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The Composition and Significance of Gas Released from Natural Diamonds from Africa and Brazil

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Abstract

Analyses of the composition of gaseous inclusions in ten samples of natural diamonds have been made by mass spectrometric techniques. The primary or secondary nature of the inclusions was not determined with certainty. Analyses were made by crushing the diamonds in the high vacuum inlet system of a research mass spectrometer. The gases observed were composed of C, H, O, N, and Ar atoms. Water was the most abundant gas, followed in decreasing abundance by hydrogen, carbon dioxide, methane, carbon monoxide, nitrogen, argon, ethylene, ethyl alcohol, butene, and oxygen. By generalizing from the results and assuming the inclusions to be primary, some suggestions are made which specify conditions under which natural diamonds may have been formed. A model consisting of reactions of H-C-O atoms is given as a possible explanation for the observed gas.

Introduction

The fact that natural diamonds often contain entrapped foreign matter has been known for many years. Williams (1932) reported evidence for primary inclusions of diamond, garnet, chrome diopside, an additional green pyroxene, biotite, ilmenite, magnetite, chlorite and graphite. Of these, however, none were confirmed by X-ray or electron microprobe techniques. He suggested olivine, phlogopite, and zircon as additional possibilities. Mitchell and Giardini (1953) subsequently substantiated olivine by means of X-ray diffraction; Sharp (1966), pyrrhotite; Harris (1968), rutile; and Meyer and Boyd (1968, 1969, 1970, 1972), garnet, chromite, and enstatite.

In addition to crystalline impurities, Bauer and Spencer (1904) found evidence for liquid- and gas-bearing microscopic inclusions within diamonds. They interpreted the liquid to be CO_2 in some cases, and either H_2O or a saline solution in others, but this was not proven. Kaiser and Bond (1959) carried out mass spectrometric analyses of the gas released by graphitization *in vacuo* of diamond at 2000°C, and suggested that nitrogen is a common substitutional impurity. Subsequently, Melton, Salotti, and Giardini (1972), also using mass spectrometry, showed that some diamonds with primary crystalline inclusions yield, upon crushing *in vacuo* at 300°C, a gas composed of N_2, H_2O, CO_2, CH_4, Ar, with H_2 and CO possibly present. In this paper a primary inclusion in diamond is defined tentatively as one completely and totally enclosed within a diamond. No fractures are seen leading to the inclusions except for internal thermal expansion cracks. The inclusion may still be secondary if the fractures are annealed or the material has been altered by some other process. The location of the gas within the diamond, *e.g.* at an interface, is not specified, and it is, therefore, not possible to determine if it is primary or secondary relative to diamond.

The significance of primary included matter relative to the genesis of a crystalline phase needs no elaboration. Diamond is generally conceded to have formed at inaccessible depth within the earth. A qualitative understanding of all included matter is significant both to natural conditions of diamond growth, and to conditions that existed within the earth at the time and place of their formation.

The principal uncertainties associated with the earlier work of Melton, Salotti, and Giardini (1972) were possible gas contamination from the crushing apparatus, possible interaction and sensitivity of the analytical system relative to minute quantities of gas, and the fact that only two samples were investigated. A systematic study has been conducted whereby the characteristics of the experimental system have been evaluated, and the gas contents of ten additional diamond samples from a variety of localities have been analyzed. The existence of an included gas phase of a generally consistent composition (including H_2 and CO) has been confirmed for diamonds that bear primary crystalline inclusions. Details of the study are given below.

Experimental

The study of included material in diamonds usually involves extremely small quantities of material; thus, high sensitivity instrumentation is essential. Furthermore, to study gaseous inclusions, the apparatus must be capable of both releasing and analyzing the gas in an uncontaminated state. The research mass spectrometer, together with the specially designed diamond crusher used in this study, meets these experimental conditions.

High sensitivity is obtained by maximizing the efficiency of the spectrometer's detector, and the transmission of ions in a combined electric and magnetic field. The relationships to be satisfied are

$$\frac{d\mathbf{p}}{dt} = e(\mathbf{E} + \mathbf{V} \times \mathbf{B}) \tag{1}$$

