Garnet-clinopyroxene amphibolite from the Takahama metamorphic rocks, western Kyushu, SW Japan: evidence for high-pressure granulite facies metamorphism

Takeshi Ikeda*, Hitoshi Yoshida*, Kazuhiro Arima*, Tadao Nishiyama**, Takeru Yanagi* and Kazuhiro Miyazaki***

*Department of Earth and Planetary Sciences, Graduate School of Science, Kyushu University, 33 Hakozaki, Fukuoka 812-8581, Japan
**Department of Earth Sciences, Kumamoto University, 2–39–1 Kurokami, Kumamoto 860–8555, Japan
***National Institute of Advanced Industrial Science and Technology (AIST), Geological Survey of Japan, AIST Tsukuba Central 7, Tsukuba 305–8568, Japan

Garnet-clinopyroxene amphibolites occur in the mylonite unit structurally at the top of the Takahama metamorphic rocks—a member of the Nagasaki Metamorphic Rocks, western Kyushu, SW Japan. Geothermoobarometry applied to an equigranular garnet-clinopyroxene amphibolite, which escaped severe mylonitization, yields the metamorphic condition of $770 \pm 70^\circ\text{C}$ and $1.1 \pm 0.2$ GPa. This condition is comparable with the high-pressure granulite facies, and is much higher-grade than those of the underlying crystalline schists of the Takahama metamorphic rocks that belong to the epidote-glaucophane schist subfacies. Despite the presence of such a gap in metamorphic condition between the crystalline schists and the mylonite unit, nearly identical muscovite $K$-Ar ages have been obtained from these units, which indicate that both units were juxtaposed at depth before they cooled below the closure temperature of the muscovite $K$-Ar system.

Keywords: Garnet-clinopyroxene amphibolite, High-pressure granulite facies, Takahama metamorphic rocks, Nagasaki Metamorphic Rocks

INTRODUCTION

The Nagasaki Metamorphic Rocks occur in three separate regions along the west coast of Kyushu, Japan, i.e., the Nishisonogi, Nomo and Amakusa-Takahama areas (Tachibana, 1955; Karakida et al., 1969) (Fig. 1). Formerly, they were considered to form a single metamorphic belt belonging to either the Sanbagawa Metamorphic Belt, to the Sangun Metamorphic Belt, or other metamorphic belts. Progress of petrological studies, however, reveals that the metamorphic facies are different in the various areas: epidote-glaucophane schist subfacies in Nishisonogi (e.g., Nishiyama, 1989); pumpellyite-actinolite facies to epidote-amphibolite facies in Nomo (e.g., Miyazaki and Nishiyama, 1989); and lower greenschist facies (close to glaucophane schist facies) to upper greenschist facies (close to epidote-amphibolite facies) in Amakusa-Takahama (Hattori and Isomi, 1976). Shigeno and Mori (2003) found tectonic blocks involving a jadeite + quartz assemblage in the Nishisonogi metamorphic rocks, which indicates the glaucophane schist facies.

In the Nomo area, Nishimura (1998) and Nishimura et al. (2004) recognized two geological units bounded by thrusts. One of the two units yielded phengite $K$-Ar ages similar to those of the Sangun (Suo) Metamorphic Rocks in contrast to the other unit that has ages similar to those of the Sanbagawa Metamorphic Rocks. Between the two units, Takeda et al. (2002) found high-grade metamorphic rocks that were mylonitized. In the Amakusa-Takahama area, one of the authors, Miyazaki, has clarified the regional occurrence of mylonitized rocks including garnet-amphibolites along the boundary between the Takahama metamorphic rocks and the Cretaceous sedimentary formations, the Himenoura Group, during geologic mapping of the Amakusa coal field (Takai et al., 1997). Takeda et al. (2002) considered that the mylonitic rocks in this
area occupied the same structural position as those in the Nomo area. These features suggest a complex history for the Nagasaki Metamorphic Rocks.

This paper describes a garnet-clinopyroxene amphibolite from the mylonite unit in the Takahama metamorphic rocks and determines the metamorphic pressure ($P$) - temperature ($T$) conditions of the amphibolite prior to the mylonitization. The estimated condition is comparable with those of the high-pressure granulite facies whereas the structurally underlying crystalline schists represent the epidote-glaucophane schist subfacies condition. Although there is a possibility that the mylonitic rocks belong to a new geologic unit that has not so far been recognized or defined, we tentatively consider them to belong to a member of the Nagasaki Metamorphic Rocks and refer to it here as the mylonite unit.

