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## An alternative interpretation of lower mantle mineral associations in diamonds

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**Abstract** Diamonds containing ferropericlase (Mg,Fe)O and other silicate (enstatite [(Mg,Fe)SiO<sub>3</sub>], in particular) assemblages are generally believed to be derived from the Earth's lower mantle. On the basis of the observed ratio between ferropericlase and enstatite inclusions and the FeO content of these ferropericlases, it is concluded that most of these minerals entrapped in diamonds may not represent the lithology of the lower mantle itself as has been suggested by many investigators. Instead, ferropericlases in these diamonds represent most likely the disproportionated product of ferromagnesite [(Mg,Fe)-CO<sub>3</sub>], which underwent a decarbonation reaction to form both diamond and ferropericlase simultaneously in the lower mantle. The wide variation in the Mg# of ferropericlase inclusions in diamonds is attributed to the decarbonation "loop" of the MgCO<sub>3</sub>-FeCO<sub>3</sub> solid solutions. Some of the enstatite inclusions coexisting with these ferropericlases in the same diamond may represent the most abundant mineral species of (Mg,Fe)SiO<sub>3</sub>-perovskite in the lower mantle. The latter mineral phase experienced a retrogressive transition into enstatite during the transport of diamonds to the Earth's surface.

### Introduction

Periclase + enstatite and forsterite + quartz (and its high-pressure variants) are the so-called "forbidden" assemblages in the upper mantle. Forsterite, enstatite, and quartz all undergo various phase transitions at high pressures and high temperatures (e.g., Liu and Bassett 1986). Regardless of the various phase transitions in the entire MgO-SiO<sub>2</sub> system, it is only possible for periclase

and MgSiO<sub>3</sub> to coexist when Mg<sub>2</sub>SiO<sub>4</sub>-spinel breaks down to periclase + MgSiO<sub>3</sub>-perovskite at pressures greater than about 27 GPa at ~1,000 °C (Liu 1975, 1976a). The above phase relationship has also been extended to include some 20–30 mol% of FeO by Liu (1976b), and therefore periclase is replaced by ferropericlase in the system MgO-FeO-SiO<sub>2</sub>. Detailed equilibrium phase assemblages in the MgO-FeO-SiO<sub>2</sub> system at 25 GPa and 1,100 and 1,600 °C have been later delineated by Ito and Takahashi (1989). Thus, ferropericlase and (Mg,Fe)SiO<sub>3</sub>-perovskite can only possibly coexist in the P, T conditions of the lower mantle.

A large amount of geophysical, geochemical and experimental evidence suggests that the Earth's upper mantle consists dominantly of peridotite or pyrolite. The Mg-number of such a chemical composition is close to 90 (Ringwood 1975). If the pyrolitic model can be extended to the lower mantle, the lower mantle should be composed of > 70 wt% of (Mg,Fe)SiO<sub>3</sub>-perovskite and ~20 wt% of ferropericlase (Liu 1979; Irifune and Ringwood 1987). If the Mg/Si ratio of the bulk mantle is similar to that in C1 chondrites, the lower mantle would be composed essentially of (Mg,Fe)SiO<sub>3</sub>-perovskite (Liu 1982). Thus, there is little doubt that (Mg,Fe)SiO<sub>3</sub>-perovskite is the most abundant mineral phase in the lower mantle lithology.

Diamonds containing ferropericlase as inclusions were discovered in several localities and suggest a lower mantle origin. On the basis of two ferropericlase [(Mg,Fe)O] and one enstatite [(Mg,Fe)SiO<sub>3</sub>] inclusions found in three separate diamonds from Orrorro, Scott-Smith et al. (1984) suggested that the enstatite inclusion might represent former (Mg,Fe)SiO<sub>3</sub>-perovskite and might have coexisted with the ferropericlase in the lower mantle. Five further ferropericlases and one magnesiowustite [(Fe,Mg)O] from Koffiefontein, Monastery, and Sloan were later reported by Moore et al. (1986). This includes a non-touching pair of ferropericlase and enstatite occurring in the same diamond from Koffiefontein. Coupling with high-pressure and high-temperature experimental data, Kesson and Fitzgerald (1991) also concluded that

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these ferropericlases and enstatites may indeed represent the lithology of the lower mantle itself.

However, in view of the many ferropericlase (in both composition and quantity) and enstatite inclusions recently found worldwide and in consideration of the plausible genesis of these lower mantle diamonds, it will be demonstrated that these inclusions do not represent lower mantle lithology as claimed.

