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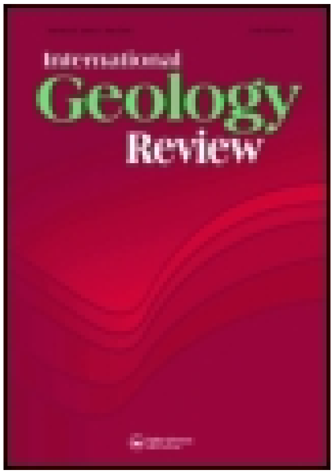

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Critique of Stability Limits of the UHPM Index Minerals Diamond and Coesite

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Critique of Stability Limits of the UHPM Index Minerals Diamond and Coesite

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Abstract

The transition boundaries for the graphite ↔ diamond and quartz ↔ coesite reactions are reviewed. The recommended linear transition boundaries are P (kbar) = 15 + 0.028 T (°C) for the graphite ↔ diamond transition, and P (kbar) = 22 + 0.009 T (°C) for the quartz ↔ coesite transition. The recommendation was made on the basis of reliable transition boundaries determined at high-pressure and high-temperature experiments and their consistency with the thermochemical calculation of the transition pressures at room temperature. The recommended boundary for the graphite ↔ diamond transition is about 2 kbar higher, and that for the quartz ↔ coesite transition is about 1.5 to 3.5 kbar higher than those adopted in the literature to constrain the P-T conditions of UHPM origin.

Introduction

ULTRAHIGH PRESSURE METAMORPHISM (UHPM) identified in gneisses of the Kokchetav Massif, Kazakhstan (Sobolev and Shatsky, 1990), in the Saxonian Erzgebirge, Germany (Massonne et al., 1998; Massonne, 1999), and in the Rhodope Metamorphic Province, northern Greece (Mposkos and Kostopoulos, 2001) was supported mainly by the discovery of microdiamonds, which exist as inclusions in garnet in the Kokchetav Massif; in garnet, kyanite, and zircon in the Erzgebirge; and in garnet and mica in the Rhodope. Other metamorphic microdiamonds are less well documented because of their scarcity (e.g., Xu et al., 1992; Dobrzhinetskaya et al., 1995; Smith, 1995). In addition to microdiamonds, the other major high-pressure mineral that suggests UHPM in Kokchetav and Erzgebirge is coesite, discovered as rare inclusions in garnet and omphacite in the nearby eclogite bodies of the aforementioned areas (Korsakov et al., 1998; Massonne, 2001) and in zircon from the Kokchetav Massif (Shatsky et al., 1995). Coesite was also found as an index mineral for UHPM terrains lacking diamonds (e.g., Chopin, 1984; Smith, 1984; Wang et al., 1989; Caby, 1994; Shatsky et al., 1995).

More recently, microdiamonds have also been found in association with other minerals in polyphase inclusions in garnet from the Erzgebirge (Hwang et al., 2001; Stockhert et al., 2001). Phologopite, quartz, and paragonite are the three major polyphases, and it is the first time that microdiamonds were found to coexist with quartz (but not coesite) in the same inclusion.

Given their importance in establishing UHPM conditions, the transition boundaries between graphite and diamond and that between quartz and coesite are crucial to our understanding of the formations of the microdiamonds and the polyphase inclusions, as well as the evolution of the P-T paths (or depths) of the burial and exhumation of continental crust. I review thoroughly the transition boundaries of these two reactions on the basis of experimental determinations and thermochemical calculations. Of course, stability fields for the α-PbO_2-type TiO_2 (Hwang et al., 2001) and other minerals in polyphase inclusions are also important, but most of them are not as thoroughly determined. Comments on the validity of the stability fields for other minor minerals are also made after transition boundaries of the graphite ↔ diamond and quartz ↔ coesite reactions are reviewed.

