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LETTER

Unusually low $\delta^{18}\text{O}$ ultra-high-pressure metamorphic rocks from the Sulu Terrain, eastern China

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Abstract—Eclogite and quartzite interlayers, containing ultra-high-pressure (UHP) metamorphic mineral assemblages, occur at Qinglongshan in the Sulu terrain, eastern China. These rocks exhibit some of the most negative oxygen isotope compositions that have ever been reported for metamorphic rocks. The $\delta^{18}\text{O}$ value of minerals in eclogite ranges from -10.4 to -9.0‰ and in quartzite ranges from -10.2 to -7.3‰ . Near isotopic equilibrium between minerals and rocks suggests that these rocks must have acquired such low oxygen isotope compositions before the UHP metamorphism, through near surface meteoric water-rock interactions. The rocks were then subducted to a great depth during continent-continent collision, forming UHP minerals with concomitant local oxygen isotope redistribution. It is significant that the rocks have retained such negative oxygen isotope compositions despite subduction into the upper mantle.

INTRODUCTION

The identification of coesite, coesite pseudomorphs, and diamond as tiny inclusions in zircon, garnet, omphacite, kyanite, epidote, and zoisite in eclogite and other metamorphic rocks of supracrustal origin is certainly one of the most important discoveries in metamorphic petrology in the past fifteen years (Liou et al., 1994, and the references cited therein). The ultra-high metamorphic pressures (>25 kbar) indicated by the presence of coesite and/or diamond are far greater than previously documented and significantly expand the known depth limits (>80 – 90 km) of metamorphic material now exposed at the Earth's surface. The coesite/diamond-bearing rocks, therefore, define a new ultra-high-pressure (UHP) metamorphic facies. Petrological as well as experimental studies on these rocks demonstrate that the stabilities of the UHP phases require abnormally low geothermal gradients, approximately $7^\circ\text{C}/\text{km}$, which can be attained only by subduction of a cold oceanic crust with pelagic sediments or old, cold continental crust (Schreyer, 1988; Liou et al., 1994). In this letter, we will show that some UHP rocks from eastern China, even having been subducted to mantle depths, can still retain negative oxygen isotope compositions, which were suggested as resulting from meteoric water-rock hydrothermal reactions prior to UHP metamorphism.

GENERAL GEOLOGY AND SAMPLE DESCRIPTION

The UHP rocks, including eclogite, marble, and gneiss, in east-central China are distributed in the Dabie and the Sulu terrains. These two terrains were separated by the left-lateral Tanlu fault for a distance of at least 500 km (Fig. 1), probably during the Cretaceous and/or Cenozoic time. Inclusions of coesite and coesite pseudomorphs are rare but widespread in both terrains (Wang et al., 1989; Okay et al., 1989; Yang and Smith, 1989; Hirajima et al., 1990; Wang

and Liou, 1991; Xu et al., 1992; Zhang et al., 1994). Diamond inclusion, however, was only reported from the Dabie area (Xu et al., 1992; Okay, 1993). The protoliths of these UHP rocks are considered to be Precambrian (Li et al., 1993). The estimated P - T conditions for these UHP rocks span a rather wide range: $P = 20$ – >35 kbar and $T = 550$ – 890°C (e.g., Wang et al., 1992; Enami et al., 1993; Okay, 1993; Zhang et al., 1994). These UHP rocks were interpreted as resulting from collision of the Sino-Korean and Yangtze cratons during 200 – 240 Ma (Li et al., 1989a,b, 1992, 1993, 1994; Ames et al., 1993; Eide et al., 1994).

The samples in the present study were collected from the Qinglongshan in the Sulu terrain (Fig. 1). Sample QL-1 is an eclogite while sample SQ-92-3 is a quartzite, which occurs as thin interlayers within the bottom part of the eclogite body. The thickness of the eclogite and quartzite interlayers varies from centimeters to meters with total thickness less than 5 m. The thickness of the entire eclogite body is about 15 m but the length is not known due to poor exposure. The relative abundance (by volume) of quartzite to eclogite is about 1 to 6 . This rock body is supposed to be embedded within the surrounding quartzofeldspathic gneiss, although the exact contact relation was not observed. The eclogite mainly consists of garnet, omphacite, and phengite, with minor amounts of rutile, kyanite, epidote, quartz, and talc. The quartzite, however, contains quartz, phengite, kyanite, garnet, epidote, and rutile. Retrograde alteration is not prominent, although incipient symplectitic replacement around omphacite grains is observed. Coesite and coesite pseudomorphs as mineral inclusions were reported in both rock types (Zhang et al., 1994). Both rock samples are foliated with phengite mainly developed along the foliation. Porphyroblastic epidotes with inclusions of coesite pseudomorphs cut the foliation, suggesting that UHP deformation occurred before the growth of epidote in the coesite stability field (Zhang et al., 1994).

