

Overpressures induced by coesite-quartz transition in zircon

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ABSTRACT

More than 120 coesite and few polycrystalline quartz (coesite pseudomorph) inclusions were identified by Raman microspectroscopy in zircons of various kinds from gneissic rocks located in the Dabieshan and Sulu ultrahigh-pressure (UHP) metamorphic terranes in eastern China. The coesite inclusions have undergone, to various degrees, the coesite-quartz transition. Raman spectra of coesite and subsidiary quartz inclusions show various shifts, which are closely correlated to the extent of the transition. The coesite-pseudomorph inclusions in zircon show the highest Raman shift. Calibrations based on the experimental work of Hemley (1987) yielded inconsistent present-day overpressures (0–24 kbar) in coesite inclusions inside zircons. The availability of fluids, which resulted in the pervasive regional retrogression of the UHP gneisses during later exhumation stages, is regarded as the main factor controlling the extent of the coesite-quartz transition. This study underscores the need for the elastic model applied by previous researchers to explain the preservation of the coesite inclusions in rigid host minerals.

INTRODUCTION

Coesite is a key index mineral used to define ultrahigh-pressure (UHP) metamorphic conditions (Liou et al. 1998) and the areal extent of UHP metamorphism in orogenic belts (Tabata et al. 1998; Parkinson and Katayama 1999; Ye et al. 2000). Coesite and its pseudomorph are easy to identify by optical means in UHP rocks. However, because of the extensive retrogression, no index UHP phases can be identified by optical methods in the Dabieshan-Sulu regional country gneisses (Carswell et al. 2000). Recent investigations have shown that zircon is a promising rigid vessel for the preservation of primary UHP phases. Laser Raman microspectroscopy has frequently been used to identify minute coesite and quartz inclusions in zircon of gneissic rocks from several UHP terranes (Sobolev et al. 1992; Tabata et al. 1998; Parkinson and Katayama 1999; Ye et al. 2000; Liu et al. 2001).

Coesite and α -quartz are distinguishable by their diagnostic Raman spectra. Coesite is characterized by a strong band at 521 cm^{-1} , together with other weaker bands at 116, 151, 176, 269, 326, 355, and 427 cm^{-1} at room temperature and atmospheric pressure (Hemley 1987; Sharma et al. 1981; Boyer et al. 1985). The main band of α -quartz is located at 464 cm^{-1} with subsidiary bands at 128, 206, and 355 cm^{-1} . However, the Raman spectra of coesite included in unfractured zircons often show a shift to higher frequency (Tabata et al. 1998; Parkinson and Katayama 1999; Ye et al. 2000).

Parkinson and Katayama (1999) reported that the 521 cm^{-1} spectra of “pure monocrystalline” coesite inclusions in

unfractured zircon and garnet of UHP metamorphic rocks show consistent shifting to 525–526 cm^{-1} . Raman spectra of coesite and quartz demonstrated a systematic pressure-dependent shift in a series of experiments by Hemley (1987). Based on the pressure calibration of Hemley (1987), Parkinson and Katayama (1999) deduced present-day ultrahigh overpressures (19–23 kbar) in coesite inclusions in unfractured zircon and garnet. Applying an elastic model, which was developed to explain the preservation of coesite inclusions in rigid-host containers (Gillet et al. 1984; van der Molen and van Roermund 1986), they concluded that the host mineral exerts an overpressure on the coesite inclusion, constraining it on, or close to, the coesite-quartz equilibria boundary, and the overpressures inhibited the coesite-quartz transformation during the exhumation of the host rocks.

Recently, we systematically analyzed Raman spectra of more than 120 coesite inclusions in zircons of various gneissic rocks from the Dabieshan and Sulu UHP terranes in eastern China. Various (0–24 kbar) present-day overpressures are deduced in coesite and its pseudomorph inclusions in zircons. Our new data indicate that the overpressures are induced by various degrees of the coesite-quartz transition, and the need for the elastic model is underscored. Effects of fluids on the coesite-quartz transition inside zircon and the P - T paths of the coesite inclusions during exhumations are discussed.

