Peak metamorphic temperature of the Nishisonogi unit of the Nagasaki Metamorphic Rocks, western Kyushu, Japan

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The Nishisonogi unit of the Nagasaki Metamorphic Rocks represents a part of a Late Cretaceous subduction complex exposed in western Kyushu, Japan. We estimate peak metamorphic temperatures using a Raman carbonaceous material (CM) geothermometer on 60 pelitic schists. No systematic regional changes were observed in the mineral assemblage of samples collected over a large area (about 30 × 15 km), which include chlorite ± garnet + white micas + albite + quartz + titanite + CM. However, the estimated peak metamorphic temperature increases structurally upward from 440 to 524 °C, suggesting an inverted thermal gradient.

Keywords: Raman carbonaceous material geothermometer, Peak metamorphic temperature, Pelitic schist, Nagasaki Metamorphic Rocks

INTRODUCTION

The Nagasaki Metamorphic Complex consists of high-pressure/low-temperature (high-P/low-T) metamorphic rocks and is exposed in the Nishisonogi Peninsula (Nishisonogi unit), Nomo Peninsula (Nomo unit), and Amakusa-shimoshima Island (Amakusa-Takahama unit) in western Kyushu, Japan (Fig. 1a; e.g., Nishiyama et al., 2017). The geotectonic setting of the Nagasaki Metamorphic Rocks has long been debated (Karakida et al., 1969; Hirokawa, 1976; Hattori, 1992; Kouchi et al., 2011) because of their peculiar NS trend perpendicular to the general trend (EW) of the Southwest Japan. A detailed understanding of the local thermal structure offers insight into this question. Miyazaki and Nishiyama (1998) and Miyazaki et al. (2013) discussed the thermal structure of the Nomo and Amakusa-Takahama units, respectively. However, the thermal structure of the Nishisonogi unit remains poorly understood because the mineral assemblages are typically unsuitable for thermodynamic geothermobarometry and do not show systematic regional variation. A Raman carbonaceous material (CM) geothermometer has been recently calibrated for low- to medium-T metapelites (150–650 °C) (Beyssac et al., 2002; Rahl et al., 2005; Lahfid et al., 2010; Aoya et al., 2010; Kouketsu et al., 2014) and provides a powerful tool for studying the thermal structure of low-T metamorphic belts (Beyssac et al., 2004; Bollinger et al., 2004; Negro et al., 2006, 2013; Robert et al., 2010; Wiederkehr et al., 2011; Mathew et al., 2013; Yoshida et al., 2016). In this study, we determine the thermal structure of the Nishisonogi unit using Raman CM geothermometry.

GEOLOGICAL SETTING

The Nishisonogi unit contains pelitic schist, psammitic schist, mafic schist, and serpentinite (Fig. 1b). The schistosity suggests a broadly NS-trending antiform (i.e., Nishisonogi Antiform after Noda and Muta, 1957). The U-Pb ages of detrital zircon suggest a depositional age of <86 Ma (Kouchi et al., 2011). The K-Ar and 40Ar/39Ar ages of phengitic muscovite in the pelitic and psammitic schists are 85-60 Ma (Hattori and Shibata, 1982; Faure et al., 1988; Miyazaki et al., 2017). The schists were formed by epidote–blueschist facies metamorphism. The representative mineral assemblage of the pelitic schist is chlorite ± garnet + white micas (phengite ± paragonite) + albite + quartz + titanite + CM ± epidote. The representative mineral assemblage of the psammitic schist is chlorite ± garnet + white micas (phengite ± paragonite) + biotite + epidote + quartz + titanite + CM ± epidote.
mitic schist is albite + phengite + quartz ± chlorite. The psammitic schist is free from CM and locally contains glaucophane + epidote/piemontite + hematite + rutile ± garnet in addition to the above minerals. The representative mineral assemblages of the mafic schist are actinolite/barroisite + epidote + chlorite + albite ± quartz and glaucophane/barroisite + epidote + chlorite + albite + hematite ± quartz, with the former assemblage being dominant. There is a minor change in the mineral assemblages of the schists within the Nishisonogi unit (Nishiyama, 1989).