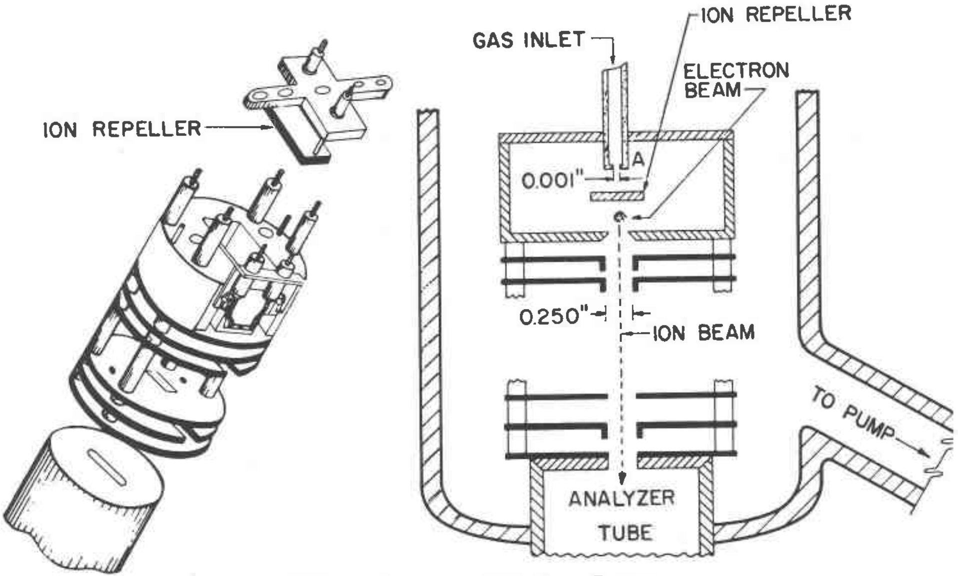
and

$$\frac{dE_e}{dt} = e\mathbf{V} \cdot \mathbf{E}, \tag{2}$$

where e is the charge on the electron, $\mathbf{p}$ is the momentum, $\mathbf{E}$ is the electric field, $\mathbf{V}$ is the velocity, $\mathbf{B}$ is the magnetic field, and E_e is the energy. In a well designed ion source, $d\mathbf{p}$ and dE_e are always in the direction of the ion exit slits.

Equation (2) is easily satisfied by requiring $\mathbf{E}$ to be in the direction of $\mathbf{V}(\cos 0° = 1)$. This restriction would also satisfy equation (1) for the special case of $\mathbf{B} = 0$. However, in electron-bombardment ion sources, the special case cannot be realized because a weak magnetic field is necessary to collimate the ionizing electrons. Consequently, the $\mathbf{V} \times \mathbf{B}$ term of Eq. (1) must be minimized. To do this, a geometrical approach is used.

Consider the high transmission ion source shown schematically in Figure 1. The effect of $\mathbf{V} \times \mathbf{B}$ is greatest in the region near the electron beam where $\mathbf{E}$ is small and $\mathbf{B}$ is large. By constructing a large ion-exit slit in the ionization chamber, large with respect to the diameter of the electron beam, ions with small momenta directed away from the normal ion path can be transmitted through the ion source.



Mass Spectrometer Ion Source

FIG. 1. Schematic diagram for the high transmission ion source used with the research mass spectrometer.

Thus, ions with a component of velocity away from the slit system can be collected.

The ion exit-slit in the present high-transmission ion-source measures 0.635 by 1.27 cm, and ions with appreciable components of velocity in various directions are transmitted. The field of the analyzer magnet subsequently corrects for small deviations in angle of incidence of the ion beam to give a high transmission through the analyzer section. These exit slits significantly improved the transmission efficiency of the ion source, and an efficiency (ions produced *vs* ions collected) of 88.3 percent has been measured. This was done by comparing the measured value of the total ion current collected on the ion repeller (Figure 1) for a negative bias with that collected at the detector for a positive bias.

A source with wide exit slits usually gives extremely low resolution. The present instrument has resolution of about 130. Fortunately, gaseous inclusions in diamonds contain small molecules (mass < 100); hence, the low resolution does not adversely affect results except for CO and N_2, which both appear at approximately 28 amu[1]. The mass difference for these molecules is about one part in 2500. To distinguish between these species, techniques other than mass separation must be used with the present instrument.

One technique is that of comparing the $14(N^+)/28(N_2^+ + CO^+)$ ratio with that of $12(C^+)/(28(N_2^+ + CO^+)$. In pure CO, the $12(C^+)/28(CO^+)$ ratio is constant to within 0.01 percent for identical experimental conditions of the instrument. The $14(N^+)/28(N_2^+)$ ratio is constant from N_2. Thus, comparison of the ion intensities of 12, 14, and 28 can be used to distinguish between equal quantities of N_2 and CO to a precision of about 0.1 percent.