The Graphite ↔ Diamond Boundary

All available experimental determinations of the transition boundary of the graphite ↔ diamond reaction are shown in Figure 1. These experimental studies were carried out mainly by the research groups at the General Electric Company and in George Kennedy's laboratory at UCLA. Earlier works prior to 1975 were all done at the General Electric Company and the belt apparatus was used. A low-friction piston-cylinder apparatus was

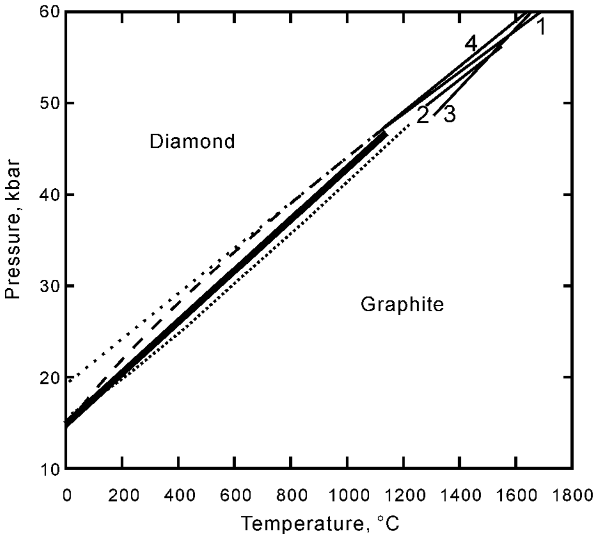

FIG. 1. Transition boundaries for the graphite ↔ diamond reaction determined by: 1 = Bundy et al. (1961); 2 = Strong and Hanneman (1967); 3 = Strong and Chrenko (1971); and 4 = Kennedy and Kennedy (1976). The boundary calculated by Berman (1979) is shown by a close-spaced dotted line. The open-spaced dotted line is a linear extrapolation of the work of Kennedy and Kennedy (1976), which was also smoothly extrapolated to room temperature at 15.6 kbar by a thin dashed line. The recommended linear transition boundary of P (kbar) = 15 + 0.028 T (°C) is shown by the heavy line.

employed in the work of Kennedy and Kennedy (1976). Except for the work of Bundy et al. (1961), the effect of pressure on the emf of the thermocouple was corrected in all later studies.

The boundaries reported by both Strong and Hanneman (1967) and Strong and Chrenko (1971) were determined utilizing two data points each. As shown in Figure 1, they all lie on the low-temperature side of all the later determinations, although the slope of the boundary determined by Strong and Hanneman (1967) is in agreement with those determined by Bundy et al. (1961) and Kennedy and Kennedy (1976). As noted by Kennedy and Kennedy, the boundary determined by Bundy et al. is almost coincident with that of their own measurement. Kennedy and Kennedy (1976) concluded that the similarity in large part is fortuitous, because the effect of pressure on the emf of the thermocouple was not corrected and the different calibration pressure for the barium transition was used in the work of Bundy et al. (1961). While the opinions of are disputable, reversal experiments were carried out by Kennedy and Kennedy (1976).

The experimental determinations of the graphite ↔ diamond transition boundary were investigated in the temperature range between 1100 and 1700°C. The UHPM identified in gneisses is believed to occur at temperatures below ~1000°C, and the microdiamonds were formed in the temperature range 800–900°C (e.g., Zhang et al., 1997; Hwang et al., 2001; Stockhert et al., 2001). Thus, in order to apply the experimentally determined graphite ↔ diamond transition boundary to the formation of the microdiamonds, the experimental data must be extrapolated to lower temperatures. Unless a linear extrapolation is assumed, a reliable extrapolation can only be done by a calculation using available thermochemical and compression (or elastic) data for both graphite and diamond via the following relation:

$$\Delta G_T^0(h) - \Delta G_T^0(l) = -\int_0^{P_t} [V(h) - V(l)]_T dP \quad (1)$$

