LETTER

Determination of SiO₂ Raman spectrum indicating the transformation from coesite to quartz in Gföhl migmatitic gneisses in the Moldanubian Zone, Czech Republic

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The Raman spectroscopy of more than 2000 SiO₂ inclusions in zircon separates from Gföhl migmatitic gneisses in the Nové Dvory area shows that most of the SiO₂ inclusions are composed of quartz with clear and intense peaks at 464, 393, 266, 207 and 125 cm⁻¹. It also reveals that a few SiO₂ inclusions have a weak but clear peak at 521 cm⁻¹, which is the most fundamental vibration of coesite, along with typical quartz vibrations mentioned above. The Raman spectrum is composed of the intense vibrations of quartz at 464, 393 and 266 cm⁻¹ of quartz and the weak vibration of coesite at 521 cm⁻¹ is obtained from the quartz proximal to the relict coesite inclusion in the pyrope of ultra-high-pressure (UHP) rocks in Dora Maira Massif. A similar Raman spectrum has been obtained for quartz transformed from coesite in UHP rocks recovered from the CCSD drillhole of the Sulu belt, eastern China. Therefore, we propose that the SiO₂ phase whose Raman spectrum shows a weak vibration at 521 cm⁻¹ existed as coesite in the past.

Keywords: Raman spectroscopy, Coesite, Quartz, Gföhl unit, Czech Republic

INTRODUCTION

Recent petrographic studies of the metamorphic rocks exposed in the root zone of the present and past orogenic belts have revealed that the occurrence of ultra-high-pressure (UHP) rock is a common phenomenon in the continent-continent collision setting (e.g., Carswell and Compagnoni, 2003). In these belts, UHP evidence mainly exists in eclogitic rocks and garnet-peridotites that commonly occur as intercalating layers and/or lenticular bodies within the felsic gneisses, which are mainly composed of amphibolite-facies mineral assemblages in their matrix. Such a mode of occurrence of UHP rocks is advantageous for the “exotic” school on the origin of the eclogite. However, micro-Raman spectroscopy recently performed in the Dabie-Sulu area (e.g., Ye et al., 2000; Liu et al., 2002) has successfully determined significant UHP evidences such as tiny inclusions of coesite and jadeite from

the zircons hosted by country felsic gneisses; this finding suggests that the felsic gneiss at least near the UHP rocks in the Dabie-Sulu area underwent UHP metamorphism on a regional scale, i.e., this is one of the potential evidences of the “in situ” model (e.g., Hirajima and Nakamura, 2003).

In the Variscan orogenic belt, diamond-bearing metapelite has been found in the Saxothuringian Zone (Erzgebirge; Massonne, 1999; Nasdala and Massonne, 2000; Massonne and O’Brien, 2003); however, the UHP evidence has not been found in metamorphosed supracrustals in the Moldanubian Zone in the Czech Republic thus far, except for garnet peridotite and some peculiar minerals/rocks such as Cpx with K-feldspar lamellae in Lower Austria (Becker and Altherr, 1992), pyrope/Opx-bearing quartzite (Kotková et al., 1997), and garnet with a minor but significant amount of Na₂O (= 0.18 wt%) and TiO₂ (0.5 wt%) in granulite (Vrána and Frýda, 2003). In the Nové Dvory area of the Moldanubian Zone, garnet peridotite and associated kyanite-eclogite, showing peak *P-T* conditions of 4.5–5.0 GPa and > 1000 °C (Medaris et al.,

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Figure 1. Geological sketch map of the Nové Dvory–Mohelno area (Medaris et al., 1990; Nakamura et al., 2004). The zircons used in this study were extracted from migmatitic gneiss around the Nové Dvory peridotite body in the Gföhl Unit.