GEOLOGICAL BACKGROUND AND SAMPLE DESCRIPTIONS

The Dabieshan-Sulu UHP terranes are located in the collision zone between the Yangtze and North China cratons. The UHP terranes consist mainly of two sets of lithologies. One set is composed of intercalated marble, eclogite, garnet-biotite

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schist, jadeite-quartzite, and dark biotite- and epidote-rich paragneiss. This set of lithologies is considered to be a metamorphic sedimentary sequence. Another set of lithologies is composed of intercalated eclogite and tonalitic-trondhjemitic-granitic orthogneisses, with rare ultramafic rocks which occur as isolated lenses in the orthogneisses. Coesite and its pseudomorph are widespread in fresh UHP rocks, such as eclogite, jadeite-quartzite, garnet-bearing marble and garnet-biotite schist (see review by Liou et al. 1998). In contrast to the UHP rocks, the regional gneissic country rocks show extensive amphibolite-facies metamorphism, and no index UHP minerals have been identified by traditional optical means. Recently, however, coesite has been identified by Raman microspectroscopy as minute inclusions in zircons from some of the gneissic rocks, and this implies a possible regional and pervasive UHP metamorphic event that predated regional amphibolitic retrogression in the Dabieshan and Sulu UHP terranes (Tabata et al. 1998; Ye et al. 2000; Liu et al. 2001). To further understand the crystallization history of zircon grains and the metamorphic evolution of the host gneissic rocks, inclusions in zircons were systematically checked using Raman microspectroscopy.

More than 120 coesite and numerous quartz inclusions were identified in zircons from all types of gneissic rocks mentioned above. The results proved that most lithologies in the Dabieshan and Sulu UHP terranes witnessed the Triassic subduction to mantle depths greater than 80 km and later exhumation to the earth's surface, as a coherent continental slab (Tabata et al. 1998; Ye et al. 2000; Liu et al. 2001). Three kinds of coesite inclusions in zircon can be identified according to their modes of occurrence: (1) Most coesites occur as grains inside unfractured zircon (Fig. 1); (2) Four coesites occur inside zircons with fractures around coesite inclusions; (3) Five coesites were polished on surface of zircon grains. Two types of quartz inclusions in unfractured zircons were identified: monocrystalline and polycrystalline (Fig. 2). For comparison, coesites in fractured garnet and omphacite in eclogites from the Dabieshan and Sulu UHP terranes were also analyzed. All coesite inclusions in garnet and omphacite are bimineralic with retrogressed palisade quartz surrounding relic coesite, and radial fractures occur in the host garnet and omphacite.

LASER RAMAN MICROSCOPY AND RAMAN BAROMETRY OF COESITE AND QUARTZ INCLUSIONS

The coesite bands at 326, 355, and 427 cm^{-1} and the α -quartz bands at 206 and 355 cm^{-1} are close to bands of the host zircon and consequently are difficult to identify precisely. Because of this, we analyzed coesite bands at 116, 151, 176, 269, and 521 cm^{-1} and quartz bands at 128 and 464 cm^{-1} , using a JASCO NRS laser raman microspectrometer at the Department of Earth and Planetary Sciences, Tokyo Institute of Technology. Details of the procedure and working conditions are given in Parkinson and Katayama (1999). Individual types of coesite inclusions show various types of Raman spectra.