where $\Delta G_T{}^0$ is the Gibbs free energy of formation, V is the volume, P the pressure, the superscript 0 denotes zero pressure and subscript T denotes a constant temperature in K, and the letters l and h inside the parentheses represent low- and high-pressure phases, respectively. All these parameters for both graphite and diamond are well determined at room temperature. Therefore, the transition pressure P_t for the graphite ↔ diamond reaction at room temperature should be well constrained. On the basis of thermochemical data compiled by Robie (1966) and the elasticity data compiled by Bass (1995), the graphite ↔ diamond transition boundary was calculated to be 15.6 ± 0.4 kbar at room temperature. The uncertainty is mainly due to the thermochemical data, and the Birch equation of state was used to computer the volume data. A linear extrapolation of the data of Kennedy and Kennedy (1976) yields 20 kbar at room temperature. The room temperature value extrapolated from the data of Bundy et al. (1961) is much too high.

The calculation employing Equation (1) can also be regarded as an independent test to discriminate the data obtained by direct experimental determination of the transition boundary at high pressures and high temperatures, which suffers from many factors such as the lack of a reliable pressure scale at elevated temperatures. The data calculated at room temperature is particularly reliable, because the thermochemical and compression data for both the low- and high-pressure phases are well determined at room temperature.

On the basis of the above criterion, the experimental curves for the transition boundary of the graphite ↔ diamond reaction determined by Bundy et al. (1961) and Kennedy and Kennedy (1976) were smoothly extrapolated to room temperature at 15.6 kbar by a thin dashed line in Figure 1. For simplicity and more practical use, a linear equation of P (kbar) = 15 + 0.028 T (°C), denoted by the heavy line in Figure 1, is recommended. This line passes through 15.6 kbar at room temperature and also is in agreement with the experimental determinations of Bundy et al. (1961), Strong and Hanneman (1967), and Kennedy and Kennedy (1976) at temperatures above 1100°C. The linear extrapolation of the Kennedy and Kennedy work to room temperature is shown by an open-spaced dotted line in Figure 1 for comparison.

On the basis of the same Equation (1), Berman (1979) calculated the equilibrium transition boundary for the graphite ↔ diamond reaction between 25 and 2727°C. This result at T < ~1200°C is also shown by a close-spaced dotted line in Figure 1. Berman (1979) calculated P_t = 16.0 kbar for the reaction at room temperature. This value, of course, is very close to that calculated in the present study. The small difference most likely results from the fact that the Murnaghan equation of state was used by Berman (1979), whereas the Birch equation of state was employed in the present study. Although Berman (1979) emphasized that the uncertainties associated with the calculated equilibrium transition boundary are almost negligible, this is probably true only in terms of the thermochemical data used. The value of $[V(h) - V(l)]_T$ in Equation (1) was assumed to be independent of temperature in the calculation of Berman (1979). Uncertainties introduced by this assumption alone were estimated to be about 1% at 200°C and about 10% at 1000°C by Liu (1979). As can be seen in Figure 1, the calculated equilibrium transition boundary for the graphite ↔ diamond reaction by Berman (1979) is close to the unreliable measurements of Strong and Hanneman (1967) and Strong and Chrenko (1971), and is ~2 kbar less than those of Bundy et al. (1961) and Kennedy and Kennedy (1976) at 1200°C.

The Quartz ↔ Coesite Boundary

The situation for the transition boundary of the quartz ↔ coesite reaction is problematic. The P-T transition boundary of the reaction is probably the most well studied, but most poorly constrained, in the high-pressure and high-temperature field. Brief summaries of the data can be found in, for example, Mirwald and Massonne (1980), Bohlen and Boettcher (1982), Kuskov and Fabrichnaya (1987), Mosenfelder and Bohlen (1997) and Liou et al. (1998).