**OUTLINE OF GEOLOGY AND METAMORPHISM**

The Takahama metamorphic rocks occur in the western part of Shimoshima Island, western Kyushu and have been regarded as a member of the Nagasaki Metamorphic Rocks (Karakida et al., 1969) (Fig. 1). They are unconformably overlain by Cretaceous sediments of the Hime-noura Group (Takeda et al., 2002; Toyohara et al., 2004), but in most places they are in fault contact with the Group.
Garnet clinopyroxene amphibolite from the Takahama metamorphic rocks (Miki, 1972; Takai et al., 1997). The Himenoura Group yields bivalve fossils of Santonian to Maestrichtian age and contains detrital grains of metamorphic minerals such as epidote, hornblende and garnet (Miki, 1972; Tashiro and Noda, 1973; Tashiro, 1976). The crystalline schists of the Takahama metamorphic rocks show muscovite K-Ar ages ranging from 83 to 89 Ma (Ueda and Onuki, 1968; Ueda et al., 1977; recalculated by Hattori and Shibata, 1982). These lines of evidence suggest that the crystalline schists of the Takahama metamorphic rocks cooled below the closure temperature of muscovite in the late Cretaceous and were uplifted to the Earth’s surface almost immediately after in time to supply clastic materials to the sedimentary basin.

The Takahama metamorphic rocks consist mainly of pelitic schists with small amounts of basic and siliceous schists. Our geological interpretation (Fig. 1) is mostly consistent with that of Takai et al. (1997) in terms of the regional distribution of lithology. Mineral assemblage of the pelitic schists is chlorite + muscovite + albite + quartz + carbonaceous materials. Garnet frequently occurs in addition to the above assemblage (Fig. 2a). Basic schists intercalating with the pelitic schists have well-defined schistosity with albite porphyroblasts up to 2-3 mm in diameter. The dominant assemblage of the basic schists is actinolite + muscovite + epidote + chlorite + albite + quartz. Occasional occurrence of sodic amphibole in the basic schists coexisting with epidote and/or garnet indicates the epidote-glaucophane schist subfacies of the metamorphism (Fig. 2b). The sodic amphiboles represent a significant zoning from glaucophane with Fe$^{3+}$(Fe$^{3+}$ + Al$^{3+}$) of ca. 0.2 at the core to crossite with ca. 0.7 Fe$^{3+}$/ (Fe$^{3+}$ + Al$^{3+}$) at the rim, through which Mg/(Fe$^{2+}$ + Mg) is almost constant at 0.6. The composition of the sodic amphiboles in the study area is similar to that in the Nishisonogi metamorphic rocks (Nishiyama, 1989) rather than that (magnesio-riebeckite to crossite) in the Nomo metamorphic rocks (Miyazaki and Nishiyama, 1989). These features suggest that the metamorphic grade is comparable with that of the Nishisonogi metamorphic rocks, which is approximately equivalent to the garnet zone of the Sanbagawa Metamorphic Belt (e.g., Banno et al., 1978).

The mylonite unit occurs in the top structural position of the Takahama metamorphic rocks (Fig. 1), as first noticed by Miyazaki (Takai et al., 1997). The unit represents a significant lithological variation from melanocratic amphibolite to leucocratic siliceous rocks, most of which are mylonitized (Hattori and Isomi, 1976; Takeda et al., 2002). In the mylonitic epidote amphibolite, Takeda et al. (2004) recently discovered kyanite, which has not previously been found in the crystalline schists. Tachibana (1967) and Ueda and Onuki (1968) reported muscovite K-Ar ages of 89 and 86 Ma respectively, for a leucocratic rock at the southern margin of the metamorphic region, which are similar to those of the crystalline schists of the Takahama metamorphic rocks.