The peak metamorphic $P$–$T$ conditions of the Nishisonogi schists have been discussed in several previous studies. Shigeno and Uda (1995, 1997) and Moribe (2013) estimated $T$ of the pelitic schist to be 400–460 °C and 350–470 °C, respectively, using the garnet–muscovite geothermometer of Hynes and Forest (1988), and Miyazaki et al. (2018) estimated ~440 °C using the Raman CM geothermometer of Aoya et al. (2010). Moribe (2013) estimated $P$ of the mafic schist to be 1.3–1.4 GPa and $T$ ~ 500 °C using the garnet–clinoxyroxene–phengite geobarometer of Ravna and Terry (2004) and the garnet–clinoxyroxene geothermometer of Ellis and Green (1979).

**METHODS**

Pelitic schist samples were collected throughout the Nishisonogi Peninsula. Polished thin sections were analyzed using optical and Raman microscopy. Minerals were identified under transmitted and reflected light using an Eclipse E600 POL polarizing microscope (Nikon Corp., Tokyo, Japan). Raman spectra of the CM were obtained using a LabRAM HR800 spectrometer (HORIBA Jobin Yvon, Edison, NJ, USA) combined with a Fandango 50 diode–pumped solid–state laser (Cobolt AB, Solna, Sweden; $\lambda$ = 514.5 nm), a BX41 microscope (Olympus Optical Co. Ltd, Tokyo, Japan), and an LMPlanFL N 100× objective (Olympus Optical Co. Ltd, Tokyo, Japan; NA = 0.80) at Kumamoto University. The spectrometer was calibrated using the Si peak at 520 cm$^{-1}$. CM grains embedded within transparent minerals were examined using a 4-mW laser at the sample surface, a 1000–µm pinhole, 50–µm slit, 1800 lines/mm grating, 30–s acquisition time, and a spectral range of 1100–1800 cm$^{-1}$. No distinct alteration of the CM was observed under these settings during continuous analysis for up to 90 s. The CM Raman spectra were decomposed into several peaks using a computer program PeakFit 4.12 software (SeaSolve Software Inc., Framingham, MA, USA) with a pseudo-Voigt function (Kouketsu et al., 2014).

**RESULTS AND DISCUSSION**

**Mineral assemblage**

The mineral assemblages identified in the pelitic schist samples are chlorite ± garnet + white micas + albite +...
quartz + titanite + CM (Table 1 and Fig. 2a). Minor amounts of accessory tourmaline, clinozoisite, calcite, and zircon are present as matrix minerals. Garnet occurs commonly throughout the Nishisonogi Peninsula. There is no apparent systematic regional variation in the mineral assemblage, which is consistent with previous studies (e.g., Nishiyama, 1989).

Peak metamorphic temperature

The Raman spectra of CM grains in the samples generally show a D1-band at ~ 1350 cm⁻¹, G-band at ~ 1580 cm⁻¹, and a small D2-band at ~ 1620 cm⁻¹ (Fig. 3). D3- and D4-bands were not observed. The ‘fitting C’ methodology, as defined by Kouketsu et al. (2014), was used for peak fitting. The ‘fitting B’ was adopted for a few samples with an undetectable D2-band.

The peak metamorphic temperatures of the collected samples were estimated using Eq. (3) of Aoya et al. (2010):

\[ T^\circ\text{C} = 91.4 (R_2)^2 - 556.3 (R_2) + 676.3, \]

where the area ratio \( R_2 = \text{D1}/(\text{G} + \text{D1} + \text{D2}). \) This equation is optimized for regional metamorphism; improving on the method of Beyssac et al. (2002). The results are listed in Table 1. Most samples exhibit a unimodal temperature distribution and a mean within the range 438–522 °C (Fig. 4; see also supplementary material Figure S1: available online from https://doi.org/10.2465/jmps.190423). Some samples, which the area ratio \( R_2 = \text{D1}/(\text{G} + \text{D1} + \text{D2}). \) This equation is optimized for regional metamorphism; improving on the method of Beyssac et al. (2002). The results are listed in Table 1. Most samples exhibit a unimodal temperature distribution and a mean within the range 438–522 °C (Fig. 4; see also supplementary material Figure S1: available online from https://doi.org/10.2465/jmps.190423). Some samples,
however, show an additional small peak at 520–610 °C. Such bimodal distribution of the temperature (or CM crystallinity) has been reported by several previous studies (e.g., Wada et al., 1994; Kouketsu et al., 2019). Kouketsu et al. (2019) proposed two possible explanations for this phenomenon: (1) a mixture of detrital and metamorphic graphite grains, and (2) a mixture of different reactive CM during short-lived metamorphism. Another possibility is (3) amorphization due to brittle deformation of CM (Nakamura et al., 2015), which reduces the estimated temperature. In our case, the effect of the different reactive CM is probably negligible because the duration of metamorphism is estimated to be ~20 Myr (Miyazaki et al., 2017). This duration is probably sufficient to reach the steady state of CM crystallinity (cf. Mori et al., 2017; Nakamura et al., 2017). Moreover, the amorphization of CM is unlikely because hexagonal CM grains, indicating high crystallinity, are not found in our samples. The high-T CM grains are therefore considered detrital and do not represent the peak metamorphic temperature of the pelitic schist.