All fractured garnet- and omphacite-hosted, bimineralic coesite inclusions yielded clear Raman bands at 116–118, 151, 176, 269–270, 326–327, 427, and 521–522 cm^{-1} (Table 1). High intensity diagnostic quartz bands are also present at 126–128 and 465–466 cm^{-1} (Table 1). These coesite and quartz spectra

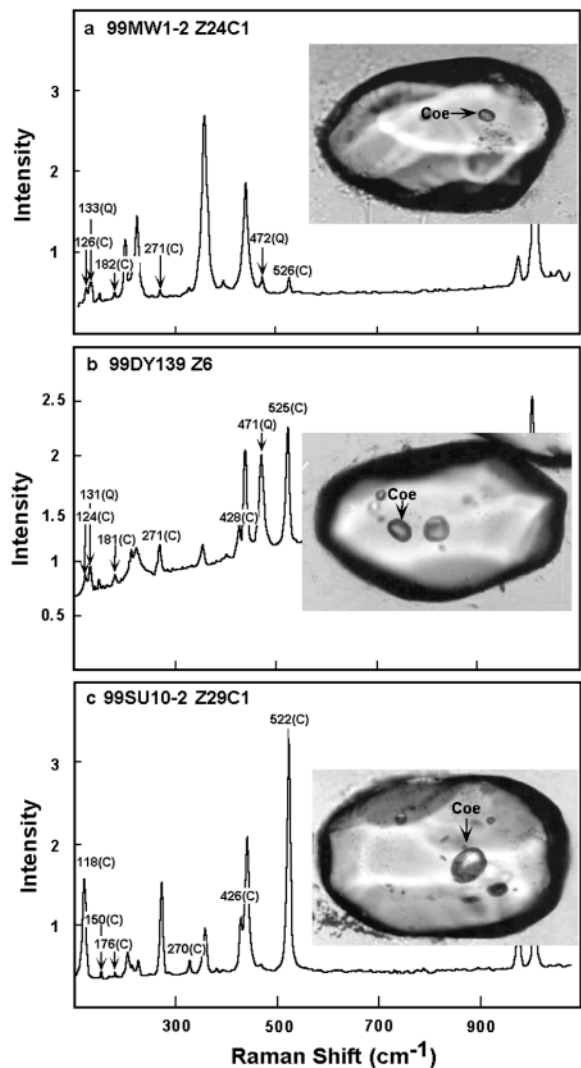


FIGURE 1. Representative microphotographs and Raman spectra for type 1 coesite inclusions in zircon. (a and b): coesite (C) with high level of transition to quartz (Q); note the intensity of the quartz bands and high Raman shift. (c) Monocrystalline coesite without a transition.

are consistent with those analyzed at room temperature and atmospheric pressure for synthetic coesite and quartz (Hemley 1987), and for natural coesite and quartz inclusions in fractured garnet and omphacite reported elsewhere (Smith 1984; Wang et al. 1989; Parkinson and Katayama 1999).

Raman bands of type 1 coesite inside unfractured zircon show various degrees of shift. The diagnostic 521 cm^{-1} band shifts in the range of 521–527 cm^{-1} , and the lower intensity coesite bands at 116, 151, 176, 269 cm^{-1} also show shifts to ranges of 116–126, 151–152, 176–182, 269–273 cm^{-1} , respectively (Fig. 1 and Table 1). Careful observations show that (1) quartz bands are always present in analyses with higher coesite Raman shift, and the higher the shift of the coesite bands, the higher the intensity of the diagnostic quartz bands (Fig. 1 and Table 1); (2) no or very low intensity quartz bands are identi-

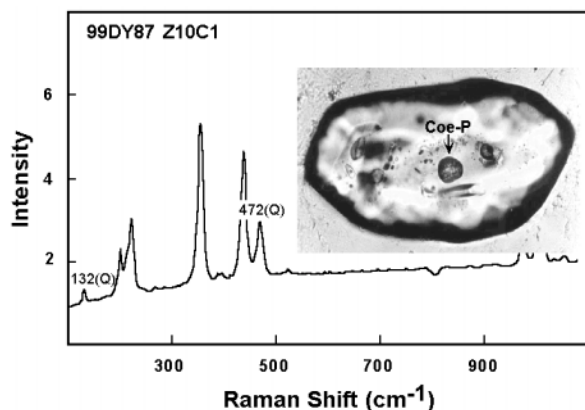


FIGURE 2. Representative microphotograph and Raman spectrum for polycrystalline coesite-pseudomorph inclusions in zircon. Note the high Raman shift.

