

## A simple model for the pressure preservation index of inclusions in diamond

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### ABSTRACT

The isovolume locus for a diamond-mineral inclusion pair occurs where the relative volumes of the two minerals respond identically to changes in pressure-temperature ( $P$ - $T$ ) conditions. Thirty potential inclusion minerals have been assessed with a simple linear model of this system. The remnant pressure on the inclusion (diamond at the Earth's surface) can be estimated by extrapolating the conditions of diamond formation to 0 °C using the slope of the isovolume locus. Remnant pressures can be positive (isovolume locus lower than formation pressure) or negative (locus higher than formation pressure), the latter indicating the inclusion has decompressed completely. When placed in order of increasing isovolume slope, this mineral list defines a pressure preservation index (PPI). Published work confirms that the model is quantitative up to diamond formation pressures of 50 Kb and qualitative beyond. Ten minerals are identified as key PPI indicators—in decreasing order of PPI they are: sanidine, coesite, dolomite, (sphen, garnet, diopside, zircon), magnesiowüstite, spinel,  $Mg_{0.9}Fe_{0.1}SiO_3$  perovskite. For most diamond formation conditions, inclusions of the first three minerals will retain high remnant pressures, whereas the last three will decompress completely. The central four minerals in brackets will have a highly variable response because their isovolume loci pass through diamond formation conditions. Preliminary calculations suggest that fluid inclusions ( $H_2O$ ,  $CO_2$ ) fall at or near the top of the list.

For inclusion-bearing diamonds at the Earth's surface, the model predicts: (1) remnant pressures; (2) confining pressure on inclusion during delivery; (3) the stability of high-pressure polymorphs; (4) retention vs. resetting of radiometric ages; (5) those minerals suited for determining the conditions of formation based on measured remnant pressure; (6) those minerals capable of stabilizing a microdiamond inclusion; and (7) that supercritical fluids preserve inclusions of microdiamond in most minerals.

### INTRODUCTION

Minerals included in diamonds have been used extensively to estimate the conditions of formation of diamonds. Mostly, this approach has depended on the mineral type and its composition. An independent technique involves estimating the remnant pressure on mineral inclusions sealed in diamond using lattice spacings determined by X-ray or Laser Raman spectra [see Liu et al. (1990), Izraeli et al. (1996), and Sobolev et al. (2000)]. To date, these measurements have indicated that some minerals are under little or no remnant pressure, whereas others remain under substantial remnant pressure. The main factor causing these differences is the contrast between the  $P$ - $T$  volume coefficients of the mineral and those of diamond. In order to characterize the diamond/inclusion system, a linear first-approximation model is used to assess thirty minerals according to their capacity to preserve the pressure of diamond formation (the pressure preservation index  $\equiv$  PPI). The minerals considered herein either have been found as inclusions in diamond, found as minerals in diamond-hosting rocks, or have been suggested as possible inclusions. However, the intention is not to focus on mineral stability, rather to provide a numeri-

cal framework showing how different minerals would respond theoretically as inclusions in diamonds during delivery to the Earth's surface. The model presumes that an inclusion is sealed in an inclusion chamber that remains pristine during delivery to the Earth's surface. Although most inclusion chambers in diamond show signs of fracturing and healing under cathodoluminescence imaging, the validity of the PPI model predictions will test whether this is an early stage growth feature or a late-stage one. Depending on these results, the model either would act as an upper limit, or would need to be modified. Initially the model is developed for silicates, oxides, and carbides. Sulfides dominate numerically as inclusions in diamonds around the world, so there is merit in extending the model to such minerals.

### BASIS FOR PPI

To a first approximation, the volume of the host mineral, diamond, varies across  $P$ - $T$  space according to the linear equation  $V_d(P, T) = V_d^0(1 + A_d T - B_d P)$  where  $V_d^0$  is the volume of diamond at the reference state (0 °C, 0 bars),  $A$  is the coefficient of thermal expansion, and  $B$  is the coefficient of compressibility ( $T$  in °C and  $P$  in Kb). The included mineral will have an analogous equation, namely  $V_i(P, T) = V_i^0(1 + A_i T - B_i P)$ . Rearranging each equation as a relative volume ( $V_d/V_d^0, V_i/V_i^0$ ).