**OCCURRENCE OF THE MYLONITE UNIT**

At the boundary between the mylonite unit and the underlying crystalline schists, a ductile shear zone of ca. 50 cm thick occurs. This shear zone consists of chlorite-actinolite schists and is commonly cut by faults, which results in a fault contact between the two units in some places. Figure 3 shows a sketch map of the mylonite unit along the southern coast of Cape Bishagoiwa located on the southern margin of the metamorphic region (boxed area of Fig. 1). The mylonite unit overlies the pelitic schists

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Figure 3. Enlargement of the boxed area in Figure 1, showing the distribution of coastal exposures. Most of the rocks in the mylonite unit were mylonitized.

with a fault dipping to the north (Fig. 3). A fractured zone defines the border between the mylonite unit and the Himenoura Group at the southeastern end of the cape (Fig. 3). The fractured zone is about 40 m wide and consists of brecciated sandstone and shale. Many normal faults occur in the Himenoura Group adjacent to the fractured zone, mostly trending NE-SW with dips of 35°-80°S. Normal faults with similar trends and dips are also frequently observed in the mylonite unit.

Amphibolite is dominant in the mylonite unit, and represents a compositional layering consisting of hornblende-rich melanocratic and plagioclase-rich leucocratic layers. Monomineralic hornblende rock also occurs as a thin layer or lens in the amphibolite (Fig. 3). Siliceous rocks occur locally, showing a gradual change from amphibolite caused by variations in the modal abundance of quartz and hornblende. Both amphibolite and siliceous rocks commonly contain garnet porphyroblasts and also local clinopyroxene. These compositional layerings have a general trend that strikes NE-SW and dips gently SE (Fig. 4).

Several calcareous layers up to 50 cm thick occur mostly sub-parallel to but locally discordant with the compositional layering (Fig. 3). The calcareous rocks occasionally include small lenses of amphibolite and garnet-amphibolite. Reaction zones consisting mostly of epidote occur between the calcareous layers and amphibolite in some places.

Most rocks are mylonitized in the outcrops along the northern and southern coasts of Cape Bishagoiwa. They show linear structures gently plunging NE (Fig. 4). The lineation is defined by the preferred orientation of epidote and quartz ribbons. Fractured garnet also elongates parallel to the lineation and the fractures are filled with chlorite.

Figure 4. Plot of structural data in the area shown in Figure 3, projected onto the lower hemisphere of a Schmidt’s net.
SAMPLE DESCRIPTIONS

Microstructures

Our petrographic study focuses on a relatively less foliated and equigranular rock occurring on the southern coast of Cape Bishagoiwa, sample #961211-2n, which was scarcely mylonitized (Fig. 5). The sample contains coarse-grained hornblende, clinopyroxene, plagioclase and garnet with minor rutile, ilmenite and quartz. Retrograde epidote, sphene and chlorite occur locally.

The garnet has a number of cracks that are oriented randomly and filled with chlorite (Figs. 6a and 7a), as well as an irregular shape; it is in direct contact with grains of clinopyroxene and plagioclase. Grains of clinopyroxene meet each other at the triple point with a dihedral angle of ca. 120° (Fig. 6b). Clinopyroxene locally contains inclusions of plagioclase and hornblende. Red-
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Dish brown hornblende with a grain size of ca. 0.5 mm is usually rimmed by pale-green actinolite of < 10 μm thick, and is in some cases replaced by chlorite along its cleavage (Figs. 6c and 7b). Rounded quartz crystals with a grain size of ca. 0.3 mm occur both as inclusions in garnet and as individual grains in the matrix, suggesting that the rock was silica-saturated during prograde metamorphism. Rutile is armoured by sphene where it occurs spatially close to hornblende rimmed by actinolite (Figs. 6a, d and 7c).

Plagioclase grains are polygonal, albite-twinned and have a grain size similar to hornblende and clinopyroxene. Fine-grained aggregates composed mainly of euhedral zoisite and sporadic muscovite appear in plagioclase grains (Figs. 6d and 7d). Quartz is absent in the aggregates.

Epidote occurs in veins composed mainly of quartz and calcite. Chlorite fills cracks, which are common in garnet and locally present in clinopyroxene. Chlorite also replaces hornblende partially along its cleavage.

These lines of microstructural evidence suggest that epidote, chlorite and sphene as well as the actinolitic rims are retrograde products. The prograde mineral assemblage is inferred to be clinopyroxene + garnet + hornblende + plagioclase + quartz + rutile. Occurrence of aggregates of zoisite and muscovite within plagioclase grains is indicative of alteration of the original plagioclase. This suggests that the original composition of plagioclase would have been more calcic than those of the present plagioclase, as will be discussed later.