### Rare and unusual mineral inclusions in diamonds

In addition to the above-mentioned ferropericlase and enstatite inclusions (enstatite is not a rare inclusion, but it is rare when it coexists with ferropericlase in the same diamond as they do in the so-called “forbidden” assemblages in the upper mantle), many more such rare and unusual mineral inclusions in diamonds have been reported in recent literature. Table 1 lists all these inclusions.

Although (Mg,Fe)SiO<sub>3</sub>-perovskite should be the most abundant mineral species in the lower mantle, Harte et al. (1999) have particularly focused their attention on the significance of diamonds that have ferropericlase inclusions. This is because enstatite is a common inclusion in diamonds, and such enstatite may not, chemically and structurally, be distinguishable from those enstatites derived from the lower mantle. Stachel et al. (2000), however, found that enstatite originating from the lower mantle may be readily recognized by its very low NiO content ( $\leq 0.03$  wt%). Some investigators believe that ferropericlase by itself is not necessarily an indication of an ultrahigh pressure origin (e.g., Eggler

and Baker 1982; Stachel et al. 1998). Following the suggestion of Harte et al. (1999) and others, I also assume that diamonds containing ferropericlase listed in Table 1 represent a lower mantle origin.

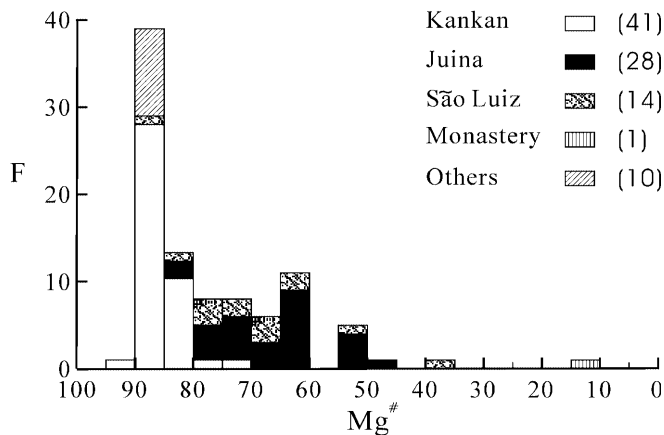
Because of the possible main lithology of the lower mantle described earlier, only the significance of the ferropericlase, enstatite, and SiO<sub>2</sub> inclusions is discussed here. All these inclusions are divided into two groups. The first group consists of ferropericlase (including three magnesiowustites) inclusions only (Fig. 1), and the second group consists of ferropericlases coexisting with enstatite or SiO<sub>2</sub> in the same diamond (Fig. 2). The chemical composition for all these inclusions is available from Harte et al. (1999), Stachel et al. (2000), and Kaminsky et al. (2001). The three-phase regions of (Mg,Fe)SiO<sub>3</sub>-perovskite + ferropericlase + stishovite at 1,100 and 1,600 °C and at 25 GPa determined in the MgO–FeO–SiO<sub>2</sub> system by Ito and Takahashi (1989) are also shown in Fig. 2.

If the chemical composition of the Earth’s mantle is approximated to be similar to pyrolite, the FeO content of ferropericlase in the lithology of the lower mantle must be  $< 20$  mol%. As shown in Fig. 1, the Kankan diamonds alone contain 41 ferropericlases, which are all relatively enriched in MgO content, and only two have FeO contents of  $> 20$  mol%. Ferropericlases of Brazilian diamonds from both Juina and Sao Luiz (42 in total) are all enriched in FeO, and only four have FeO contents of  $< 20$  mol%. Thus, worldwide, there are about 44% ferropericlase inclusions that have FeO contents of  $> 20$  mol%. Not only are these ferropericlases enriched in FeO, but the FeO content is widely spread (FeO = 6–88 mol%). Thus, it is highly unlikely that all these ferropericlases represent the main lithol-

**Table 1.** Rare inclusions discovered in diamonds. The numbers in parentheses after the minerals are the number of diamonds in which the mineral assemblage is found