fied in analyses of the coesite inclusions with diagnostic band located at 521–522 cm^{-1} (Fig. 1 and Table 1); (3) individual coesite and quartz bands show various degree of shifting; in order of decreasing shift they are: 116 (Coe) \rightarrow 464 (Qtz) \rightarrow 176 (Coe) \rightarrow 521 (Coe) \rightarrow 269 (Coe) \rightarrow 151 (Coe) (Table 1). This order is consistent with the experimental results of Hemley (1987). In a series of diamond-anvil cell experiments to 40 GPa, Hemley (1987) demonstrated that the room temperature Raman spectra of coesite and α -quartz show a clear pressure-dependent shift, and individual bands show various degree of shifting: the shifts of coesite bands at 116, 176, and 521 cm^{-1} and

the quartz band at 464 cm^{-1} are more sensitive than other bands to the change in pressure. We conclude that overpressures must exist on the coesite inclusions in zircon that we have studied and this is responsible for the shifts of the coesite and subsidiary quartz bands. Applying the calibration of Hemley (1987) to our samples, the shift observed in our samples requires a 0–24 kbar present-day overpressures. This result is quite different from that reported by Parkinson and Katayama (1999). Parkinson and Katayama (1999) observed that the coesite inclusions enveloped in unfractured zircon consistently yielded principal coesite Raman bands displaced to 525–526 cm^{-1} , which requires a 19–23 kbar overpressure (Hemley 1987). Our new data show that individual coesite inclusion in unfractured zircon bears various overpressures.

Although some type 2 and 3 coesites in zircon also show a high degree of subsidiary quartz bands (Table 1), the analyzed coesite (116–118, 151, 176–178, 269–270, 521–522 cm^{-1}) and quartz bands (128–129, 464–466 cm^{-1}) are generally consistent with those of the coesite and quartz at ATP (Hemley 1987), and thus imply no, or very low, overpressures.

All monocrystalline quartz inclusions in unfractured zircons give ATP Raman spectra at 128 and 464–466 cm^{-1} . However, the 128 and 464 cm^{-1} bands of all polycrystalline quartz inclusions in unfractured zircons are displaced consistently to 131–134 and 470–472 cm^{-1} , respectively (Fig. 2 and Table 1). No coesite spectrum is identified in the analyses of polycrystalline quartz inclusions (Fig. 2). Again applying of the calibration of Hemley (1987), the shift in Raman spectra yields overpressures of 20–24 kbar for polycrystalline quartz inclusions in unfractured zircons.

TABLE 1. Frequencies (cm^{-1}) of coesite and quartz Raman bands in spectra of representative inclusions