To reduce the influence of these high-T CM grains, we adopt median T values rather than the mean. The peak metamorphic temperatures of the samples are then estimated to range between 440 and 524 °C with a standard deviation (1σ) range of ±15–51 °C. The difference between the mean and median was less than 17 °C. Although the estimated peak metamorphic temperature is associated with a large error, significant variation is observed among the samples. Within the accuracy of the measurements, the results of this study are consistent with those of Shigeno and Uda (1995, 1997) and Miyazaki et al. (2018) (Fig. 2b). Moribe (2013) reported lower peak metamorphic temperatures than those found here although the reason remains unclear.

### Thermal structure

Our data show a regional variation in the peak metamorphic temperature (Fig. 2b). Lower T-values typically occur in the northwestern part of the Nishisonogi Peninsula. In particular, peak metamorphic temperatures below 460 °C dominate in the western limb of the Nishisonogi Antiform (Fig. 5). On the other hand, T >490 °C are confined to southeastern Nishisonogi Peninsula. The peak meta-

| Sample | Location | Mineral assemblage | R2c | T (°C)d |
|--------|----------|--------------------|-----|---------|
| H11    | 32°52′31.4″N 129°45′14.2″E | Grt + Chl | 0.347 ± 0.084 | 495 ± 42 |
| H13    | 32°50′32.3″N 129°45′26.8″E | Chl | 0.374 ± 0.063 | 482 ± 31 |
| H14    | 32°50′32.3″N 129°45′02.4″E | Grt + Chl | 0.389 ± 0.062 | 474 ± 30 |
| I04    | 32°59′09.6″N 129°46′12.9″E | Chl | 0.363 ± 0.059 | 487 ± 29 |
| I07    | 32°56′33.2″N 129°46′46.2″E | Chl | 0.405 ± 0.089 | 467 ± 43 |
| I09    | 32°54′41.2″N 129°46′32.9″E | Grt + Chl | 0.372 ± 0.071 | 483 ± 35 |
| I12    | 32°51′19.3″N 129°46′41.4″E | Grt + Chl | 0.399 ± 0.099 | 470 ± 49 |
| I14    | 32°49′13.6″N 129°46′35.9″E | Grt + Chl | 0.391 ± 0.083 | 473 ± 41 |
| I15    | 32°48′30.4″N 129°46′21.2″E | Chl | 0.347 ± 0.085 | 495 ± 42 |
| I16    | 32°47′30.0″N 129°46′50.7″E | Chl | 0.380 ± 0.067 | 479 ± 33 |
| J03    | 33°00′27.3″N 129°47′02.9″E | Chl | 0.432 ± 0.078 | 454 ± 37 |
| J05    | 32°58′17.9″N 129°47′22.6″E | Chl | 0.346 ± 0.075 | 495 ± 37 |
| J06    | 32°57′11.8″N 129°47′34.4″E | Grt + Chl | 0.356 ± 0.055 | 490 ± 27 |
| J09    | 32°54′48.8″N 129°47′26.7″E | Chl | 0.322 ± 0.070 | 507 ± 35 |
| J11    | 32°52′24.2″N 129°47′08.9″E | Chl | 0.398 ± 0.094 | 470 ± 46 |
| J13    | 32°50′11.4″N 129°47′03.5″E | Grt + Chl | 0.379 ± 0.086 | 479 ± 42 |
| J15    | 32°48′04.9″N 129°47′04.8″E | Chl | 0.293 ± 0.077 | 522 ± 38 |
| K05    | 32°58′43.0″N 129°48′08.2″E | Chl | 0.457 ± 0.041 | 441 ± 19 |
| K07    | 32°56′19.4″N 129°48′05.3″E | Chl | 0.383 ± 0.047 | 477 ± 23 |
| K10    | 32°53′18.0″N 129°48′29.6″E | Grt + Chl | 0.328 ± 0.069 | 504 ± 32 |
| K12    | 32°51′48.7″N 129°48′48.5″E | Grt + Chl | 0.364 ± 0.076 | 487 ± 37 |
| K14    | 32°49′11.0″N 129°48′01.9″E | Chl | 0.319 ± 0.056 | 508 ± 28 |
| K16    | 32°47′35.6″N 129°48′40.5″E | Grt + Chl | 0.324 ± 0.078 | 506 ± 39 |
| K17    | 32°46′29.9″N 129°48′01.8″E | Chl | 0.348 ± 0.079 | 495 ± 39 |
| L05    | 32°58′23.7″N 129°49′14.4″E | Grt + Chl | 0.357 ± 0.079 | 490 ± 39 |
| L13    | 32°50′30.4″N 129°49′15.0″E | Grt + Chl | 0.354 ± 0.060 | 491 ± 30 |