The high transmission ion source is a component of a 15.24 cm radius 90°-sector type of magnetic scanning research instrument constructed in the University of Georgia shops. The gas-handling system of the instrument is made of borosilicate glass. A three-stage oil (silicone) diffusion pump evacuates the gas-handling system to a base pressure of better than 10^{-7} torr. Base pressure in the analyzer section of the instrument, which is evacuated by two three-stage oil diffusion pumps, is about 5×10^{-9} torr with liquid N_2 trapping. The analyzer section of the instrument is constructed of inconel. All other components are constructed of stainless steel.

The ion-detection circuit of the high-sensitivity research instrument consists of a 14-stage electron multiplier, attached to a vibrating-capacitor amplifier. A strip-chart recorder attached to the amplifier gives a permanent record of each signal. Calibration for linearity is done by measuring the isotopic abundance of neon on three different scales and comparing the experimental results with the known value. Linearity is better than 0.25 percent over all ranges.

The background ion intensity at each of the mass positions under study is determined both before and after each diamond is crushed. No significant change in background was observed during the course of a study. Thus, the experimental data could be easily corrected for background.

The released gas from the diamond flows from the crusher into the ionization chamber by means of a pinhole leak of about 10^{-4} cm diameter. A glass two-liter sample reservoir between the leak and the crusher maintains a rather constant pressure over the time of an experiment (~5 minutes). The gas pressure in the sample system is measured with an MKS baratron gauge.

Ions are formed by bombarding the gas with a 50 μ-amp beam of electrons of 100 eV energy. Electronic regulation is used to maintain constant electron emission and energy.

A possible major source of contamination of the sample is gas released from the elements of the diamond crusher described by Melton *et al* (1972). Several materials have been tested for use as crushing pistons. This was done both with no diamond in the crusher, and with a 3 carat, well-crystallized, type 1-a diamond which has remarkable resistance to crushing. Little gas was released from the crusher until the pistons were struck very hard blows, resulting in significant piston damage. This observation provided evidence that the observed gas was released from the diamond rather than from the crusher since inclusion-bearing diamonds crush "easily," and with little or no damage to the pistons. Experiments in which partial destruction of the pistons occurred were discarded because of possible contamination from the pistons.

Several materials were tested for use as crusher pistons. Stainless steel pistons gave the largest amount of gas contamination, and hydrogen-sintered high-temperature vacuum-annealed tungsten gave the least. Results for H_2-sintered tungsten carbide and for degassed tungsten are given in Tables 1 and 2. Those for stainless steel are in Table 3. Results in

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[1] Atomic mass units.

TABLE 1. Typical Volumes and Composition of Gases Released from H_2-Sintered Tungsten Carbide (6 wt percent Cobalt) Pistons Installed in Diamond Crusher

	Gas from typical shock needed to crush diamonds, with no detectable damage to pistons	Gas from shocks imparting some fragmentation of the pistons
Volume released**	8.6×10^{-7} cc	3.2×10^{-5} cc
Gas	Volume %	Volume %
H_2	86.7	35.4
CH_4	6.8	6.2
H_2O	0.0	0.0
C_2H_4	0.0	41.0
N_2	0.0	0.0
CO	1.7	12.8
C_2H_5OH	0.0	0.0
O_2	0.0	0.2
C_3H_6	0.0	0.0
Ar	0.3	0.9
CO_2	4.5	3.5

*Pistons and crusher were baked at 200°C for at least 72 hours in a vacuum of 10^{-8} torr prior to testing.
**Based on the following standards for calibration: 8.9×10^{-5} cc sealed capillaries of air crushed in instrument; 1.7×10^{-4} cc sealed capillaries of air crushed in instrument; 3.35×10^{-4} cc sealed capillaries of air crushed in instrument.

Tables 1 and 2 will be discussed further in connection with the analyses of gases from crushed diamond crystals. Experience with type 1-a diamonds with no microscopically visible inclusions suggests that these diamonds would be the ideal material for crusher pistons. They are extremely durable.

The apparatus was calibrated by crushing sealed borosilicate glass capillaries having known volumes of air. Mass spectrometric results agreed to within ±5 percent for both volume and composition to results obtained by other methods.

Since this study was made to explore gas included within diamonds, only structurally-sound crystals that contained completely-enclosed inclusions were used. A careful microscopic examination of the larger crystalline inclusions in one specimen revealed irregular opaque regions on the diamond-inclusion interface. These particular opaque areas could indicate either an additional solid phase (Harris, 1972; Harris and Vance, 1972), or a fluid-containing discontinuity (possibly secondary) across which total internal reflection of light has occurred. Bauer and Spencer (1904) described such phenomena according to the latter interpretation.

