

Genesis of diamond: a mantle saga

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Abstract

A model for the genesis of natural diamond is presented based on the physical, chemical and mineralogical properties and features of diamond. Optical studies suggest that individual diamonds have had complex growth histories in which growth and dissolution may have occurred. Growth was not always continuous nor did diamonds grow in necessarily similar chemical environments. Evidence for this is provided by variation in the nitrogen and trace element contents in diamonds as well as information from studies of the minerals included in diamond. Isotopic data suggest that diamonds formed from carbon whose sources varied isotopically. The possibility exists that some diamonds may be products of recycled subducted carbon, whereas others have formed from primordial material either through magmatic or metasomatic processes. It is also likely that most diamonds formed in the Archaean or Proterozoic. The cognate host rocks for diamond in the mantle were several but can be broadly grouped into eclogitic and ultramafic (peridotitic); however, in mineralogical and chemical detail these rocks are quite diverse. Although diamond is commonly found in kimberlite and in lamproite at the earth's surface, these two rocks are not genetically related to diamond formation. Instead they are the transporting vehicles in which diamond ascended rapidly from mantle depths to the crust.

Introduction

Although diamond has been a source of fascination, the origin of this mineral has for centuries perplexed man. Greek philosophers and medieval alchemists ascribed many mystical properties to diamond. When taken as a powder, voluntarily or involuntarily, it could, among other things, cure diseases, poison ones enemies or make the honest strong and agile. An unusual belief, especially held in Greece and India was that diamond could procreate itself—a boon to the owner of a diamond mine.

In 1772 Lavoisier demonstrated that diamond, like carbon, would burn in air. However, it was only later in 1797 that Smithson Tennant proved that diamond consisted of carbon. This led several gentlemen scientists of the nineteenth century to suggest that diamond was formed through the action of heat and pressure on plant remains (Des Cloizeaux, 1855; Goppert, 1862).

The discovery of diamonds in a volcanic rock (kimberlite) at Kimberley, South Africa in 1871 led to more scientific, and less philosophical studies. This did not, however, deter various authors from presenting opposing viewpoints, as summarized by Williams (1932). For example, Lewis (1887) considered that diamond formed in the crust as the kimberlite host rock solidified—the carbon being derived from coal and other carbonaceous material.

In contrast others maintained that diamonds had originally formed in ultrabasic rocks at depths, and were subsequently released as the rocks fractured upon incorporation into the kimberlite melt (Harger, 1905; Holmes, 1936). The

initial evidence for this latter idea was the discovery of diamond in an eclogite xenolith from kimberlite (Bonney, 1899). Du Toit (1906), Wagner (1914) and Sutton (1928) modified this idea and suggested that the eclogite and peridotite xenoliths were cognate with the kimberlite. Diamond was thus genetically related to the early crystallization of kimberlite magma.

The above two hypotheses have lasted until the present and have developed with some modifications into the phenocryst versus xenocryst schools (Dawson, 1980). For example, Gurney et al. (1979) and Harte et al. (1980) maintain that diamonds are genetically related to early crystallization products of kimberlite within the upper mantle and are thus phenocrysts. In contrast, Meyer and Tsai (1976a), Robinson (1978), and Meyer (1982 a,b) have argued that diamonds are accidental inclusions in kimberlite and thus are xenocrysts; the association of diamond and kimberlite being one of passenger and transporting vehicle.

Most scientists familiar with diamond concede that diamond has grown stably within the upper mantle (Kennedy and Nordlie, 1968; Meyer and Boyd, 1972; Orlov, 1973; Sobolev, 1974; Robinson, 1978). Omitted for purposes of this discussion are the polycrystalline aggregates of diamond (carbonado, framesite, boart) which have received little scientific study (Trueb and DeWys, 1969; 1971; Trueb and Barrett, 1972; Gurney and Boyd, 1982) and whose origin is even more uncertain than the single crystal diamond considered here.

Suggestions as to the source of carbon from which diamond forms have been diverse and range from coal and plant remains as favored in the 1800's, to carbon dioxide and methane today. However, whether or not the carbon is primitive or from recycled crustal material is a necessary question. Current studies on carbon isotopes (Deines, 1980; 1982; Milledge et al., 1983), as well as on nitrogen (Becker, 1982) and rare gases (Ozima and Zashu, 1983) bear on this question.

An important factor in understanding the formation of natural diamond is afforded by detailed examination of minerals included in diamond. These studies, mostly crystallographic and mineralogical, have been reviewed by Sobolev (1974), Meyer and Tsai (1976a), Harris and Gurney (1979) and Meyer (1982a). Isotopic studies of these small inclusions in diamond are now possible and future work should provide significant results concerning diamond and the evolution of the upper mantle.