Reference	Locality	Inclusions
Newton et al. (1977)	Arkansas, USA	Periclase + magnetite
Scott-Smith et al. (1984)	Orroroo, S. Australia	Ferropericlase (2) <sup>a</sup> , enstatite
Moore et al. (1986)	Monastery, S. Africa	Moissanite (1), moissanite + garnet (2), magnesiowustite (1)
	Sloan, Colorado	Moissanite (1), moissanite + diopside (1), moissanite + sulfide (?) (1), ferropericlase (1)
	Koffiefontein, S. Africa	Ferropericlase (3), ferropericlase + enstatite (1)
Moore and Gurney (1989)	Monastery, S. Africa	Ferropericlase
Otter and Gurney (1989)	Sloan	Ferropericlase
Harte and Harris (1994)	Sao Luiz, Brazil	Ferropericlase (14), ferropericlase + SiO <sub>2</sub> (1) Ferropericlase + enstatite (3), ferropericlase + CaSiO <sub>3</sub> (1), ferropericlase + TAPP (3), ferropericlase + enstatite + TAPP (1)
Harte et al. (1999)		
McDade and Harris (1996)	Letseng-La-Terrai, Lesotho	Ferropericlase
Kopylova et al. (1997)	River Ranch, Zimbabwe	Ferropericlase
Hutchison (1997)	Guinea	Ferropericlase (2)
Stachel et al. (1998)	Mwadui, W. Tanzania	Ferropericlase (1)
Stachel et al. (2000)	Kankan, Guinea	Ferropericlase (27), ferropericlase + enstatite (2), ferropericlase + SiO <sub>2</sub> (1), ferropericlase + enstatite + siderite (1), ferropericlase + enstatite + Mg-hornblende (1), ferropericlase + enstatite + CaSiO <sub>3</sub> + MgSiO <sub>3</sub> -Mg <sub>2</sub> SiO <sub>4</sub> (1), enstatite + CaSiO <sub>3</sub> (2) + .....
Kaminsky et al. (2001)	Juina, Brazil	Ferropericlase (23), ferropericlase + SiO <sub>2</sub> (1), ferropericlase + ilmenite (2); ferropericlase + “olivine” (1), ferropericlase + spinel (1)

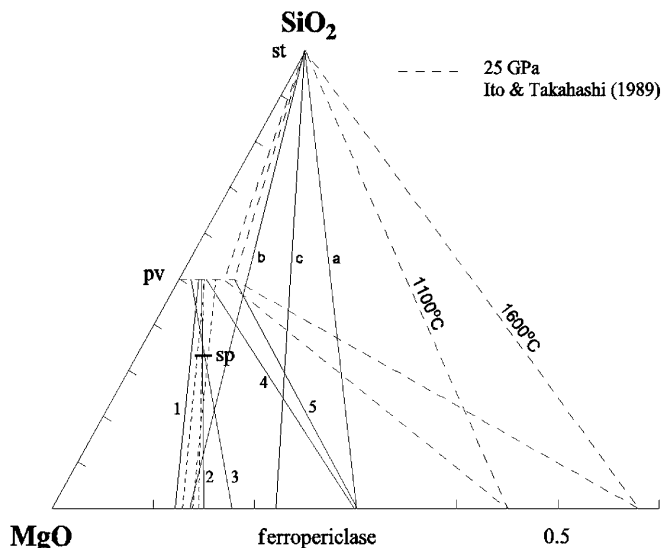
<sup>a</sup>Number of diamonds



**Fig. 1.** Histogram of the Mg# of ferropericlasite inclusions in diamonds from various localities. The numbers inside the parentheses represent the number of inclusion at each locality

ogy of the lower mantle. In order to avert this difficulty, Harte et al. (1999) proposed that the FeO-rich ferropericlasites may originate from the D'' zone at the mantle-core boundary, and are transported to the shallow parts of the lower mantle by plumes and then entrapped by diamonds at the top part of the lower mantle. The D'' zone has limited temperature and pressure ranges. Therefore, the FeO content of ferropericlasites formed from the D'' zone should be clustered in a narrow range. In other words, the FeO content of ferropericlasites found in diamonds should concentrate in two narrow regions (bimodal): one represents the lower mantle lithology and the other represents the D'' zone source. But, as shown in Fig. 1, except in the region  $\text{FeO} = 0.11\text{--}0.16$  (44 ferropericlasites), there are 41 ferropericlasites (mostly from Brazil) spreading widely in the range 0.20–0.88. Thus, the suggestion of a D'' zone origin is not substantiated by the data. Furthermore, Harte et al. (1999) proposed that both diamonds and their inclusions from Sao Luiz might grow simultaneously (see late discussion) in the top parts of the lower mantle. The latter suggestion does not seem to agree with the D'' zone scenario proposed for the FeO-rich ferropericlasites.