TABLE 2. Typical Volumes and Compositions of Gases Released from H_2-Sintered High-Temperature Vacuum-Annealed 95%-W 3.5%-Ni 1.5%-Fe Pistons in the Diamond Crusher

	Gas from typical shock needed to crush diamonds with minor piston deformation	Gas from shock giving significant piston deformation
Volume released*	2.3×10^{-7} cc	1.8×10^{-6} cc
Gas	Volume %	Volume %
H_2	75.0	39.4
CH_4	0.0	7.2
H_2O	0.0	26.9
N_2	0.0	13.7
O_2	0.0	0.6
CO_2	25.0	12.2

*Pistons and crusher were baked at 200°C for 72 hours in vacuum of 10^{-8} torr prior to testing.
**Calibration same as for Table 1.

Any "leak" from the present atmosphere into enclosures within diamond specimens would most likely be a reversible process; that is, the gas would be evacuated during "preconditioning" of the diamond in vacuum. The latter presently consists of maintaining the crusher containing the crystal at a constant temperature of 200°C while open to the high-vacuum of the spectrometer over a period of many hours. If ancient leakage of gas into the diamond from the kimberlite had occurred, and then sealed by some mechanism, then this source of possible contamination cannot be eliminated. Further details of the experimental techniques are given by Melton (1970, 1972).

The diamond specimens to be used were first examined under a 45-power binocular microscope to be sure that the inclusions appeared to be completely enclosed within the crystal. They were then cleaned with HF, boiled in aqua regia, and rinsed at least ten times with distilled, deionized water. The glass vials in which the diamonds were stored were washed and similarly rinsed.

Results

When diamonds were crushed in the diamond crusher at a pressure of 10^{-8} torr and a temperature of 200°C, the mass spectra revealed a relatively

TABLE 3. Typical Volume and Composition of Gas Released from Stainless Steel Pistons Installed in the Diamond Crusher

Gas from typical shock needed to crush diamonds, with significant piston deformation. Volume released** 2.7×10^{-4} cc

Gas	Volume %	Gas	Volume %
H_2	33.2	CO	5.4
CH_4	12.1	O_2	2.3
H_2O	7.4	CO_2	15.6
N_2	12.6	Other hydrocarbons	11.3

*Pistons and crusher were baked at 200°C at least 72 hours in a vacuum of about 10^{-8} torr prior to testing.
**Calibration same as for Table 1.

TABLE 4. Volume Percent Composition and Volumes of Gases Released from Diamonds Crushed at 200°C under a Vacuum of about 10^{-8} Torr

	SOURCE OF DIAMONDS					
	Africa					South America
	39 small octahedral flats with inclusions*	Type Ia with inclusions**	Type Ia with inclusions**	Type IIa with a few small inclusions+	Black cube, Congo	Translucent Tan cube, Brazil++
	Volume percent					
Volume (cc)	1.9×10^{-5}	4.8×10^{-6}	1.6×10^{-6}	1.8×10^{-6}	4.0×10^{-5}	3.5×10^{-5}
Gas						
H_2	1.8	29.4	43.1	59.0	1.6	1.6
CH_4	0.6	8.2	12.9	8.4	5.8	4.0
H_2O	9.6	33.2	17.2	22.5	63.6	85.1
C_2H_4	0.5	0.7	0.0	0.0	0.4	0.2
N_2	38.4	3.7	2.6	0.0	2.4	1.7
CO	45.8	9.4	14.7	0.0	0.0	0.0
CH_3CH_2OH	0.05	2.0	0.0	0.0	3.0	0.1
O_2	0.0	0.0	0.0	0.0	0.0	1.9
C_3H_6	0.3	1.2	0.0	0.0	2.0	0.0
Ar	0.9	1.2	0.0	0.0	0.4	0.05
CO_2	2.0	10.0	9.5	10.1	20.8	5.4

*African production
**Kimberly Mine: light-yellow rounded hexoctahedron (~3 cts.).
+Premier Mine: colorless rectangularly-shaped fragment (~3 cts.).
++Donated by B. J. Nameth, Kennametal, Inc.

complex gas phase. The major constituents were H_2, CH_4, H_2O, C_2H_4, N_2, CO, C_2H_5OH, O_2, Ar, C_3H_6, and CO_2. Water was especially abundant in some of the diamonds that have been crushed, as shown in Table 4.