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$V_i^0$ ) and subtracting yields the equation of the isovolume locus along which the relative volumes of both minerals respond identically to a change in pressure and temperature, namely  $P/T = (A_d - A_i)/(B_d - B_i)$ . As shown on Figure 1, these loci (for 30 minerals) pass through the origin with a positive slope, and represent “zero” remnant pressure for a reference state corresponding to the origin of the graph (0 °C, 0 Kb).

Liu et al. (1990) and Izraeli et al. (1996) have shown that a line parallel to this isovolume locus but offset by pressure  $P_r$  represents the approximate locus of diamond formation conditions that would result in a remnant pressure of  $P_r$ . Thus, within the framework of the simple PPI model, the slope of the isovolume locus can be used to generate lines of constant  $P_r$ . If a diamond forms at a pressure above the isovolume locus of a mineral, there will be positive remnant pressure on the mineral inclusion at the Earth’s surface. Conversely, if the diamond forms below the locus, the remnant pressure will be “less than zero,” and the mineral inclusion will be completely decompressed. Because of this relationship, minerals with a low slope on Figure 1 will have relatively high values of remnant pressure, whereas minerals with a steep slope will have low or negative values of remnant pressure. Thus the minerals, when sorted by increasing slope values, represent inclusions in a sequence with progressively lower remnant pressures (see Table 1). For the treatment of dense H<sub>2</sub>O and CO<sub>2</sub> fluids as inclusions, the volume coefficient data were selected from a  $P$ - $T$  region near the resultant isovolume locus, but well into their supercritical fields. Of course, these are fluids and not solids, so the calculations produce a “virtual” isovolume locus as if they were solids (see Table 1). Because they are supercritical fluids, positive remnant pressures >2 Kb will be representative, but anything below critical pressures (including calculated negative remnant pressures) actually will be replaced by reduced positive pressures. However, supercritical H<sub>2</sub>O and CO<sub>2</sub> have such low isovolume slopes (Table 1) that treating them like the other solid minerals is justified by the high remnant pressures the fluid inclusions invariably generate.

For the particular  $P$ - $T$  conditions of diamond formation, it is possible to convert the calculated remnant pressure to a factor that represents the degree of preservation of the pressure of formation, namely the ratio of the remnant pressure to the pressure of formation (hereafter, pressure preservation factor). Minerals toward the top of the list are better at retaining the

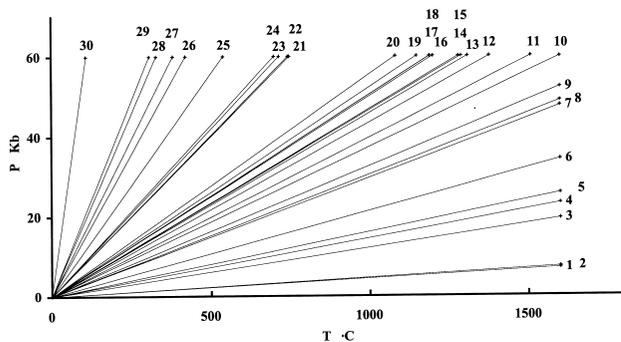


FIGURE 1. Pressure temperature ( $P$ - $T$ ) space with thirty isovolume loci labeled with mineral identification numbers (MN in Table 1).

pressure of diamond formation—the top six entries (CO<sub>2</sub> to phlogopite) produce remnant pressures greater than 50% of the pressure of formation. As Sobolev et al. (2000) stated, coesite is one of the best minerals in this regard, but supercritical fluid inclusions of CO<sub>2</sub> and H<sub>2</sub>O are predicted to be better than any solid minerals.

## DISCUSSION

Theoretically, measurements of the remnant pressure on a sealed mineral inclusion can be combined with the pressure preservation factor of the mineral to estimate the pressure of formation of the diamond. In practice, only the top 6 entries in Table 1 have preservation factors high enough to determine the pressure of formation reliably—these minerals belong to a “heritage” group of inclusions. In contrast, the minerals 21–30 (Table 1) will decompress completely for most diamond formation conditions. The isovolume loci for minerals 12–20 (Table 1) pass through the region of dominant diamond formation, so the decompression aspects of these minerals depends on the exact conditions of diamond formation. Preliminary cal-