1990; Nakamura et al., 2004), occur as a lenticular body (approximately 1 km × 3 km) surrounded by Gföhl migmatitic gneisses (Fig. 1), from which no HP/UHP evidence has been reported, except for the occurrence of kyanite. Therefore, there is a significant pressure gap between UHP ultramafic/mafic rocks and the surrounding gneiss. Such a geological setting is similar to the mode of occurrence of UHP rocks in the regional gneiss terranes mentioned above. To determine the *P–T* history of Gföhl migmatitic gneiss and to consider the timing of juxtaposition of crustal materials and mantle rocks, we have carried out a petrological study in the Moldanubian Zone in the Czech Republic. We have found a peculiar Raman spectrum suggesting the transformation of coesite to quartz from a few SiO₂ inclusions in the zircons extracted from the migmatitic gneisses of the Nové Dvory area. In this paper, we provide a detailed explanation on this peculiar Raman spectrum. Mineral abbreviations are after Kretz (1983).

GEOLOGICAL SETTING AND PETROGRAPHY

The Moldanubian Zone of the Bohemian Massif represents a Variscan tectonic assemblage of medium to high-grade metamorphic associations with mineral assemblages in amphibolite and granulite facies, which are intruded by numerous granitoids. The Gföhl Unit is mainly composed of the garnet-, kyanite-, and biotite-bearing felsic granulite (upper) units along with minor amounts of pyroxene-bearing granulite and the lower migmatitic gneiss unit (e.g., Carswell and O'Brien, 1993). Peridotite

bodies sporadically occur both in the Gföhl granulite and Gföhl migmatitic gneiss. Eclogites in the Gföhl Unit are associated with the peridotite bodies, except for one locality near Spačice (Medaris et al., 1998). Previous studies show that the Gföhl granulites have experienced a clockwise *P–T* path with peak conditions of approximately 1.6–2.0 GPa and 900–1100 °C, followed by nearly isothermal decompression to medium-*P* and high-*T* conditions (0.6–0.8 GPa, 700–800 °C) (Vrána, 1989; Carswell and O'Brien, 1993; Cooke, 2000; Cooke and O'Brien, 2001). In contrast, thermodynamic modeling by Štípská and Powell (2005) has yielded a metamorphic peak temperature of 750–850 °C at 1.6–1.8 GPa that is distinctly lower than that mentioned in previous studies. Prince et al. (2000) and Svojtka et al. (2002) reported the Sm–Nd garnet ages of 354 ± 6 Ma as the high-*P* and high-*T* phases of granulite evolution. Sm–Nd dating of the prograde zoned garnets obtained from felsic granulite also constrains their minimum crystallization age to 354 Ma (Faryad and Košler, 2007). However, Kröner et al. (2000) reported a concordant U–Pb age of 339.8 ± 2.6 Ma for zircon in granulites in the Czech Republic. Inherited zircons in the Moldanubian granulites have yielded the Paleozoic age maxima at ca. 360–400 Ma and 450–470 Ma as the protolith of the granulite (Kröner et al., 2000). Medaris et al. (2005) has summarized the available Sm–Nd ages of the peridotites and associated HP mafic rocks from the Gföhl Unit. The garnet peridotites constrain the cooling ages averaging 339 ± 10 Ma, and some of the garnet pyroxenite and eclogites yielded a similar mean Sm–Nd age of 336 ± 7 Ma; however, several samples are older, ca. 370–380 Ma.

Nové Dvory is located in the southeastern part of the Czech Republic, where a peridotite body intercalating with eclogite layers with/without kyanite occurs within the migmatitic gneiss of the Gföhl Unit (Medaris et al., 1990; Nakamura et al., 2004) (Fig. 1). Gföhl migmatitic gneisses show various types of partial melting structures such as leucosomes, parallel to foliation or existing in interboudin partitions, and leucocratic veins. These rocks are mainly composed of garnet, biotite, K-feldspar, plagioclase, and quartz with minor amounts of kyanite, sillimanite, spinel, and zircon. Both kyanite and sillimanite occur in the matrix. Kyanite is commonly surrounded by plagioclase and is rarely in direct contact with quartz (Fig. 2a). Sillimanite (fibrolite) occurs in a composite pool that is elongated parallel to the migmatitic foliation along with biotite and quartz (Fig. 2b). Most of the garnets are fine-grained (< 1 mm in diameter). However, some of them are coarse-grained (up to 3 mm). The latter type is abundant in inclusion phases, which are mostly quartz and rarely antiperthitic plagioclase, rutile, and kyanite. Fine-