Some 20 independent experimental determinations and thermochemical calculations of the transition boundary are known in the literature. The results of these studies, however, show considerable discrepancies concerning the equilibrium pressures and temperatures (see Fig. 2). There is no simple explanation for the wide discrepancies and it is even more difficult to discriminate one against the other. For high-pressure and high-temperature experimental studies, however, the reaction rate to reach equilibrium at low temperatures and the uncertainties in our knowledge of pressure at high temperatures are probably the two main reasons for the reported differences, in addition to other difficulties such as shear strength of the pressure medium, friction, and pressure gradients existing in the various high-pressure apparatus. The discrepancy cannot even be resolved by reversal studies. Reaction reversals were conducted in each run by Bohlen and Boet-

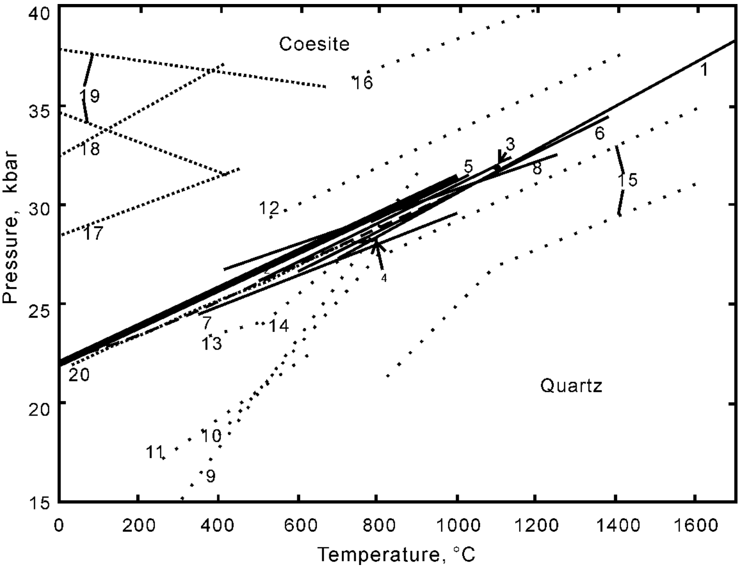

FIG. 2. Transition boundaries (solid lines) for the quartz ↔ coesite reaction determined using the piston-cylinder apparatus: 1 = Boyd and England (1960); 2 = Kitahara and Kennedy (1964); 3 = Green et al. (1966); 4 = Boettcher and Wyllie (1968); 5 = Akella (1979); 6 = Mirwald and Massonne (1980); 7 = Bohlen and Boettcher (1982); 8 = Bose and Ganguly (1995). Those determined using other pressure devices are shown by open-spaced dotted lines: 9 = Griggs and Kennedy (1956); 10 = MacDonald (1956); 11 = Dachille and Roy (1959); 12 = Takahashi (1963); 13 = Bell et al. (1965); 14 = Roy and Frushour (1971); 15 = Naka et al. (1972); and 16 = Bohler and Arndt (1974). Thermochemical calculations of the transition boundary (shown by close-spaced dotted lines) were performed by: 17 = Holm et al. (1967); 18 = Liu (1979); 19 = Weaver et al. (1979); and 20 = Kuskov et al. (1991). See text for the calculations performed by Akaogi and Navrotsky (1984), by Akaogi et al. (1995), and Hemingway et al. (1998). For clarity, the transition boundary between low- and high-quartz is not shown.

tcher (1982) and Bose and Ganguly (1995) in their study of the quartz ↔ coesite reaction (lines 7 and 8, respectively, in Fig. 2). Their results, as shown in Figure 2, are nearly parallel to each other, but the transition pressure determined in the latter study is about 1.5 kbar higher than that determined in the former study at the same temperature. Bose and Ganguly (1995) stated that they are unable to sort out the reason for the pressure discrepancy.