TABLE 1. Minerals, identification numbers, and data

Mineral	MN	A <sub>i</sub>	B <sub>i</sub>	Source no.	Isovolume at 1600 °C
		×10 <sup>6</sup>	×10 <sup>6</sup>	A <sub>i</sub> , B <sub>i</sub>	$P$ (Kb)
Diamond		0.708	0.238	3,3	
CO <sub>2</sub>	1	520	120	25,25	6.9
H <sub>2</sub> O	2	860	188	25,25	7.3
sanidine	3	19	1.75	14,4	19.4
coesite	4	11.8	1.002	24,17	23.2
SiC	5	4	0.4425	1,2	25.8
phlogopite	6	32	1.7	23,10	34.2
quartz	7	59	2.2	12,12	47.5
glaucophane	8	27	1.1	13,13	48.8
tremolite	9	31.3	1.176	26,6	52.2
dolomite	10	37.4	1.22	22,19	59.8
sillimanite	11	14.4	0.585	29,30	63.1
enstatite	12	32.2	0.9615	15,9	69.6
lawsonite	13	34.7	0.98	14,14	73.3
sphene	14	25	0.76	14,14	74.5
jadeite	15	24.7	0.75	5,4	75.0
clinozoisite	16	27.8	0.78	14,14	80.0
grossular	17	25	0.72	24,10	80.6
diopside	18	33.4	0.886	28,28	80.7
fosterite	19	26.4	0.73	8,21	83.6
zircon	20	12.3	0.447	20,11	88.7
periclae	21	31.6	0.6238	8,8	128.1
magnesiowustite	22	33.2	0.641	31,16	129.0
ilmenite	23	30.2	0.59	28,28	134.1
kyanite	24	30	0.58	7,7	137.0
rutile	25	26.5	0.47	24,4	177.9
spinel	26	25.2	0.41	24,24	227.8
corundum	27	23	0.38	8,4	251.2
MgSi-perovskite	28	27.1	0.3831	8,8	291.0
stishovite	29	16.4	0.3185	8,18	311.9
Fe <sub>3</sub> C	30	43.4	0.3158	27,27	877.6

Notes: Data source numbers: 1 = Accuratus Web Site (2002); 2 = Ames Laboratory Web Site (2002); 3 = Berman (1994); 4 = Birch (1966); 5 = Cameron et al. (1973); 6 = Comodi et al. (1991); 7 = Comodi et al. (1997); 8 = Fei (1995); 9 = Flesch et al. (1998); 10 = Hazen and Finger (1976a,1976b); 11 = calculated from data in Hazen and Finger (1979); 12 = Helgeson (1978); 13 = Holland (1988); 14 = Holland and Powell (1985); 15 = Hugh-Jones = (1997); 16 = Jacobsen et al. (1999); 17 = Levien and Prewitt (1981); 18 = Luo et al. (2002); 19 = Martens et al. (1982); 20 = Wang (2002); 21 = Olinger (1977); 22 = Reeder and Markgraf (1986); 23 = Robie et al. (1979); 24 = Skinner (1966); 25 = Smith (1963); 26 = Sueno et al. (1973); 27 = Vaccadolo et al. (2002); 28 = Wechsler and Prewitt (1984); 29 = Winter and Ghose (1979); 30 = Yang et al. (1997); 31 = Zhang et al. (2002).

culations suggest that minerals 1–10 and 14–25 in Table 1 may be used to identify uniquely the mode of diamond formation using measured remnant pressures.

### Complexities

This model is simple because various complexities have been ignored. Some complexities are qualitative or even indeterminate, such as a multiphase inclusion in a diamond. Despite this latter complexity, there may be insights if the other minerals in the inclusion chamber have isovolume loci that lie on the same side of the diamond formation conditions (see below). Another qualitative complexity occurs if the inclusion capsule is locally damaged but rehealed; if this occurred during formation of the diamond, the model would act as an upper limit. Future application of the PPI model probably will show the significance of damaged inclusion capsules.

There are other types of complexities that are quantitative, i.e., they will change the actual remnant pressure value through some sort of correction process. Firstly, there is an algebraic correction, at the temperature of diamond formation, if the difference between the diamond formation pressure and the isovolume locus pressure is defined as  $P_a$ , then actual  $P_r = -(B_d - B_i) \cdot P_a / B_i$ , which reduces to  $P_r \approx P_a$  (used herein) because  $B_d$  is typically much smaller than  $B_i$  (Table 1). This approximation cannot change the sign of  $P_r$ , only its relative size. Tests have shown that the geometric developments and conclusions are the same regardless of whether the exact or approximate relation for  $P_r$  is used, while the geometric development is simpler with the approximation.