To obtain clear evidence that the gases observed were in fact released from the diamonds and not from the apparatus, several tests were performed. First, direct comparison to calibration data showed the amount of gas released from the crusher was an order of magnitude too low to account for the gas observed from inclusion-bearing diamonds. Furthermore, the composition of gas released from the crusher was quite different from that released from diamonds. For example, no H_2O was released from the crusher (Tables 1 and 2), whereas H_2O is a persistent constituent in gases released from diamonds that contained inclusions (Table 4).

To determine gas volumes by mass spectrometry, the response of the instrument to pure gases is utilized. The method involves measuring the ion intensities from a gas of known composition at several different pressures as measured by a MKS baratron gauge. A calibration curve of volume *vs* intensity is thereby established. Since the volume of the instrument is known, a knowledge of the pressure can then be used to determine the volume of the gas sample. All experimental results showed the volume *vs* intensity curve to be linear over the pressure range from 10^{-7} to 10^{-8} torr except for H_2O. At higher pressures, $>10^{-6}$ torr, the H_2O intensity became nonlinear, increasing more slowly than the H_2O pressure.

A complete analysis of the gas released from diamond also required an accurate knowledge of instrumental background (ion beams produced from residual gases in the instrument). The background was determined before and after each crushing experiment. If it changed significantly, indicating either electronic instability or atmospheric contamination during an experiment, the experiment was terminated. In addition, the ion intensities of masses 28 N_2 and 32 O_2 were monitored with the system closed for at least 5 minutes before each experiment to preclude the possibility of leaks into the system from the atmosphere. Further evidence that leaks from the present atmosphere did not occur is gleaned from the fact that significant amounts of O_2

were observed only from diamonds of cube form which crush very easily (see Tables 4 and 5).

Before a comparison can be made between the observed gases in diamonds from different sources, the absolute accuracy of the data from each experiment also must be examined. As noted in Table 1, gas volumes as large as about 8.6 × 10^{-7} cc were released from the crushing apparatus in typical experiments necessary to crush diamonds with no or possibly minute inclusions. Consequently, an experiment with a diamond which releases less than 1 × 10^{-6} cc of gas is not interpreted quantitatively.

Another aspect of the experimental conditions which must be considered is that of heat generated by the crushing of the diamond. The actual amount of heat generated upon crushing a diamond cannot be measured by present experimental techniques. However, indirect evidence indicates that the intensity of heat generated within the diamond is below 1200°C. This is based on the fact that no evidence was observed for the conversion of the crystal to graphite. Graphitization occurs rapidly at 1200°C under vacuum conditions. Furthermore, even if the temperature reached 1200°C, little reaction between the gases would occur in the brief time needed for fracture of the crystal. It is concluded, therefore, that the measured composition of the gas is probably close to that entrapped in the diamond at present. It should be pointed out, however, that the composition of the gas within the diamond most likely has changed subsequent to its entrapment.

The pressure of the gas within the diamonds that have been crushed could not be determined. This is true because the pressure of the gas within the diamond is related to the volume it occupies. This volume is unknown. Part of it may be thermal expansion cracks. A calculation of the relative volume change for olivine and for garnet in diamond upon reduction of an assumed high temperature and pressure predicts that the internal gas pressure should be relatively low. The small volumes of gas released from the diamonds upon crushing (~10^{-5} cc at standard conditions) is not inconsistent with the calculation.

TABLE 5. Percent Composition and Volume of Gases Released from Diamond Cubes Crushed at about 200°C under a Vacuum of about 10^{-8} Torr

	SOURCE OF DIAMONDS				
	Congo			Brazil	
	Yellow, Translucent	Green, Translucent	Black, Opaque	Tan, Translucent	
	Volume %				
Volume (cc)	9.1 x 10^{-5}	9.6 x 10^{-5}	4 x 10^{-5}	3.5 x 10^{-5}	1.1 x 10^{-5}
Gas					
H_2	1.0	2.9	1.6	1.6 *	8.4**
CH_4	3.7	2.8	5.8	4.0	5.1
H_2O	46.4	72.5	63.6	85.1	76.1
C_2H_4	0.0	0.0	0.4	0.1	0.0
N_2	2.6	1.6	2.4	1.7	2.5
CO	12.9	0.0	0.0	0.0	0.0
CH_3CH_2OH	2.5	0.0	3.0	0.1	0.0
O_2	1.2	0.1	0.0	0.9	2.0
Ar	0.3	0.0	0.4	0.05	0.0
1-butene	0.0	0.0	2.0	0.0	0.0
CO_2	28.1	20.1	20.8	5.4	5.9

*first crushing, carbide pistons (see Table 1).
**second crushing, tungsten pistons (see Table 2).