Sample no.	Lithology	Mineral	Host	Inclusion type	Band in Raman spectra (in cm^{-1})								
					Coe	Coe	Coe	Coe	Coe	Coe	Qtz	Qtz	
99DY134	eclogite	Coe+Qtz	Omp	Bimineralic	117	151		270	326	427	521	128	466
99DY134	eclogite	Coe+Qtz	Grt	Bimineralic	118	151		270	327	427	521	128	465
99MW1-2	Tonalitic. G	Coe+Qtz	Z24C1	1	126	152	182	271	328		526	133	472
99DY9	Paragneiss	Coe+Qtz	Z5	1	126	152	182	271	328		525		471
99MW1-2	Tonalitic. G	Coe+Qtz	Z30C1	1	124	152	180	270			524		470
99MW1	Tonalitic. G	Coe+Qtz	Z1C3	1	123	152	180	271	328	427	524		470
99DY87	Trondhjemitic. G	Coe+Qtz	Z6C1	1	122	151	180	271	327	524		470	
99MW1	Tonalitic. G	Coe+Qtz	Z8C1	1	122	152	180	270			523		469
99DY139	Trondhjemitic. G	Coe+Qtz	Z1	1	122	150	179	270	427	523		469	
99SU1-10	Granitic. G	Coe+Qtz	Z5C1	1	119	151	180	271	327		523		468
99MW1	Tonalitic. G	Coe+Qtz	Z9C1	1	120	151	179	271	328	428	522		468
99SU10-2	Granitic. G	Coe+Qtz	Z10C1	1	118	151	177	270			522	129	468
9DY137	Granitic. G	Coe+Qtz	Zir	2	118		178	270		428	522		468
99SU10-2	Granitic. G	Coe+Qtz	Z7C1	3	118	152	177	270		429	522		467
99SU10-2	Granitic. G	Coe+Qtz	Z58C1	3	118	150	176	270	328		522		467
99DY87	Trondhjemitic. G	Coe	Z2C2	1	119	151	177	270			522		
99DY9	Paragneiss	Coe	Z3	3	118	151	178	270			522		
99DY67	Tonalitic. G	Coe	Z2	1	118	151	177	270	326	427	522		
99SU10-2	Granitic. G	Coe	Z29C1	1	118	151	176	270	327	427	522		
99SU1-10	Granitic. G	Coe	Z2C1	1	117		177	270		428	522		
99MW1-2	Tonalitic. G	Coe-P	Z36	Polycrystalline								132	471
99DY87	Trondhjemitic. G	Coe-P	Z36	Polycrystalline	Z8							132	471
99ND4	Trondhjemitic. G	Coe-P	Z4	Polycrystalline								132	471
99DY87	Trondhjemitic. G	Coe-P	Z10C1	Polycrystalline								131	471
99MW1	Tonalitic. G	Coe-P	Z9	Polycrystalline								131	471
99MW1	Tonalitic. G	Coe-P	Z11	Polycrystalline								130	470

Note: Tonalitic. G = Tonalitic gneiss, Trondhjemitic. G = Trondhjemitic gneiss, Granitic. G = Granitic gneiss, Coe-P = coesite pseudomorph.

DISCUSSION

The coesite inclusion and the host zircon are considered to form at the same initial mantle lithostatic temperature and pressure conditions. Differential expansion is expected to take place between the coesite inclusion and the host zircon in response to falling pressures and temperatures on exhumation of the host rocks to the Earth's surface, and this results in thermal expansion elastic overpressures (Gillet et al. 1984; Van der Molen and van Roermund 1986; Zhang 1998). The thermal expansion induced elastic overpressure can be precisely estimated by the analysis of the differences in compressibilities and thermal expansivities of the inclusion-host systems (Zhang 1998). The initial coesite-zircon formation temperature of 700–800 °C at 30 kbar as estimated by chemical equilibria between coexisting peak phases in nearby coesite eclogite (Cong et al. 1995; Zhang et al. 1995). Using this initial *P-T* condition and bulk moduli and thermal expansion data of Holland and Powell (1998), we calculated that the elastic thermal expansion induced overpressure of less than 0.5 kbar in the coesite inclusion and zircon host system, based on stress model of Zhang (1998). This result is consistent with the very low Raman barometry estimated overpressures for the coesite inclusions with no, or very low, Raman shift, but is much lower than the overpressures of coesite inclusions with higher coesite Raman shift (Fig. 1 and Table 1). Overpressures higher than those induced by the elastic thermal expansion require other formation mechanisms.

The presence of diagnostic quartz bands, with various intensities, together with coesite bands indicate that some coesites are not “pure monocrystalline,” but instead reflect various degrees of the coesite-quartz transformation in the coesite inclusions. Taking into consideration the positive relationship between the intensity of the subsidiary quartz bands and the degree of the Raman shifting, we conclude that the overpressures are mainly controlled by the extent of the coesite-quartz transition in the inclusions. Because coesite is 10% denser than quartz, the coesite-quartz transition will result in volume enlargement, and this is regarded as the reason for the formation of radial fractures in host garnet and omphacite in natural UHP rocks (Gillet et al. 1984; van Molen and van Roermund 1986).