Ferropericlasites coexisting with enstatite or  $\text{SiO}_2$ -inclusion in the same diamond are shown as tie-lines in Fig. 2. If these diamonds were derived from the lower mantle, then enstatite and  $\text{SiO}_2$  inclusions should be former  $(\text{Mg,Fe})\text{SiO}_3$ -perovskite and stishovite, respectively. There are four Kankan diamonds containing both ferropericlasite and enstatite; three of these also contain other minerals such as siderite, Mg-hornblende, etc. The latter diamonds and their inclusions appear to have a complicated history (Stachel et al. 2000). However, the FeO content for both ferropericlasite and enstatite inclusions in these four diamonds is very similar to each other and is shown by three short dashed tie-lines in Fig. 2. In addition to the above Kankan diamonds, there are only five diamonds containing both ferropericlasite and enstatite



**Fig. 2.** The MgO-rich and  $\text{SiO}_2$ -rich portion of the phase relations in the MgO–FeO– $\text{SiO}_2$  system (in molar composition) at 1,100 and 1,600 °C and 25 GPa determined by Ito and Takahashi (1989). The solid tie-lines connect the compositions of ferropericlasite and enstatite inclusions found in the same single diamonds from Koffiefontein (marked by 1) and Sao Luiz (marked by 2–5), and of ferropericlasite and  $\text{SiO}_2$  inclusions found in Sao Luiz, Kankan and Juina (marked by a, b, and c, respectively). Three short dashed tie-lines connect the compositions of ferropericlasite and enstatite inclusions found in four Kankan diamonds. st Stishovite; pv perovskite-type structure; sp  $(\text{Mg,Fe})_2\text{SiO}_4$ -spinel having  $\text{FeO}/(\text{MgO} + \text{FeO}) = 0.09\text{--}0.11$ , which is not a stable phase at the P,T conditions determined by Ito and Takahashi (1989), but is given for the convenience of discussion

as inclusions, and three diamonds containing both ferropericlasite and  $\text{SiO}_2$ -inclusion (all shown by solid tie-lines). These inclusions are discrete, single crystals that are not in physical contact inside the diamond and, therefore, they cannot chemically equilibrate or re-equilibrate with one another in response to changing pressure and temperature conditions. If they are real tie-lines, these inclusion pairs must be once in chemical equilibrium at certain P, T conditions in the lower mantle.

Combining the data of both Figs. 1 and 2, there are 94 ferropericlasites in total. Among them, ten (five in the Kankan diamonds) coexist with enstatite in the same diamond. If these inclusions represent the lithology of the lower mantle as suggested, the ratio of occurrence between ferropericlasite and enstatite observed in diamonds that have the lower mantle origin is in gross disagreement with the possible lithology of the lower mantle, which should be dominated by  $(\text{Mg,Fe})\text{SiO}_3$ -perovskite, unless the intercrystalline surface energies between diamond and these inclusion minerals changes the situation drastically. The latter is rather doubtful, however.

In summary, the data of Fig. 1 show that ~47% of the ferropericlasites have a  $\text{FeO}/(\text{MgO} + \text{FeO})$  ratio in the narrow range 0.11–0.16, and the remainder spread in the wide range of 0.06–0.88. The data of both Figs. 1 and 2

show that the occurrence of ferropericlasite is much more frequent than that of enstatite in these diamonds. Both facts do not seem to be appropriately explained in terms of the main lithology of the lower mantle. Thus, it is highly unlikely that all these inclusions represent the lithology of the lower mantle.

### Implication for lower mantle lithology

Ferropericlasite in the lower mantle should be derived from the breakdown of  $(\text{Mg,Fe})_2\text{SiO}_4$ -spinel, which exists in the transition zone at the high pressures of the lower mantle. The  $\text{FeO}/(\text{MgO} + \text{FeO})$  ratio for either olivine or  $(\text{Mg,Fe})_2\text{SiO}_4$ -spinel in the mantle is close to 0.1. In Fig. 2,  $(\text{Mg,Fe})_2\text{SiO}_4$ -spinel that has  $\text{FeO}/(\text{MgO} + \text{FeO}) = 0.09\text{--}0.11$  is marked by a horizontal bar denoted by “sp”. The composition of the pair of ferropericlasite and enstatite of the Koffiefontein diamond (marked by “1”) appears to be too rich in MgO compared with the upper mantle chemistry. The composition of the five ferropericlasites and six enstatites found in four Kankan diamonds (marked by short dashed lines), and that of the pair of ferropericlasite and enstatite found in two Sao Luiz diamonds (marked by “2” and “3”) appear to be consistent with that of the  $(\text{Mg,Fe})_2\text{SiO}_4$ -spinel in the transition zone. It is noticed that these tie-lines cross each other. If these are real tie-lines representing the breakdown product of  $(\text{Mg,Fe})_2\text{SiO}_4$ -spinel at pressures greater than that of the 650-km seismic discontinuity (it may have moved downwards slightly after the formation of the Earth), they should not cross each other because the pressure at the discontinuity is a fixed value. Thus, the crossing of the tie-lines seems to suggest that some pairs of inclusions in the same diamond are not once chemically equilibrated, or may not be entrapped by the diamond at the same time and/or in the same place (i.e., a repeated growth of diamonds).