ANALYTICAL METHOD

Oxygen was extracted from mineral separates, following the method described by Rumble and Hoering (1994), with the use of a stable isotope laser probe located at the Geophysical Laboratory. A CO_2 laser beam was employed to burn the mineral grains completely

FIG. 1. Simplified geologic map showing the Dabie and the Sulu ultra-high-pressure terrains in east-central China. The eclogite and quartzite samples in this study were collected from the Qinglongshan in the Sulu terrain.

in a BrF_5 atmosphere. Oxygen gas, instead of CO_2 gas, was collected for oxygen isotope measurements. This method avoids the possible isotopic fractionation during the O_2 - CO_2 conversion reaction as documented by Mathey and Macpherson (1993). The precision of the analyses is generally better than $\pm 0.2\text{‰}$. The isotopic reference material NBS-28 has a $\delta^{18}\text{O}$ value of $+9.3 \pm 0.2\text{‰}$, for 4 aliquots (Rumble and Hoering, 1994).

Each mineral separate in this study was analyzed at least twice (Table 1) and all yielded similar results. Two mineral samples, 71-59B quartz and KBH-1 olivine, that have been analyzed conventionally and by laser fluorination were employed as intralaboratory standards. The results obtained in the present work are comparable to previous analyses (Table 2).

RESULTS AND DISCUSSION

The results of the analyses are shown in Table 1. $\delta^{18}\text{O}$ values of minerals in eclogite range from -10.4 to -9.0‰ and

in quartzite range from -10.2 to -7.3‰ . The whole-rock oxygen isotope compositions of eclogite and quartzite were not analyzed but may be very close to that of garnet and quartz, respectively, because these two minerals are the major component in the respective rock sample. The unusually low $\delta^{18}\text{O}$ values of these two samples are rather astonishing. As a comparison, Sharp et al. (1993, Fig. 7) compiled all the published $\delta^{18}\text{O}$ values of quartz from various metamorphic rocks and showed that none of the metamorphic quartz has $\delta^{18}\text{O}$ value lower than $+5\text{‰}$. In addition, published $\delta^{18}\text{O}$ values for all eclogites are also larger than zero, ranging from $+1.5$ to $+12.0\text{‰}$ (Taylor and Coleman, 1968; Vogel and Garlick, 1970; Garlick et al., 1971; Desmons and O'Neil, 1978; Matthews and Schliestedt, 1984; Agrinier et al., 1985; MacGregor and Manton, 1986; Ongley et al., 1987; Schliestedt and Matthews, 1987; Nadeau et al., 1993; Jacob et al., 1994). It is noted that the $\delta^{18}\text{O}$ values of most eclogites from the Dabie and the Sulu areas also fall into the above reported range (Xie, 1993; Baker et al., 1994; Yui et al., 1994).

Table 1. Oxygen isotope compositions of mineral separates and estimated temperatures

Sample	Mineral	$\delta^{18}\text{O}(\text{‰})$	$\Delta(\text{pairs})$	T(°C) ¹				
				R-H	H-H-F	J	M	Z
QL-1	phengite	$-9.0 \pm 0.3(3)^2$	ph-om ³	42	-31	684		521
eclogite	omphacite	$-10.2 \pm 0.3(6)$	ph-gar	488	515	649		686
	garnet	$-10.4 \pm 0.1(4)$						
SQ-92-3	quartz	$-7.3 \pm 0.1(2)$	qz-ph	871	1023	775		879
quartzite	phengite	$-8.7 \pm 0.0(2)$	qz-ky	738	990			726
	kyanite	$-9.1 \pm 0.0(2)$	qz-ep	721			640	615
	epidote	$-10.0 \pm 0.1(2)$	qz-gar	706	781	723	749	781
	garnet	$-10.2 \pm 0.0(2)$	ph-gar	470	489	596		649

1. Temperature estimates are based on the fractionation relations proposed by: R-H, Richter and Hoernes (1988); H-H-F, Hoffbauer et al., (1994); J, Javoy (1977); M, Matthews (1994); and Z, Zheng (1993a, 1993b).

2. Number in parentheses gives number of analyses.

3. Abbreviations: ep, epidote; gar, garnet; ky, kyanite; om, omphacite; ph, phengite; qz, quartz.

Table 2. Oxygen isotope compositions of intra-laboratory standards

Sample	$\delta^{18}\text{O}(\text{‰})$ (this study)	$\delta^{18}\text{O}(\text{‰})$ (previous average)
71-59B	$10.19 \pm 0.33(6)^1$	$10.40 \pm 0.30(3)^1$
quartz		
KBH-1	$5.25 \pm 0.11(5)^1$	$5.30 \pm 0.19(4)^1$
olivine		

1. Number in parentheses gives number of analyses.

FIG. 2. The isotherm plot of mineral separates from sample QL-1 (eclogite) and SQ-92-3 (quartzite). The data points should fall on a straight line if they are in isotopic equilibrium (Javoy et al., 1970). The a and b are coefficients of the fractionation relation $\Delta^{18}\text{O} (\text{mineral-garnet}) = b + a(10^6/T^2)$ reported by Javoy (1977). The slope of the regression line corresponds to a temperature of $709 \pm 33^\circ\text{C}$.