To analyze the data, I first divided the determined transition boundaries of the quartz ↔ coesite reaction shown in Figure 2 into two groups: in one group the boundaries were determined by employing the piston-cylinder apparatus (shown by solid lines from 1 to 8 in Fig. 2) and in another they were determined by another pressure apparatus (shown by open-spaced dotted lines from 9 to 16 in Fig. 2). This is because the piston-cylinder apparatus is believed to be the most suitable device for determination of the P-T transition boundary of this kind. As shown in Figure 2, the latter proposal seems to be supported by the concordance of the results (they all cluster in a very narrow pressure range of 29.6 ± 1 kbar at 900°C). Except for the work of Mirwald and Massonne (1980; line 6 in Fig. 2), however, the effect of pressure on the emf of the thermocouple was not corrected in studies employing the piston-cylinder apparatus. Thus, according to the comments by Kennedy and Kennedy (1976) on the graphite ↔ diamond transition, the general agreement may be fortuitous. The other reason for the concordance is because some studies have attempted to determine the transition boundary and to calibrate the pressure of their piston-cylinder apparatus at high temperature simultaneously (e.g., Green et al., 1966; Boettcher and Wyllie, 1968).

The results of Mirwald and Massonne (1980) and Bohlen and Boettcher (1982) are most commonly quoted in the studies of UHPM (e.g., Zhang et al., 1997; Hwang et al., 2001; Stockhert et al. 2001).

Although the general results of these two studies are very close to each other, the dT/dP slopes of the transition boundaries determined are rather different. When these results are extrapolated to temperatures below 600°C, very different values of transition pressure are obtained (see Fig. 3).

The boundaries of the quartz ↔ coesite transition determined by high-pressure devices other than the piston-cylinder apparatus spread widely in the pressure range from 15 to 40 kbar, as shown in Figure 2. Except for the very early work of Griggs and Kennedy (1956; line 9), MacDonald (1956; line 10), and Dachille and Roy (1959; line 11), the dT/dP slope of the transition boundaries are generally in good agreement with those determined in the piston-cylinder apparatus (excluding lines 7 and 8). This suggests that the absolute pressure calibration used in the various studies is in gross disagreement, but the variation of pressure with temperature is the same for most of these studies.

Among all the studies shown in Figure 2, only the results obtained by Bohler and Arndt (1974; line 16) were carried out by *in situ* X-ray measurements using the NaCl pressure scales simultaneously at high pressures and high temperatures. These authors observed that the pressure increased with increasing temperature at constant loading pressure in all of their conducted experiments. All other studies shown in Figure 2 must also be subjected to the same difficulty (as admitted by Bose and Ganguly; 1995), which might be the real cause of the discrepancies displayed in Figure 2. However, despite the superiority of the *in situ* study using the NaCl pressure scale at elevated temperatures, the pressure of the transition boundary determined by Bohler and Arndt (1974) is too high in comparison with all other data shown in Fig. 2. This discrepancy is not well understood.

As it has been demonstrated for the graphite ↔ diamond boundary, Equation (1) provides additional constraints on the quartz ↔ coesite transition. This has been used several times in the literature, and the results of these studies are displayed by closely spaced dotted lines in Figure 2. The first such study was performed by Holm et al. (1967; line 17), who measured the thermochemical data for the quartz ↔ coesite transition in the temperature range 5 to ~1000 K. However, their calculation of the quartz ↔ coesite transition boundary was limited at the time by the unavailability of compression data for coesite. Using the same thermochemical data of Holm et al. (1967), Liu (1979) and Weaver et al. (1979) performed the same calculation using Equation (1) on the basis of the experimentally determined compression curve of coesite then available to them. As shown in Figure 2, however, they obtained quite different results (lines 18 and 19). The results obtained by Weaver et al. (1979; line 19) have particularly attracted attention in the literature (e.g., Mirwald and Massonne, 1980; Bohlen and Boettcher, 1982; Akaogi and Navrotsky, 1984) because a negative dT/dP slope of the quartz ↔ coesite transition boundary was reported. The details of these studies are not elucidated here. Unless the transition boundary determined by Bohler and Arndt (1974) was correct, the calculations of Liu (1979) and Weaver et al. (1979) all suggest that the values of ΔG_T^0 for the quartz ↔ coesite transition determined by Holm et al. (1967) are probably too high.