The compositions of two of the diamonds that have ferropericlasite rich in FeO [ $\text{FeO}/(\text{MgO} + \text{FeO}) = 0.3$ ] from Sao Luiz (marked by “4” and “5”) are too FeO-rich for the composition of the lower mantle. Thus, in brief, the compositions for most pairs of ferropericlasite and enstatite inclusions found in the same single diamond do not appear to represent the lithology of the lower mantle.

If the diamonds from Sao Luiz were all formed in equilibrium at similar P and T conditions, then the tie-lines of “5” and “a” (ferropericlasite-SiO<sub>2</sub>) in Fig. 2 should represent the two sides of the three-phase region of stishovite,  $(\text{Mg,Fe})\text{SiO}_3$ -perovskite and ferropericlasite. Compared with the three-phase regions determined at 1,100 and 1,600 °C by experiment, it would suggest that the Sao Luiz diamonds were formed at a temperature much lower than 1,100 °C at ~25 GPa. This is a very unlikely temperature for the Earth’s lower mantle. The pairs of ferropericlasite and SiO<sub>2</sub>-inclusion found in the same diamond at Kankan and Juina are marked by “b”

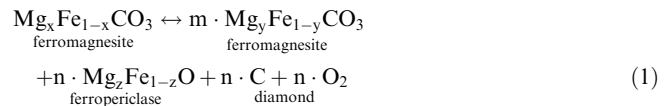
and “c”, respectively, in Fig. 2. In fact, the composition of all pairs of ferropericlasite and SiO<sub>2</sub> inclusions cannot be in equilibrium under any P, T conditions known by experiment (e.g., Liu 1976b; Yagi et al. 1979). Thus, all these facts suggest that the various mineral inclusions found in the same diamond are not in equilibrium at lower mantle P, T conditions and, instead, indicate a repeated diamond growth.

### Genesis of diamonds and ferropericlasites in the lower mantle

One may wish to know how the diamonds in the lower mantle were formed in the first place. Stachel et al. (2000) speculated that these diamonds may be derived from subducted oceanic plates accumulating at the top of the lower mantle, but recognized that the carbon isotopic composition of lower mantle diamonds indicates a primitive signature. Liu (1999) proposed that decarbonation of ferromagnesite in the P, T conditions of the lower mantle may form diamonds. Haggerty (1999) also pointed to the dissociation of magnesite in plumes as the possible origin of diamonds in the lower mantle.

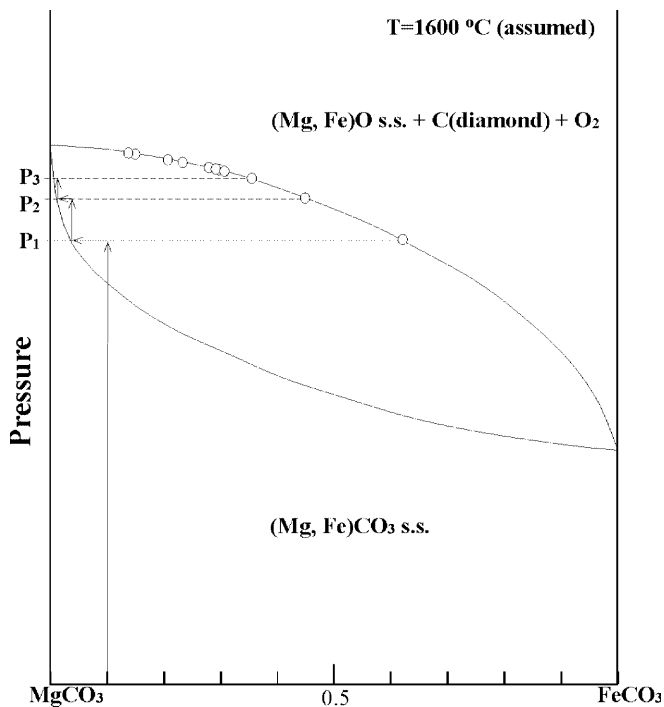
If lower mantle diamonds were indeed formed by decarbonation, one would expect that diamond and ferropericlasite grow simultaneously, and that ferropericlasite should be the most likely inclusions in these diamonds. The latter is substantiated by the data of Figs. 1 and 2, and the former appears to be supported by the observation of Harte et al. (1999) who reported that the mineral inclusions released from Sao Luiz diamonds have shown shapes indicating imposed diamond morphology, which demonstrates that the growth of diamond and inclusion occurred simultaneously.