Compared with the limited $\delta^{18}\text{O}$ range (i.e., from +5 to +6‰) of mantle rocks (Taylor, 1968), the published positive and large range of $\delta^{18}\text{O}$ value for eclogite mentioned above was attributed to (1) high-pressure fractional crystallization (Garlick et al., 1971), (2) isotopic exchange with (meta)sedimentary rocks (Vogel and Garlick, 1970; Desmons and O'Neil, 1978), or (3) interaction with meteoric waters (Vogel and Garlick, 1970). In the light of oxygen isotope studies of oceanic lithosphere and ophiolites (e.g., Muehlenbachs and Clayton, 1972a,b; Spooner et al., 1974; Gregory and Taylor, 1981), however, these "anomalous" oxygen isotope compositions of eclogites can now be more properly interpreted as largely resulting from metamorphism of hydrothermally altered oceanic crust (Gregory and Taylor, 1986; MacGregor and Manton, 1986; Ongley et al., 1987). The oceanic crust may be the most probable protolith of the eclogite. The positive and large $\delta^{18}\text{O}$ range of eclogite can then be accounted for by hydrothermal reactions between seawater and oceanic crust under different temperature conditions with variable water/rock ratios (e.g., Elthon, 1981). Such reactions, of course, took place before subduction. Because the oxygen isotope composition of seawater may not deviate too much from that of the present-day ocean during geologic time (e.g., Muehlenbachs, 1986) and because prograde metamorphic effects, even considering the possible pressure effect on isotopic fractionations (Polyakov and Kharlashina, 1994), normally do not lower the $\delta^{18}\text{O}$ value of a rock by more than 1–2‰ (Valley, 1986), the scenario of seawater-oceanic crust hydrothermal reaction obviously cannot explain the eclogite with the unusually negative $\delta^{18}\text{O}$ value reported here. Hydrothermal exchange with meteoric waters under high temperatures and high water/rock ratios at shallow crustal levels must be invoked as a working hypothesis. The Holocene basalt in Iceland (Hattori and Muehlenbachs, 1982) or the Tertiary rift systems of the proto-North Atlantic (e.g., Taylor and Forester, 1979) may be a good example as the possible analog for this proposed hydrothermal system. In the former example, the drill cuttings from the active Krafla central volcano in Iceland show an $\delta^{18}\text{O}$ depletion trend with depth. The lowest $\delta^{18}\text{O}$ value of -10.5‰ was observed for basalt at a depth of 2100–2150 m. The $\delta^{18}\text{O}$ value

of local meteoric water ranges from -10 to -13‰ . Model calculations showed that the hydrothermal alterations for the Krafla basalt at the depth around 2100 m might have occurred at $300\text{--}400^\circ\text{C}$ with a water/rock ratio larger than 50.

Considering the geologic situations in the Qinglongshan area, there are only two settings where the proposed meteoric water-rock hydrothermal reactions could have occurred. First, there are numerous Jurassic-Cretaceous granitic intrusions postdating continental collision in the Sulu terrain (Lin et al., 1994). These granitic intrusions could have induced large-scale hydrothermal systems (e.g., Taylor, 1977), which may have caused severe meteoric water-rock interactions for the nearby eclogite. If this is the case, the negative $\delta^{18}\text{O}$ value of eclogite reported here would have nothing to do with the UHP metamorphism. However, such extreme ^{18}O depletion could not have occurred without the complete obliteration of the UHP mineral assemblages. Alternatively, and more likely, the negative $\delta^{18}\text{O}$ value of eclogite might have been caused by meteoric water-rock reactions prior to UHP metamorphism. The igneous activity, which caused this hydrothermal system, may or may not be genetically related to the protoliths of the eclogite. In any case, the protoliths were then subducted to great depths with concomitant oxygen isotope redistribution among newly formed UHP minerals. The fact that these rocks still retain negative oxygen isotope compositions would imply an extremely limited isotopic exchange between the subducted rocks and their surroundings during the UHP metamorphism.