Figure 2. (a) A typical macroscopic texture of the Gföhl migmatitic gneiss. (b) Photomicrograph of kyanite surrounded by plagioclase in migmatitic gneiss. Plane-polarized light. (c) Photomicrograph of sillimanite (fibrolite) in a compositional pool along with biotite and quartz in migmatitic gneiss. Plane-polarized light. (d) Cathodoluminescence image of host zircon grain, including the SiO₂ inclusion, extracted from a migmatitic gneiss.

grained garnets consist of Ca-poor almandine with a composition range of $\text{alm}_{77-84}\text{prp}_{8-16}\text{grs}_{2-4}\text{sps}_{7-8}$. However, some coarse-grained garnets show chemical zoning, defined by Ca- and Mg-rich cores ($\text{alm}_{72-75}\text{prp}_{13-16}\text{grs}_{6-9}\text{sps}_{3-4}$) and a Ca-poor rim ($\text{alm}_{77-80}\text{prp}_{12-17}\text{grs}_{2-4}\text{sps}_{3-4}$). The Fe/Mg ratio is almost constant in the core; however, it increases toward the outermost rim of the coarse-grained garnets. The rim compositions of the coarse-grained garnet are identical to those of the fine-grained garnets. The chemical characteristics of the coarse-grained garnets are identical to those of Gföhl felsic rocks in Lower Austria (Cooke and O'Brien, 2001), although the grossular content exhibits a bell-shaped pattern in the core of the studied samples. Plagioclase and biotite are rather homogeneous in each crystal. However, their compositions are slightly different, correlating with the mode of occurrence, such as $\text{An}_{20-24}\text{Ab}_{73-79}\text{Or}_{1-2}$ for the matrix phase and $\text{An}_{18-23}\text{Ab}_{73-81}\text{Or}_{1-3}$ for the inclusion phase in the garnet, and $\text{Mg}\# [= \text{Mg}/(\text{Fe} + \text{Mg})]$ of biotite ranges from 0.30 to 0.42 for the matrix phase and from 0.38 to 0.47 for the inclusion phase in the garnet.

RAMAN SPECTROSCOPY AND CATHODOLUMINESCENCE (CL) OBSERVATION

More than 2000 SiO₂ micro-inclusions in zircon separates obtained from 10 samples of Gföhl migmatitic gneisses in the Nové Dvory area were analyzed using a laser Raman spectrophotometer JASCO NRS-3100 (Jasco, Tokyo, Japan) at Kyoto University using the 514.5 μm line of an

Ar-ion laser at 30 mW and a spot size of 1.0 μm . The spectrograph was obtained using 1800 groove/mm holographic grating. The accumulation times were between 30 and 60 s. Calibration was performed using 520 cm^{-1} Si-wafer band and neon (Ne) spectrum.

Because of the limited size of the SiO₂ inclusions, most of the Raman spectrograph is a mixture of signals from the SiO₂ inclusions and host zircon. Host zircons show clear and intense peaks at 1004, 972, 438, 355, 224 and 202 cm^{-1} (Fig. 3). Both quartz and coesite show a typical Raman peak at 355 cm^{-1} (Hemley, 1987). However, the 355 cm^{-1} Raman peak of the SiO₂ phases is masked by an intense zircon Raman peak at the same position in this study (Fig. 3). After the overlap of the zircon peak, it is found that most SiO₂ inclusions are composed of quartz with clear and intense peaks at 464, 393 and 266 cm^{-1} ; however, a few SiO₂ inclusions obtained from 4 samples show a weak but clear peak at 521 cm^{-1} , which is the most fundamental vibration of coesite, along with the typical quartz vibrations mentioned above (Fig. 3).