Both the high value and wide range of FeO content in ferropericlasites can also be reasonably interpreted in terms of the same genesis of diamonds proposed above. Figure 3 displays the “two”-phase region of a plausible phase diagram for the  $\text{MgCO}_3\text{--FeCO}_3$  system at a given temperature for the following reaction:



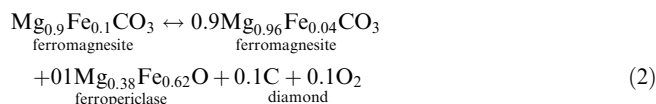
where  $m + n = 1$ ,  $x = my + nz$ ,  $m/n = (x - z)/(y - x)$  and  $1 > y > x > z > 0$  [Note that O<sub>2</sub> in Reaction (1) is in solid state.] Reaction (1) yields both ferropericlasite and diamond in the “two”-phase region at higher pressures. Thus, as concluded earlier, ferropericlasite should be the most likely inclusions in diamonds in the lower mantle. The oxygen produced in reaction (1) must have either reacted with other components in the mantle or escaped to the surface.

The FeO content of ferropericlasites found in the Sao Luiz diamonds spreads most widely. Thus, these samples are used here as a demonstration. The smallest



**Fig. 3.** Hypothetical “two”-phase region of the decarbonation reaction of the  $\text{MgCO}_3\text{--FeCO}_3$  system with increasing pressure at a constant temperature (assumed to be 1,600 °C). The *open circles* represent the chemical compositions of ferropericlase inclusions in diamonds found in Sao Luiz. It represents a case where ferropericlases were entrapped as inclusions in diamonds at certain pressures (such as  $P_1$ ,  $P_2$ , and  $P_3$  ...), but maintained in equilibrium with  $(\text{Mg,Fe})\text{CO}_3$ , diamond and  $\text{O}_2$  at other pressures

$\text{FeO}/(\text{MgO} + \text{FeO})$  ratio of the Sao Luiz ferropericlases is 0.14; therefore, 0.86 is the lower limit of  $x$ . Let us assume  $x = 0.90$ , which is close to that of the lower mantle. When pressure reaches  $P_1$  in Fig. 3,  $\text{Mg}_{0.9}\text{Fe}_{0.1}\text{CO}_3$  decomposes to 10% diamond plus ferropericlase [ $n = 0.1$  in reaction (1)]. We further assume that the ferropericlase that has the largest  $\text{FeO}/(\text{MgO} + \text{FeO})$  ratio is the disproportionate product (i.e.,  $z = 0.38$ ), then the values of  $m$  and  $y$  in reaction (1) can be calculated to be 0.90 and 0.96, respectively. Therefore, the diamond containing ferropericlase with the richest FeO content from Sao Luiz may be formed according to the following reaction:



Once  $(\text{Mg}_{0.38}\text{Fe}_{0.62})\text{O}$  is entrapped inside the diamond as an inclusion in the above reaction, the more MgO-rich ferromagnesite ( $\text{Mg}_{0.96}\text{Fe}_{0.04}\text{CO}_3$ ) becomes the new starting material. When pressure increases (or at an increasing depth) again, more MgO-rich ferropericlases may be entrapped in diamonds at  $P_2$ ,  $P_3$ ...

Thus, the FeO-rich ferropericlase inclusions do not have to be invoked to have a  $D''$  zone source in the

mantle-core boundary. FeO-rich ferropericlase and diamond can grow simultaneously at shallow parts of the lower mantle. More importantly, the observed large spread in the FeO content of ferropericlases can also be explained by the same scenario.

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