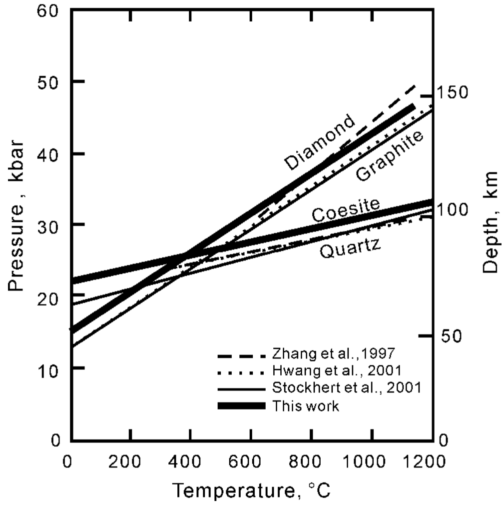

FIG. 3. Transition boundaries for the graphite ↔ diamond and quartz ↔ coesite reactions adopted by Zhang et al. (1997), Hwang et al. (2001) and Stockhert et al. (2001) are compared with those recommended in this study.

The thermochemical data of the quartz ↔ coesite transition were redetermined by Akaogi and Navrotsky (1984), Kuskov et al. (1991), and Akaogi et al. (1995), who also calculated the quartz ↔ coesite transition boundary on the basis of their new data. The calculated curves of the transition boundary by Akaogi and Navrotsky (1984) and Akaogi et al. (1995) are almost identical to those determined by Bohlen and Boettcher (1982) and by Mirwald and Massonne (1980) in the temperature range measured. For clarity, the calculated curves are not shown in Figure 2. On the other hand, the good agreement is not surprising, because a particular P,T point in the experimental determinations was chosen as a base point in the calculations of Akaogi and Navrotsky (1984) and Akaogi et al. (1995). Actually, what is really demonstrated by these investigators is that the dT/dP slope determined in the experiments of Mirwald and Massonne (1980) and Bohlen and Boettcher (1982) is in good agreement with that of the calculations. However, as pointed out earlier, the dT/dP slopes in the determinations of Mirwald and Massonne (1980) and Bohlen and Boettcher (1982) are rather different from each other (see lines 6 and 7 in Fig. 2). The transition pressures for the quartz-coesite reaction between 25 and 727 °C calculated by Kuskov et al. (1991) are slightly higher (by ~0.5 kbar) than those determined by Mirwald and Massonne (1980) and Bohlen and Boettcher (1982), but the dT/dP slope of the calculated boundary is closer to that of Mirwald and Massonne (1980) than to that of Bohlen and Boettcher (1982).

The thermochemical data of coesite has been further revised by Hemingway et al. (1998), who recalculated the quartz ↔ coesite transition boundary again. However, the way the transition boundary was calculated is rather peculiar. First, they used the transition boundary determined by Bohlen and Boettcher (1982) as a base, then calculated the ambient enthalpy values for the quartz ↔ coesite reaction from the transition boundary using Equation (1). The calculation was repeated until the calculated ambient enthalpy value nearly approached a constant. Only the values of bulk modulus and thermoexpansivity of quartz and coesite were cited in the calculation. This is not enough to warrant the calculations, however, because how the values of $[V(h) - V(l)]_T$ at higher temperatures were estimated was not explicitly stated. In fact, all the thermochemical calculations above 200°C experience the same difficulty because the compression (or elastic) data at elevated temperatures are not available for these minerals. A simple assumption that $[V(h) - V(l)]_T$ on the right-hand side of Equation (1) is independent of temperature at T < 200°C is probably appropriate (Liu, 1979).