Other quantitative complexities include non-linear corrections. For example, Zhang (1998) has carefully investigated the influence of  $P$ - $T$  variation in the volume coefficients, the shape of the inclusion, and the elastic response of the diamond to the inclusion. Zhang (1998) and Izraeli et al. (1996) showed that remnant pressures greater than 5–10 Kb require significant non-linear corrections. Sobolev et al. (2000) considered all of these quantitative corrections for a coesite inclusion in a diamond from Venezuela, and determined a remnant pressure of 36.2 Kb. For this setting, the PPI model predicts an extrapolated value of 38.4 Kb, indicating the modest scale of the total quantitative correction required.

Significantly, these types of quantitative corrections are not required for the isovolume loci. For instance, Lui et al. (1990) and Izraeli et al. (1996) demonstrated that the fully corrected isovolume loci of garnet or olivine are basically straight lines up to 45 Kb, with a gentle curvature at higher pressures (concave toward higher temperature). Because this curvature is relative to diamond, the other minerals will respond in the same way. Thus, the simple linear model is reasonably accurate up to formation pressures of 50 Kb with regard to the placement of the mineral isovolume loci, and retains a relative rather than absolute reliability for higher-pressure regimes.

### Delivery path and confining pressure on inclusions

Delivery of a diamond from the conditions of its formation to the Earth's surface causes the confining pressure on an inclusion in the diamond to change from the diamond formation conditions to the remnant pressure at the Earth's surface. How

does the confining pressure develop between these end points during delivery? A simple volcanic delivery model involves two stages, namely rapid pressure drop to near zero, followed by a rapid drop in the temperature to near 0 °C. At the end of the first stage, diamond controls the volume available for the inclusion. This control results in the equation  $V_d/V_d^0 = 1 + A_d \cdot T_f = 1 + A_j \cdot T_f - B_j \cdot P_{OT}$ , where  $P_{OT}$  is the confining pressure on the inclusion at  $T_f$  (the temperature of diamond formation) at the Earth's surface ( $P = 0$  bar). Rearranging yields  $P_{OT} = -T_f \cdot (A_d - A_j) / B_j$ . Values for  $P_{OT}$  are greater than 2 Kb and increase down the mineral list in Table 1. These minerals never decompress completely during the first stage of the volcanic delivery path because the thermal expansion of diamond is lower than all other minerals, hence the sign of  $P_{OT}$  from the last equation must remain positive. However, on the second stage of the delivery path, the confining pressure on the inclusion would change from positive  $P_{OT}$  to the remnant pressure determined previously. Thus, minerals with an isovolume locus below the diamond formation conditions cannot decompress completely on any delivery path.

If the isovolume locus is at a higher pressure than the diamond-formation conditions, the confining pressure must become negative on the second stage of the delivery path. Figure 2 illustrates an example of extrapolation for a clinopyroxene inclusion from cratonic diamond-formation conditions, yielding a crossover temperature of 150 °C at  $P = 0$  Kb. At the Earth's surface (0 °C, 0 Kb), this inclusion will be under minus 7.6 Kb remnant pressure. The diamond at 0 °C would have to be placed under +7.6 Kb load pressure for the confining pressure on the inclusion to be returned to zero. Consider the triangle defined by the above values [(150 °C, 0 Kb on the inclusion, so  $P_c = 0$ ) – (0 °C, 0 Kb) – (0 °C, 7.6 Kb on the diamond so  $P_c = 0$ ), see Fig. 2]. Because the PPI model is linear, the hypotenuse of this triangle corresponds to a confining pressure of 0 Kb. Hence, any delivery path that enters this "cavity" triangle must result in complete decompression of the clinopyroxene inclusion. Analogous to the  $P_r$  approximation, a line parallel to the hypotenuse of the cavity triangle, but at a pressure 10 Kb higher, represents the condition where the confining pressure on the inclusion is 10 Kb. Thus, the PPI model shows how to contour  $P$ - $T$  space in terms of confining pressure for any delivery path.