Mineral chemistry

The chemical composition of the constituent minerals of
sample #961211-2n was determined using an energy dispersive X-ray analyzer (LINK ISIS) attached to a scanning electron microscope (JEOL 5800LV) at Kyushu University. A 20 kV accelerating voltage and 500 pA beam current were employed with 100 s livetime. Ferric contents in clinopyroxene and garnet estimated from stoichiometry are negligible. Estimation of the ferric content in amphiboles based on 13 total cations, exclusive of Ca, Na and K, yields ferric iron less than 6.0% among the total iron for all analyses with an average of 2.3%. This calculation assumes that Mn, Mg and Fe$^{2+}$ do not enter into the M4 site. However, this assumption is not valid for amphiboles formed under high-temperature conditions (e.g., Ross et al., 1968). The maximum estimate in the amount of Mn, Mg and Fe$^{2+}$ at the M4 site is obtained by setting the total cation exclusive of K and Na as 15 where no Na enters into the M4 site. This calculation yields negative values of ferric iron for all analyses. This suggests that the former estimate provides the maximum ferric content, which is still low. Indeed, the ferric content of the analysis used to estimate metamorphic conditions shown in Table 1 is 0.5% of total iron in maximum. To avoid subjective estimates of ferric content, this study treats amphibole as ferric−free. The difference in the P−T estimation resulting from not considering the ferric iron of hornblende is negligible.

The garnet is chemically homogeneous except at the 50 μm thick margin where Ca and Fe contents increase and Mg content decreases outward. No compositional gradient is detected at the vicinity of the randomly oriented cracks that are filled with chlorite.

The clinopyroxene is chemically homogeneous, where Al$_2$O$_3$ content reaches 6.1 wt% and TiO$_2$ content is less than 1.0 wt%. The margin of the grains represents outward decrease in Al combined with increase in Ca, Mg and Si. Jadeite content is ca. 8.0 mol% and does not show significant variation within the grains.

Pleochroic variation of hornblende detected under the microscope reflects significant variation in the chemical composition. The reddish−brown interior has TiO$_2$ and

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<th>Mineral Analysed point</th>
<th>Clinopyroxene (Cpx1)</th>
<th>Garnet (GA16)</th>
<th>Hornblende (Hb9)</th>
<th>Plagioclase (Pl 39)</th>
<th>Average of areal analyses</th>
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*total Fe as FeO.

**Plagioclase shown in this table was not employed for the P−T estimation.
Al$_2$O$_3$ contents of 3.0 and 14 wt%, respectively, whereas the pale-green to colourless rim portrays virtually zero and 2.0 wt%, respectively. The former is tschermakite and the latter is actinolite based on the classification after Leake et al. (1997). A positive correlation between Na and six-coordinated Al is observed.

Plagioclase is partly replaced by aggregates of zoisite and muscovite. The anorthite content of the remaining plagioclase ranges from 1.0 to 16 mol% even in one grain (the analysis with a maximum anorthite content shown in Table 1). Zoisite replacing plagioclase is virtually free from Fe$^{3+}$. Phengite content in muscovite occurring in plagioclase is as low as Si = 6.0 - 6.1 based on 22 oxygen. The contents of FeO and MgO in muscovite are mostly below the detection limit or less than 1.0 wt%. Na/(K + Na) is 0.07 - 0.12.

Epidote occurring in quartz–calcite veins has Fe$^{3+}$/(Al + Fe$^{3+}$) of 0.16 - 0.19. Mg/(Fe + Mg) of chlorite in contact with hornblende is ca. 0.41 with Si = 6.1 based on 28 oxygen, while that in contact with garnet is ca. 0.63 with Si = 6.4.

Representative analyses of the homogeneous interior of garnet, clinopyroxene and hornblende with their calculated formulae are shown in Table 1.

**DISCUSSION**

**Estimation of original composition of plagioclase**

Microstructural features described above suggest that the original composition of the plagioclase may have been significantly different from that shown in Table 1 because of the retrograde formation of grains, mainly of zoisite and sporadic muscovite. Formation of such hydrous minerals at the expense of plagioclase requires introduction of H$_2$O component into the system. Assuming a closed system excluding H$_2$O, the isochemical decomposition of plagioclase can be described as:

$$4\text{CaAl}_2\text{Si}_2\text{O}_6 + 4\text{KAlSi}_3\text{O}_8 + 1.5\text{H}_2\text{O} \rightarrow 2\text{Ca}_2\text{Al}_2\text{Si}_3\text{O}_10\text{(OH)} + \text{KAl}_3\text{Si}_3\text{O}_10\text{(OH)}_2 + 2\text{SiO}_2$$

This reaction does not deal with the albite component, which results in the residual plagioclase becoming albite-rich. Reaction (1) is, however, inconsistent with the fact that quartz is absent within the present plagioclase. This indicates that the zoisite and muscovite within the plagioclase were formed in an open system.