To consider the significance of the Raman spectrum composed of a weak peak at 521 cm^{-1} and intense peaks at 464, 393 and 266 cm^{-1} , Raman spectroscopy was performed on a coesite-quartz composite inclusion present in a pyrope of Dora-Maira UHP rock (Fig. 4). The relic coesite shows a Raman spectrum with a strong 521 cm^{-1} peak and weak 462, 355, 270 and 204 cm^{-1} peaks (1 in Fig. 4b). The proximal quartz (approximately up to 50 μm away from the coesite-quartz boundary) shows a Raman spectrum with a strong 464 cm^{-1} peak and weak 521 cm^{-1}

Figure 3. (a) Raman spectra of a common (bottom) and rare (upper) SiO₂ inclusion in zircon. (b) The enlarged version of the boxed part in (a).

peaks (2 in Fig. 4b); however, the distal quartz (approximately greater than 50 μm from the coesite-quartz boundary) does not show the 521 cm⁻¹ peak (3 in Fig. 4b). These data suggest that the SiO₂ phase showing the Raman spectrum with a strong 464 cm⁻¹ peak and weak 521 cm⁻¹ peaks in the Gföhl migmatitic gneiss may be once coesite. A similar Raman spectrum has been obtained for quartz transformed from coesite in UHP rocks recovered from the CCSD drillhole of the Sulu belt, eastern China (Liu et al., 2002), and in eclogite from the Lanterman Range, Antarctica (Ghiribelli et al., 2002).

To consider the trapped timing of the SiO₂ inclusions in zircon, optical and cathode luminescence (CL) image observations were carried out. This result revealed that

Figure 4. (a) Photomicrograph of quartz-coesite composite inclusion in pyrope-quartz schist of the Brossasco-Isaska unit of the Dora Maira Massif. Plane-polarized light. (b) (1) Raman spectrum of relict coesite in pyrope-quartz schist in the Dora Maira Massif. (2) Raman spectrum of the quartz proximal to relict coesite in pyrope-quartz schist in the Dora Maira Massif. (3) Raman spectrum of the quartz distal to relict coesite in pyrope-quartz schist in the Dora Maira Massif.

most zircon grains were prismatic and exhibited oscillatory zoning from the core to the rim. No clear discontinuous/irregular pockets indicating the later stage growth/replacement of the host zircon crystals (e.g., Gebauer et al., 1997) were detected (Figs. 2c and 2d). All possible coesite

inclusions were detected from the core, although all the SiO₂ inclusions in the rim were composed of quartz. This observation is in contrast to the normal observation in the other UHP terranes, including the Kaghan Valley in the Pakistan Himalaya (Kaneko et al., 2003). This issue is beyond the scope of this paper, and it will be discussed elsewhere.

DISCUSSION

Why is a weak 521 cm⁻¹ peak identified in the quartz transformed from coesite? We tentatively propose two plausible reasons: (1) an effect of trapped strain due to the phase transformation from coesite to quartz (e.g., Ikuta et al., 2007) and (2) existence of sub-microscopic coesites in the transformed quartz. Ikuta et al. (2007) have succeeded in determining the crystal structure of coesite and its transformed polymorph, quartz, in a UHP eclogite of Sulu belt, China, by the newly developed in situ X-ray diffraction method using a rock thin section mounted on a glass slide. They also pointed out that the displacement ellipsoid for the quartz transformed from coesite is significantly larger than that of the previously reported natural quartz at room temperature and have envisaged that this may be caused by the trapped strain due to the phase transformation from coesite to quartz. The peak height ratio of 521 cm⁻¹/464 cm⁻¹ of the transformed quartz gradually decreases from the quartz near the coesite grain to the quartz at the outermost rim of the quartz-coesite composite SiO₂ inclusion (Kobayashi, unpublished data). This could suggest that the trapped strain is higher in the proximal quartz nearby the coesite than the distal quartz. If so, the Raman shift of the representative band such as 464 and/or 393 cm⁻¹ will be expected. However, we did not detect such a Raman shift among the proximal and distal quartz in our samples. Alternative idea is what sub-microscopic size coesite still remains in the quartz nearby the transformation boundary between coesite and quartz by some kinetic reason. Monsenfelder et al. (2005) pointed out that the infiltration of H₂O occurring under conditions with relatively higher *T* critically enhanced the transformation from coesite to quartz rather than the rapid exhumation; however, this is not the case for infiltration under lower-*T* conditions. The thermodynamic calculation and synthetic experiments for high-*P* and high-*T* granulite (White et al., 2001; Tropper et al., 2005) and the inferred *P-T* history of the studied rock, as mentioned later, suggest that the studied rock was almost free from the hydrous phase during the high-*P* and high-*T* granulite stage, 800–1200 °C and 1.2–1.6 GPa. Then, the biotite is formed from garnet + other phases + infiltrated H₂O or partial melt at the later stage of the decompression history under