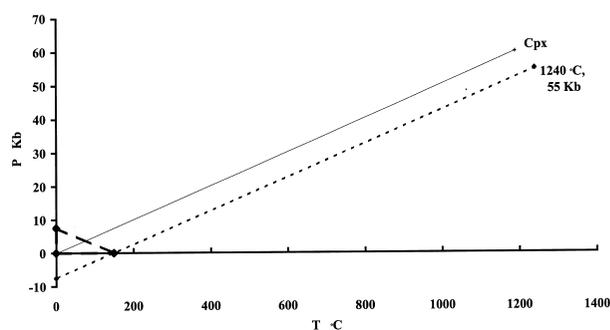


FIGURE 2. Pressure temperature ( $P$ - $T$ ) space with isovolume locus of diopside (MN = 18), extrapolation (dotted line) of cratonic diamond formation conditions to determine remnant pressure at 0 °C, and cavity triangle (heavy dashed line).

### Inclusions of high-pressure polymorphs

Magnesiowustite inclusions are found in some transition-zone diamonds (Scott-Smith et al. 1984), and the PPI model predicts a remnant pressure in excess of 100 Kb (for a diamond formation pressure of 180 Kb) so this mineral should retain the original high-pressure form. Coesite inclusions are reported as single crystals in diamonds from cratonic regimes (South Africa, South America), from mobile belts (Argyle Australia), and are common in diamonds from “deep leads” at Copeton (New South Wales, Australia, a non-traditional source). The remnant pressure on these coesite inclusions must remain high enough (18 Kb) to prevent coesite from inverting (the extrapolated remnant pressures are greater than 17Kb, indicating independent geological support for the simple PPI model).

Conversely, a high-pressure polymorph with an isovolume locus above the pressure of diamond formation will enter a cavity triangle that will allow it to invert on all delivery paths. Thus, the model predicts that stishovite and  $Mg_{0.9}Fe_{0.1}SiO_3$  perovskite (MgSi-perovskite in Table 1) inclusions should invert during delivery of a transition-zone diamond to the surface, and this behavior has been found by Kaminsky et al. (2002) in superdeep diamonds from Brazil.

### Mineral hosting inclusion of diamond

van Roermund et al. (2002) reported finding a microdiamond within a spinel grain formed in peridotite during a subduction collisional event that caused a doubling in the thickness of the crust in Norway. In the same rock, they reported interstitial graphite blebs, and speculated that spinel protected the microdiamond from graphitization during exhumation. The PPI model shows that spinel shrinks faster than diamond during delivery to the surface. As spinel is actually the host for the diamond, this volume change would generate a substantial remnant pressure on the microdiamond. The PPI model predicts that minerals 16–30 (Table 1) provide the “right” numerical environment for protecting most inclusions of diamond.

Can the remnant pressures be predicted if a mineral hosts a polymineralic inclusion including diamond? For instance, the microdiamond reported by van Roermund et al. (2002) was actually in a sealed chamber that included seven mineral grains (in approximate order of decreasing vol%: high density fluid, phlogopite,  $BaCaMgCO_3$ , spinel, diamond, periclase, dolomite, spinel, monazite). At first glance, this appears to be an indeterminate system. However, the five largest included minerals/fluids have isovolume loci that lie below the diamond formation conditions and below the spinel isovolume locus. During delivery to the Earth’s surface, these relationships should result in a high remnant pressure on the microdiamond, stopping it from inverting. In fact, preliminary calculations using the PPI model show that almost any mineral is capable of preserving microdiamond if the microdiamond is trapped in an inclusion chamber with supercritical fluid.

### Isotopic age dating of inclusions in diamond

The model has implications for interpreting isotopic age dates on mineral inclusions in diamonds. If a mineral inclusion is surrounded by a partial cavity during delivery to or cooling at the Earth’s surface, daughter products of the radioactive de-

cay could diffuse to this cavity, assisted by the high-vacuum equivalent of negative remnant pressure. Under these circumstances, it is highly likely that the age date will at least partially reset to the age of emplacement.

The isovolume loci of minerals 11–21 (Table 1) overlap with diamond-formation conditions. Thus, minerals in this range may reset in some diamonds and not in others, the result critically dependant on whether the formation conditions are just above or below the particular isovolume locus. As Figure 2 shows, a diopside inclusion will decompress completely for most cratonic diamonds, and so may reset. However, some diamonds may form above the isovolume locus of diopside, in which case they should retain the age of crystallization. Age dating of inclusions of mica or sanidine should give the age of crystallization for all diamonds formed within the Earth, due to low isovolume slopes on Table 1.

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