It is difficult to assess precisely the amount and the species of elements that were added into or subtracted from the original plagioclase. Al and Si are generally thought to be immobile during decomposition of plagioclase to form symplectite (e.g., Mongkoltip and Ashworth, 1983). Assuming that Al and Si were immobile during the plagioclase alteration, the original composition of plagioclase was estimated based on the bulk composition of the area including plagioclase present together with aggregates of zoisite and muscovite.

Determination of modal fraction and composition of the constituent minerals is valid to estimate the bulk composition if each mineral is chemically homogeneous (e.g., Hokada, 2001). This is not the case in this study; the plagioclase exhibits significant compositional variation. This study employed instead the areal analyses of a square area of 100 $\mu$m x 100 $\mu$m by scanning an electron beam. This method involves a systematic matrix-effects error, as pointed out by Raase (1998). He compared the results based on the above two methods and revealed that the relative error due to matrix effect is ca. 10% for the anorthite content. Thus the following discussion is valid only within this uncertainty.

Results of the measurement of the 100 $\mu$m x 100 $\mu$m area differ from area to area, mainly because of significant variation in the modal abundance of zoisite between the areas (Fig. 8). The FeO and MgO contents of each analysis are less than 1.0 wt% and below detection limit, respectively. The bulk compositions lie on a line connecting zoisite and albite-plagioclase, which is consistent.
with the fact that zoisite and host albitive plagioclase are dominant phases. This line is depleted in Al and Si as compared with the line representing stoichiometric plagioclase solid solution (Fig. 8). The average of the analyses is shown in Table 1, and has Si/Al atomic ratio of 1.97. Considering analytical uncertainties the original composition of the plagioclase was estimated to be in the range of An 0.31–0.38.

Geothermobarometry

Several geothermobarometers were applied to the sample to estimate the metamorphic conditions. They include Fe–Mg exchange reactions between garnet and clinopyroxene (Råheim and Green, 1974; Ellis and Green, 1979; Powell, 1985; Pattison and Newton, 1989) and those between garnet and hornblende (Graham and Powell, 1984) for geothermometers, and the net-transfer reactions among garnet, clinopyroxene, plagioclase and quartz (Newton and Perkins, 1982) and among garnet, hornblende, plagioclase and quartz (Kohn and Spear, 1990) for geobarometers. The ferric contents of garnet, clinopyroxene and hornblende are negligible, as described above, therefore all the calculations were performed with ferric-free compositions. The anorthite content employed in the calculation is based on the Al–Si ratio of the average of the areal analyses shown in Table 1. These calculations provide temperatures ranging from 700 to 850°C and pressures from 0.98 to 1.15 GPa (Fig. 9). As pointed out by Newton and Perkins (1982), the use of their geobarometer would yield a pressure estimate systematically lower than the other barometers by ca. 0.2 GPa. This correction results in a pressure range of 1.00–1.26 GPa. The uncertainty in estimating the original composition of the plagioclase would yield an error in the pressure estimation of 0.06 GPa. The use of the actual composition of the plagioclase, instead of the reconstructed composition, would result in an overestimation of pressure by up to 0.5 GPa. Our best estimate of the metamorphic condition is, therefore, 770 ± 70°C and 1.1 ± 0.2 GPa.

The estimated condition is within the stability field of kyanite (Bohlen et al., 1991), and also within the stability field of zoisite + kyanite assemblage (Perkins et al., 1980). This result is consistent with the occurrence of kyanite that coexisted with clinzoisite together with garnet and hornblende in the same outcrop described by Takeda et al. (2004).

The assemblage, garnet + clinopyroxene + plagioclase, characterizes a transition between the granulite and eclogite facies in terms of continuous reactions that produce garnet at the expense of orthopyroxene and plagioclase with increasing pressure (e.g., O’Brien and Rötzler, 2003; Pattison, 2003). This assemblage with or without hornblende and quartz occurs in the terranes not only of high-pressure granulite facies but also of amphibolite facies (e.g., Pattison, 2003). Figure 9 compares the estimated condition with the P-T estimates of metabasites possessing these assemblages for granulite, transitional amphibolite–granulite and amphibolite occurrences compiled by Pattison (2003). The estimated condition is similar to those of garnet + clinopyroxene + plagioclase ± hornblende ± quartz assemblages found in terranes of the granulite facies in the world. This suggests that the mylonite unit from the Takahama area would belong to the high-pressure granulite facies.