This paper suggests a model for the genesis of diamond, and its subsequent passage to the earth's surface. Current interest in the evolution of the upper mantle and magma generation considers diamond to be an unreactive chemical probe from the depths. One aim of the discussion and model presented herein is to place the genesis of diamond within the correct context of mantle processes. It is also hoped that the discussion will remove various misunderstandings that are prevalent with respect to diamond and its relationship to kimberlite and other rocks. A subsidiary aim of this paper is to bring to the attention of mineralogists the large amount of important information contributed by physicists to diamond research, and equally to expose physicists to geological processes attendant on diamond formation and the subsequent history of diamond.

Although the host rocks for diamond at the earth's surface are kimberlite and lamproite, it is believed that these rocks are not genetically related to diamond. Accordingly, it is not the aim of this paper to dwell on the nature of the chemical and mineralogical differences between various kimberlites, and between kimberlites and lamproites. The interested reader is referred to Dawson (1980) for kimberlites, Mitchell (1984) for lamproites, and the proceedings volumes of the three International Kimberlite Conferences.

Physical features of diamond

A considerable amount of detailed study into the physics of diamond has been undertaken over the past 35 years (Berman, 1965; Field, 1979). Much of this research has significance to mineralogy and bears on the formation of diamond.

Figure 1a is a photograph of a typical clear and colorless diamond without any visible flaws. This clarity, shown by many diamonds, suggests to the observer crystallization in a single uninterrupted event. This is not the case. In Figure 1b is shown a polished and etched surface of a diamond displaying a series of geometrical layers—referred to as the stratigraphy of diamond (Harrison and Tolansky, 1964; Seal, 1965). These patterns were interpreted by Frank (1966) as being due to periodic growth on octahedral and

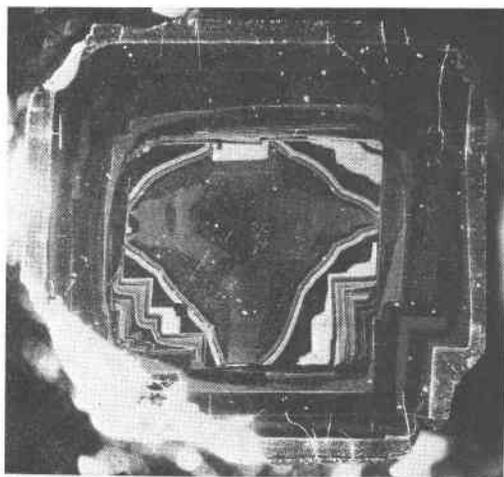
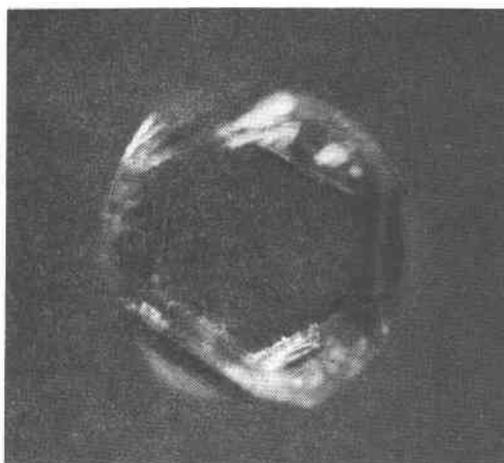


Fig. 1. (a) A transparent, clear and well shaped octahedron of diamond. Octahedral edge 1 mm. (b) A polished and etched section through an octahedral diamond (2.4 mm on edge), showing the internal stratigraphy of diamond. Various growth layers are either Type I or Type II diamond (Figure 3.1; Harrison and Tolansky, 1964).

cuboid surfaces. More detailed descriptions of this phenomenon are to be found in Suzuki and Lang (1976) and Lang (1979). The stratigraphy of diamond can also be illustrated by cathodoluminescence on polished surfaces of diamond (Moore, 1979) and by X-ray topography (Lang, 1979). The growth stratigraphy is observed because various layers consist of either Type I or Type II diamond, and these two types have different chemical and physical properties (Table 1).

The presence of Type I and II diamond was first demonstrated by Robertson et al. (1934) based on differences in UV and IR absorption. Lonsdale (1942) showed that Type I diamond produces extra X-ray diffraction reflections, or spikes. These spikes were interpreted to be due to platelets within the diamond structure (Frank, 1956). Kaiser and Bond (1959), and later Lightowers and Dean (1964) proved

the presence of nitrogen in Type I diamonds and showed a correlation between nitrogen content and optical absorption at $7.8 \mu\text{m}$ (1282 cm^{-1}). In contrast Type II diamond contains very little nitrogen and no platelets.

The above evidence suggests that the stratigraphy of diamond records periodic growth in chemically different environments, at least with respect to the amount of nitrogen present. Alternatively, the rate of crystallization, or length of residence time after partial growth may also contribute to differences in nitrogen content.