a Grt, garnet; Chl, chlorite; with white micas + albite + quartz + titanite + CM ± clinozoisite ± tourmaline ± calcite.
b Number of the analyzed grains.
c Area ratio: $R^2 = D1/(G + D1 + D2)$.
d Peak metamorphic temperature estimated using Eq. (3) of Aoya et al. (2010).
morphic temperature broadly increases from northwest to the southwest. However, despite the temperature difference, the mineral assemblage of the pelitic schist shows no systematic regional variation (Fig. 2). For example, our highest $T$-values are close to the temperature of the

Figure 2. (a) Mineral assemblages and sample numbers of the pelitic schists. (b) Peak metamorphic temperature of the pelitic schist samples. Values reported by previous studies are also shown. The gray areas indicate the distribution of the Nishisonogi unit.

Figure 3. Representative Raman spectra of CM in selected samples. The amplitude is not to scale. The temperature estimated from each spectrum is indicated in parentheses.

Figure 4. Histograms of temperature estimations for selected samples. (a) Unimodal distribution. (b) Bimodal distribution including a higher-$T$ peak (indicated by an arrow).
albite–biotite zone in the Sanbagawa belt (~ 520 °C; Enami et al., 1994), but no biotite was detected. Although the cause of this inconsistency is unclear, it could be related to variation in bulk chemical composition.

Notably, the higher-\(T\) area corresponds to the structurally upper layers of the Nishisonogi unit. Although there is a lack of information on the metamorphic pressure, these findings suggest an inverted thermal gradient in the Nishisonogi unit. An inverted thermal structure has been inferred in some low-\(T\) and high-\(P\) metamorphic belts. For example, higher-\(T\) rocks lie on the structurally upper part of the Sanbagawa metamorphic belt (Banno et al., 1986; Hara et al., 1990; Enami, 1994). Such a thermal structure may record the original thermal regime of the subduction zone or may be a consequence of the formation of a piled nappe structure during exhumation. The origin of the inverted thermal structure in the Nishisonogi unit is unclear. A boundary between the higher-\(T\) (>490 °C) and lower-\(T\) areas is not apparent (Fig. 2b) and no structural discontinuity has been reported between them. The inverted thermal structure described here should be interpreted on current evidence as a continuous temperature gradient within the Nishisonogi unit.

**CONCLUSIONS**

The mineral assemblage of pelitic schists within the Nishisonogi unit of the Nagasaki Metamorphic Rocks (western Kyushu, Japan) shows no systematic regional variation. However, Raman CM geothermometry results indicate peak metamorphic temperatures ranging from 440 to 524 °C. The peak metamorphic temperature increases structurally upward (northwest to southeast), which suggests an inverted thermal gradient.

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**SUPPLEMENTARY MATERIAL**

Figure S1 is available online from https://doi.org/10.2465/jmps.190423.

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