Gap in metamorphic grade between the mylonite unit and the crystalline schists

The P-T conditions of the crystalline schists of the main part of the Takahama metamorphic rocks have not been
well constrained. Nevertheless, the occurrence of the mineral assemblage, sodic amphibole + epidote + albite, suggests that the crystalline schists belong to the epidote–glaucophane schist subfacies, as described above. Evans (1990) examined the compositional effect on the stability field of sodic amphibole + epidote and revealed that the stability field expands towards both higher temperatures and lower pressures when sodic amphibole changes composition from glaucophane to crossite. The stability field, however, would not exceed 600°C at 1.0 GPa, even if the sodic amphibole were crossite. In addition, features such as the actinolite being stable in the basic schists and the presence of garnet in the pelitic schists but not biotite suggest that the metamorphic conditions of the crystalline schists are comparable with those of the garnet zone of the Sanbagawa Metamorphic Rocks where the estimated temperatures are below 500°C (Enami et al., 1994). We, therefore, conclude that a significant gap (or a steep gradient) exists in metamorphic conditions between the mylonite unit and the crystalline schists below. The muscovite K–Ar ages of the mylonite unit and the crystalline schists are similar, as described before, suggesting that both units were juxtaposed at a depth above the closure temperature of the muscovite K–Ar system.

The microstructures such as rutile armoured by sphene, and hornblende rimmed by actinolite may have been produced by the following reactions among the pre-existing matrix minerals:

\[
\begin{align*}
5\text{CaMgSi}_2\text{O}_8 + 3\text{TiO}_2 + \text{SiO}_2 + \text{H}_2\text{O} & \rightarrow 3\text{CaTiSiO}_3 + \text{Ca}_2\text{Mg}_5\text{Si}_8\text{O}_{22}(\text{OH})_2 + 22 \text{H}_2\text{O} \\
n & \text{sphe} + \text{tremolite} \\
\end{align*}
\]

and

\[
\begin{align*}
25\text{Na}_2\text{Si}_2\text{Al}_3\text{Si}_6\text{O}_{22}(\text{OH})_8 + 86 \text{SiO}_2 + 22 \text{H}_2\text{O} & \rightarrow 12\text{Ca}_2\text{Al}_2\text{Si}_2\text{O}_{10}(\text{OH})_8 + 7\text{Mg}_8\text{Al}_2\text{Si}_1\text{O}_{10}(\text{OH})_8 + 13\text{Ca}_2\text{Mg}_5\text{Si}_8\text{O}_{22}(\text{OH})_2 + 25\text{NaAlSi}_2\text{O}_6 \\
& \text{hornblende} + \text{epidote} + \text{chlorite} + \text{tremolite} + \text{albite} \\
\end{align*}
\]

The temperatures of reaction (2) range from 740 to 780°C at pressures of 1.0 to 1.2 GPa based on the thermodynamic calculation of Holland and Powell (1990). The corresponding reaction in the Fe-endmember system takes place at lower temperatures by ca. 20°C. Reaction (3) defines the low-grade limit of the amphibolite facies. Therefore, the temperatures of these two reactions that the garnet-clinopyroxene amphibolite underwent during retrograde metamorphism are significantly higher than those of the crystalline schists belonging to the epidote–glaucophane schist subfacies. These considerations support the conclusion shown above that the two units were metamorphosed under significantly different conditions.

**CONCLUSIONS**

The Takahama metamorphic rocks of western Kyushu consist of two units separated by shear zones and metamorphosed under different conditions. One unit comprises crystalline schist that have undergone the epidote–glaucophane schist subfacies metamorphism. The other is a mylonitized unit that occurs structurally at the top of the Takahama metamorphic rocks and was metamorphosed under high-pressure granulite facies conditions (ca. 1.1 GPa and 770°C). We consider that these two units were juxtaposed at a depth above the closure temperature of the muscovite K–Ar system.

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Garnet-clinopyroxene amphibolite from the Takahama metamorphic rocks


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