In summary, regardless of the various high-pressure devices used, the different corrections applied, and the different pressure scales employed in each experimental study, the experimental data shown in Figure 2 seem to suggest that the dT/dP slope of 110 ± 10°C/kbar is well established for the quartz ↔ coesite transition. By adopting the same principle used to establish the graphite ↔ diamond transition boundary, a transition pressure of 22.3 ± 0.5 kbar at room temperature was calculated for the quartz ↔ coesite reaction using Equation (1). $\Delta G_{298}{}^{0} = 890 \pm 20$ cal/mole from the work of Akaogi et al. (1995) and Hemingway et al. (1998) and the elastic moduli of both quartz and coesite recommended by Liu (1993) were used in the calculation. The bulk moduli of coesite have recently been remeasured by Angel et al. (2001), who concluded that the new data do not affect the earlier calculations of the quartz ↔ coesite transition. Thus, the experimental curves for the transition boundary of quartz ↔ coesite reaction determined by using the piston-cylinder apparatus were smoothly extrapolated to room temperature at 22.3 kbar by a thin dashed line in Figure 2. This line is coincident with that calculated by Kuskov et al. (1991; line 20 in Fig. 2) at T > 200°C. As before, a linear equation of P (kbar) = 22 + 0.009 T (°C), which is denoted by the heavy line in Figure 2, is recommended for the quartz ↔ coesite transition.

Comparison with Data in the Literature

As mentioned earlier, the transition boundaries for both the graphite ↔ diamond and quartz ↔ coesite reactions have been widely used to study the formation of the microdiamonds and to infer the P-T conditions of UHPM of the continental crust. Figure 3 compares the transition boundaries used in three recent studies (Zhang et al., 1997; Hwang et al., 2001; Stockhert et al. 2001) with those recommended here. As can be seen, the dT/dP slopes for both the graphite ↔ diamond and quartz ↔ coesite reactions adopted in the literature are in general agreement with those recommended in this work. In view of the experimental data reviewed in Figure 1, except for the work of Zhang et al. (1997), the pressure of the transition boundary of the graphite ↔ diamond reaction adopted in the literature is certainly too low by at least 2 kbar. Although the transition pressure of the graphite ↔ diamond boundary used by Zhang et al. (1997) agrees with that recommended in this study in the vicinity of 800°C, their transition boundary for the graphite ↔ diamond reaction would yield much too low pressures at lower temperatures because of the small dT/dP slope.

The transition pressures of the quartz ↔ coesite reaction from the literature are all about 1.5 to 3.5

kbar lower than those recommended from this study. This would increase the inferred depth of UHPM about 8 km toward the interior.

The transition boundaries of other minor minerals such as the rutile ↔ α-PbO_2–type TiO_2 and the paragonite ↔ jadeite + kyanite + H_2O reactions were also used to study the formation of polyphase inclusions and to constrain the P-T conditions of UHPM origin (e.g., Hwang et al., 2001). In view of the extensive experimental determinations of the graphite ↔ diamond and quartz ↔ coesite transitions and the existing discrepancies reviewed in this study, the reliability of the transition boundaries of these minor minerals is quite doubtful. The P-T evolution of the three metamorphic stages for the diamondiferous gneiss in the Erzgebirge proposed by Massonne (1999) has been shown to be consistent with the stability field of the nanophase α-PbO_2–type TiO_2 by Hwang et al. (2001). On the other hand, the α-PbO_2–type TiO_2 has so far been found once only between twinned rutile bicrystals by Hwang et al. (2000), who also demonstrated that the basic unit of atoms at the twin boundary of rutile is identical to that of the α-PbO_2–type TiO_2. Thus, it would be interesting to find out whether very high pressure is required to form the α↔-PbO_2–type TiO_2 between twinned rutile bicrystals.

Acknowledgments

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