II formed in island arc and/or back-arc basin environments. Suda et al. (2014) inferred that the crustal evolution of the Yakuno ophiolite was characterized by intra-oceanic island arc and back-arc basin settings (Figs. 5c and 5d). Amphibolites I and II from the Tenzan area could have experienced the crustal evolution in the island arc and back-arc basin settings similar to the Yakuno ophiolite.

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