The volumes of gas released upon crushing the diamonds appear to be a direct function of the number and size of crystalline inclusions. Typically, diamonds without noticeable inclusions yield the smallest amount of gas relative to the carat weight of the diamond. Diamonds with a large number of readily noticeable inclusions yield much larger amounts of gas per carat.

The most accurate results are probably those from experiments with diamond cubes. They crush very easily and release large volumes of gas (see Table 5). It is interesting to note the good reproducibility obtained with the tan-colored cube from Brazil[2] (Table 5). Essentially the only difference between the original crushing and a subsequent crushing of large recovered fragments was in the percentages of H_2 and H_2O. The two experiments were carried out under different conditions as separate experiments. Consequently, the observed difference in the percentage of hydrogen and water cannot be attributed to the crusher elements, because the carbide pistons (see Table 1) used in the former experiment release more gas upon shock than do the tungsten pistons (see Table 2) used in the latter experiment. One may conclude, therefore, that the concentration of H_2 (and other gases) may possibly vary with location within a given diamond. This observation on one sample agrees with fluctuations in impurity concentration reported by Frank and Lang (1965), and may imply either environment changes during the diamond growth, discontinuous growth, or both.

In summary, our results show that H_2, H_2O, CO_2, and CH_4 are persistent molecular compounds of the gas included in diamonds. Large (~3 carats) well-crystallized octahedra and hexoctahedra thus far have been consistently high in hydrogen and low in nitrogen. Small (~0.005 carat) flattened octahedra,

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[2] Kindly donated by B. J. Nemeth, Director of Research, Kennametal, Inc., Greensburg, Pennsylvania.

on the other hand, have thus far shown low H_2, but high N_2, content (refer also to the earlier analysis of small flattened octahedra). Cubes are found to contain large volumes of gas relative to other crystal forms, abundant H_2O, and often also a small quantity of molecular oxygen. The oxygen could have been due to contamination from the atmosphere, but we have no experimental evidence to indicate that this is the case.

Conclusions

The results of this investigation are not inconsistent with diamond formation by a gas phase-solid reaction under a variable environment. Water was observed as a major component of the included gas and may suggest that free water was in the upper mantle during diamond crystallization.

By postulating gas-solid reactions between atoms C, H, and O as a mechanism for diamond formation, a series of equilibria can be assumed to occur. Initial reactions are:

$$C + O_2 \rightleftarrows CO_2 \qquad \langle 1 \rangle$$

$$H_2 + \tfrac{1}{2}O_2 \rightleftarrows H_2O \qquad \langle 2 \rangle$$

$$C + \tfrac{1}{2}O_2 \rightleftarrows CO \qquad \langle 3 \rangle$$

$$C + 2H_2 \rightleftarrows CH_4 \qquad \langle 4 \rangle$$

Since the above reactions do not yield gas compositions (see French, 1966) consistent with those observed in the present study, secondary reactions among products (CO_2, H_2O, CO, and CH_4) from initial reactions are needed to explain the experimental data. Such reactions begin to occur immediately after formation of the initial products and continue in time until equilibrium is established. Preliminary calculations indicate that the following secondary reactions combined with the primary reactions and explain the observed data qualitatively:

$$H_2 + CO_2 \rightleftarrows H_2O + CO \qquad \langle 5 \rangle$$

$$4H_2 + CO_2 \rightleftarrows CH_4 + 2H_2O \qquad \langle 6 \rangle$$

$$CH_4 + CO \rightleftarrows H_2O + H_2 + C_{\text{diamond}} \qquad \langle 7 \rangle$$

Reaction 5 is the well known water-gas reaction, and reaction 6 is the Giardini-Salotti (1969) reaction which occurs at high pressure.

These seven reactions are adequate to explain the composition of the gas released from natural diamonds, except for nitrogen and argon. The latter gases are assumed inert to the diamond-growth process. A thorough study is presently under way to calculate expected abundances of the gaseous inclusions using known and estimated equilibrium constants, the seven postulated reactions, and a modified French (1966) model for the C-H-O system.

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It should be pointed out that the observed gases may have been "spectator" molecules and in no way influenced the diamond growth process. We believe this not to be the case, but the present data do not eliminate this possibility.

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