The presence of high intensity quartz bands together with coesite bands in type 2 and 3 coesite inclusions in zircon indicate that the coesite-quartz transition must have occurred in those coesite inclusions, but Raman barometry gives no, or very low, overpressures. The previously existing overpressures induced by the coesite-quartz transition might have been released along fractures or during polishing to the surface.

Polycrystalline quartz inclusions in unfractured zircon yield the highest Raman shift and overpressures. We interpret the polycrystalline quartz inclusions as pseudomorphs after coesite, with the overpressure reflecting the volume enlargement after the coesite-quartz transition. The absence of coesite spectra in analyses of the coesite-pseudomorphs indicates that all the original coesite was completely consumed.

Gillet et al. (1984) and Van der Molen and van Roermund (1986) proposed an elastic model to explain the preservation of coesite in rigid hosts, and this elastic model was applied by Parkinson and Katayama (1999). According to their elastic model, the overpressure, induced by the volume change of the

coesite-quartz transition during the exhumation of the host rocks, constrains the coesite inclusion on, or close to, the coesite-quartz equilibrium boundary. This inhibits the coesite-quartz transformation. If fluids were available, this elastic model might work at high temperature conditions (echoed by the initial exhumation stage) such that the reaction kinetics are quick enough to let the overpressures buffer the reaction (Mosenfelder and Bohlen 1997). However, the inconsistent overpressures exerted on coesite enclosed in zircon and the occurrence of coesite pseudomorphs reported here, as well as the occurrence of intergranular coesite in eclogites (Liou and Zhang 1996; Ye et al. 1996) underscore the need for the elastic model (Mosenfelder and Bohlen 1997). Our new data show that, except for the few inclusions with the highest Raman shift, the overpressures exerted on most coesite inclusions are lower than pressures required on the coesite-quartz equilibria, and thus the overpressures are unlikely to inhibit the coesite-quartz transformation. The occurrence of coesite pseudomorphs with high overpressures in unfractured zircon further demonstrates that the overpressure failed to prevent the coesite-quartz transition during the exhumation of the host rocks.

The varying degrees of coesite-quartz transition in coesite inclusions in zircon reported here imply that some factors other than overpressure must have controlled the preservation of the coesite inclusions. Fluid is well known to have a catalytic effect on reactions (e.g., Rubie 1986). The experiments of Mosenfelder and Bohlen (1997) suggest that coesite is unlikely to be preserved on a geologic time scale if it reaches the quartz stability field at temperatures above 375–400 °C with high fluid activity. The *P-T* conditions and availability of fluids during exhumation of the host rocks are therefore considered to play a crucial role in controlling the extent of the coesite-quartz transition. The preservation of coesite in fractured garnet and omphacite, and coesite in the interstice of more than one UHP phase in eclogites (Liou and Zhang 1996; Ye et al. 1996) requires a relatively low fluid activity at high temperature conditions. The preservation of extremely low oxygen isotopic protolith compositions (e.g., Zheng et al. 1999) also implies a very limited fluid flux in the gneissic rocks during exhumation at high temperature conditions. The occurrence of coesite pseudomorphs with high overpressures as reported here also implies that the fluid, which catalyzed the coesite-quartz transformation, must access the previously coesite at relatively low temperature conditions, possibly during the later regional retrogression stage. Otherwise the elastic model might work, that is, the overpressures might buffer the coesite inclusions on, or close to, the coesite-quartz boundary, and inhibit the coesite-quartz transformation (Gillet et al. 1984; Van der Molen and van Roermund 1986). Based on the above argument, we conclude that the coesite inclusions in zircon suffered only a very limited transformation to quartz during the high-temperature exhumation, and consequently, the coesite inclusions might follow a *P-T* path consistent with the host rocks in this stage. Subsequently, fluid may access the coesite inclusions at various degrees during the pervasive amphibolitic retrogression, and this may explain the observed range in the degree of transformation to quartz, and hence the range of the calculated overpressures. Individual coesite inclusions might follow different

P-T paths, which are controlled by the inclusion overpressures in the later exhumation stage of the host rocks.

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