Quartzite, interlayered with the eclogite in the present study, also shows negative oxygen isotope composition (Table 1). The ^{18}O -enrichment trends for minerals in both eclogite and quartzite samples are in correct order for equilibrium fractionation (e.g., Richter and Hoernes, 1988; Zheng, 1993a,b; Hoffbauer et al., 1994). The $\delta^{18}\text{O}$ values of phengite and garnet from quartzite are almost identical to those from eclogite (Table 1). These may indicate a near oxygen isotope equilibrium within and between rock samples over scales of a few meters. In the isotherm plot of Javoy et al. (1970; Fig. 2), the minerals from both eclogite and quartzite closely fall on a straight line, depicting near isotopic equilibrium. Depending on the geothermometers employed, the estimated

temperatures, however, cover a very wide and unrealistic range (Table 1), especially for those temperature estimates involving phengite and omphacite derived from the theoretical fractionation relations suggested by Richter and Hoernes (1988) and Hoffbauer et al. (1994). Possible drawbacks, which may limit the reliability of the temperature estimates, of the theoretical calculations proposed by these workers were discussed by Zheng (1993a). The calibrated fractionation relations given by Javoy (1977) and Matthews (1994) and the theoretical ones postulated by Zheng (1993a,b), on the other hand, yield a more consistent and narrower temperature range, i.e., from 521 to 879°C (Table 1). Considering the possible stoichiometric effect on oxygen isotope compositions of omphacite, phengite, epidote, and garnet (e.g., Matthews et al., 1983), this temperature range seems reasonable. The isotherm plot of Fig. 2 also yields a temperature estimate of 709°C $\pm$ 33°C. These estimates are slightly lower than the proposed temperature conditions for the UHP peak metamorphism derived from cation partitioning between coexisting phases for the study area (i.e., 700–890°C, Zhang et al., 1994), a common phenomenon generally interpreted as resulting from retrograde oxygen exchange during the cooling of the rock (e.g., Matsuhisa et al., 1979; Matthews et al., 1983). The near oxygen isotope equilibrium among minerals and between rock types under high-temperature conditions strongly suggests that the ^{18}O -lowering event occurred before UHP metamorphism, because any hydrothermal reactions with high water/rock ratios postdating UHP metamorphism would tend to disturb isotopic equilibrium (Gregory and Criss, 1986). In this regard, Rb-Sr and Sm-Nd mineral isochrons for sample QL-1 yielding concordant Triassic ages (Li et al., 1994) supports the present conclusion.

The eclogite sample QL-1 was collected from the middle part of the eclogite body, while sample SQ-92-3 was collected from a 30 cm-thick quartzite layer in the bottom interlayer part. The distance between these two samples is about 5 m. Near oxygen isotope equilibrium between these two samples is therefore significant. It is probable that the protoliths of both rock types have negative oxygen isotope compositions before the UHP metamorphism, although the possibility of complete isotopic equilibration between rock types with distinct initial $\delta^{18}\text{O}$ compositions during the UHP metamorphism cannot be completely excluded.

Because the UHP rocks were suggested to have been subducted to mantle depths (Liou et al., 1994), the survival of these unusually negative oxygen isotope compositions in eclogite/quartzite from the Qinglongshan reported is really surprising, but is consistent with the preservation of other anomalous ^{18}O eclogites. These values are even more significant when compared with the oxygen isotope data from other UHP terrains. For example, the UHP eclogites from the Western Gneiss Region of Norway exhibit $\delta^{18}\text{O}$ values ranging from +4 to +8‰ (Agrinier et al., 1985). The minerals in the UHP whiteschists from Dora Maira Massif, western Alps show oxygen isotope composition in the range from +4.5 to +8.6‰ (Sharp et al., 1993). Note that all these published data either cluster around the mantle $\delta^{18}\text{O}$ value (i.e., from +5 to +6‰) or are close to the oxygen isotope composition of the country rocks (e.g., Sharp et al., 1993). Any possible

interactions between these subducted rocks and their surroundings may, therefore, not be easily detected due to the small $\delta^{18}\text{O}$ contrast, despite the fact that the isotopic fractionations among minerals in these two UHP terrains indicate the preservation of high-temperature stable-isotope signatures (Agrinier et al., 1985; Sharp et al., 1993).

CONCLUSIONS

UHP eclogite and quartzite interlayers from the Qinglongshan in the Sulu terrain, eastern China are found to have negative oxygen isotope compositions. $\delta^{18}\text{O}$ values of mineral separates range from -10.4 to -7.3‰. These are the lowest $\delta^{18}\text{O}$ values ever reported for metamorphic rocks. Near isotopic equilibrium between minerals and rocks demonstrates that these rocks must have acquired such low oxygen isotope compositions before the UHP metamorphism. The only environment where such ^{18}O -depletion processes occurred is in hydrothermal systems involving meteoric waters. The protolith of the eclogite is therefore not submarine oceanic crust. These negative oxygen isotope compositions also imply an extremely limited isotopic exchange between the subducted rocks and their surroundings during the UHP metamorphism.

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