Figure 5. Newly proposed *P-T* path for the Gföhl gneiss in this study. For comparison, the *P-T* path of the Gföhl granulite (Carswell and O'Brien, 1993; Cooke and O'Brien, 2001) and the peak *P-T* conditions of garnet peridotite and kyanite-eclogite of the Nové Dvory body (Medaris et al., 1990; Nakamura et al., 2004) are inserted. *P-T* conditions inferred from the garnet core and the matrix stages of the Gföhl gneiss are also inserted. Grt-Bt geothermometer; Ia: Ferry and Spear (1978); Ib: Hodges and Spear (1982); Ic: Dasgupta et al. (1991); GASP geobarometer; IIa: Koziol and Newton (1988); IIb: Hodges and Spear (1982); and IIc: Perchuk et al. (1985).

700–880 °C and 0.4–0.9 GPa. The temperature condition, under which H₂O infiltration/partial melting occurred, should be sufficiently high to transform coesite to quartz, even in zircon grains.

On the basis of the mineral composition and texture, the following two equilibrium stages are identified in the Gföhl migmatitic gneiss: a Ca-rich garnet core stage and a matrix stage. The Ca-rich core of some coarse-grained garnet may coexist with K-feldspar, plagioclase, biotite and kyanite. The mineral assemblage of the matrix stage can be identified according to the presence of Ca-poor garnet, K-feldspar, plagioclase, biotite and sillimanite. Cordierite has not been detected in the studied samples. The pressure calculated by the garnet-plagioclase-quartz-Al₂O₃ (GASP) geobarometer (Koziol and Newton, 1988; Hodges and Spear, 1982; Perchuk et al., 1985), utilizing the most Ca-rich garnet core composition and plagioclase inclusion in garnet, provided the pressure conditions of 1.2 GPa at 800 °C and 1.6 GPa at 1000 °C. The coarse-grained garnet shows a bell-shaped pattern of grossular content in the core that may be reset during the later stage of the LP/HT event. Therefore, the estimated pressure may represent a minimum value for the early stage. The *P-T* conditions estimated from the matrix mineral assemblage are 700–880 °C and 0.4–0.9 GPa; these conditions are obtained using a combination of the garnet-biotite (Grt-Bt) geothermometer (Ferry and Spear, 1978; Hodges

and Spear, 1982; Dasgupta et al., 1991) and GASP geobarometer. The estimated P - T conditions for the matrix stage are almost identical to those for the amphibolite-facies stage of the Gföhl migmatitic gneiss in Lower Austria (Carswell and O'Brien, 1993; Cooke, 2000; Cooke and O'Brien, 2001) (Fig. 5). Such a P - T path is apparently identical to that of the granulite/gneiss of the Gföhl Unit in Lower Austria (Cooke and O'Brien, 2001). However, we have found a few SiO_2 inclusions suggesting the transformation from coesite to quartz by using the Raman spectrum in the Gföhl migmatitic gneiss at Nové Dvory. From this fact, we may infer that the Gföhl migmatitic gneiss once underwent UHP metamorphism before the HP-granulite stage.

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