At high pressure it is possible to cause migration of the substitutional nitrogen in diamond so that the nitrogen aggregates (including platelets) found in natural diamonds are produced (Evans and Qi, 1982). Based on the results of this study Evans and Qi suggest that Type Ia diamond must have existed for between 200 million and 2000 million years. This range in time is admittedly large but is due to insufficient knowledge of the activation energy of migration of certain aggregates in diamond at high pressure and temperature. Nevertheless, the data indicate that some diamonds have had gestation periods in excess of the age of the kimberlite eruption that transported them to the crust.

In summary, although individual diamonds are grossly similar in physical properties detailed examination shows subtle differences resulting in four distinct types of diamond (Table 1). Growth of diamonds is crystallographically discontinuous and reflects possible variation in chemistry of the growth environment. Experimental aggregation of nitrogen to form platelets and other nitrogen aggregates in diamond suggests very long residence times for diamond within the upper mantle prior to reaching the earth's surface.

Chemical features of diamond

The presence of nitrogen as a major impurity in diamond has been noted, as also has the occurrence of boron (<20 ppm) which accounts for the semi-conducting properties of Type IIb diamond (Table 1). As a chemical sink for various elements, diamond is fairly poor with regard to concentration levels, although 58 elements have been recognized at the trace impurity level (Sellschop, 1979). Most of the results reported by Sellschop were obtained using instrumental neutron activation analysis, but early studies used emission spectrographic techniques (Chesley, 1942; Raal, 1957). Elements occurring in amounts greater than 1 ppm are listed in Table 2 (Sellschop, 1979), and most of these elements are typically present in silicate and sulfide magmas.

In a significant contribution, Fesq et al. (1975) suggested that in certain instances the trace elements in diamond were contained in sub-microscopic inclusions that represented quenched, or temperature re-equilibrated, melt from which diamond had crystallized; composition of the melt was thought to be picritic. At the time of the study (1972-73) Fesq and coworkers were able only to examine batches of diamonds, and thus the data represent diamond of different type and origin. Recent developments in analytical

Table 1. Some properties of diamond

Classification	
Type Ia	- Most common, approx. 98% of natural diamonds. Contains nitrogen up to 2500 ppm by wt. as aggregates and platelets
Type Ib	- Rare in nature but most synthetic diamonds are of this type. Nitrogen < 20 ppm by wt. in dispersed substitutional form.
Type IIa	- Very rare. Nitrogen < 20 ppm by wt. Often the very large gem diamonds are this type.
Type IIb	- Extremely rare and generally blue in color. Semi-conducting (B < 20 ppm by wt.). Most pure type of diamond.
Space Group: $Fd\bar{3}m - O_h^7$	
Unit cell: 3.56683 ± 0.00001 to $3.56725 \pm 0.00003 \text{ \AA}$	
Density: Type I - $3.51537 \pm 0.00005 \text{ gm cm}^{-3}$ Type II - $3.51506 \pm 0.00005 \text{ gm cm}^{-3}$	
Ultraviolet and Infrared	
Type I	- Strong absorption <340 nm and between 6 to 13×10^3 nm
Type II	- Transparent to 225 nm and between 6 to 13×10^3 nm
Color	
Type Ia	- Various (e.g. colorless, pale yellow, brown)
Type Ib	- Various (e.g. yellow, dark brown)
Type IIa	- Colorless, brown
Type IIb	- Blue
Thermal Conductivity	
Type Ia	- $600-1000 \text{ Wm}^{-1} \text{ K}^{-1}$ (at 293°K)
Type II	- $2000-2100 \text{ Wm}^{-1} \text{ K}^{-1}$ (at 293°K)
Resistivity	
Type I	- $>10^{18} \text{ ohm m}$
Type IIa	- $\text{ca.}10^{18} \text{ ohm m}$
Type IIb	- $10^3 - 10^5 \text{ ohm m}$

techniques (e.g. nuclear activation and ultra sensitive gamma ray spectroscopy) have enabled trace elements to be studied in a single diamond, and the results should provide significant information regarding the chemical differences between the various types of diamond.

Various diamonds have different trace element contents (Sellschop, 1979). This variation in trace element content suggests that different diamonds have not always formed under identical chemical conditions even though the diamonds may have been obtained from the same kimberlite pipe. If color is due to variation in trace element content,

Table 2. Elements present in diamond (maximum ppm by weight)

Element	Amount	Element	Amount	Element	Amount
H	1100	S	90	Co	2
B	10	Cl	4	Ni	80
N	5500	K	48	Cu	40
O	1700	Ca	195	Ag	30
Na	34	Ti	8	Ba	58
Mg	370	Cr	1000	La	14
Al	100	Mn	5	Ce	17
Si	80	Fe	140	Hg	5

Elements present in diamond < 1 ppm by wt: F, Sc, V, As, Rb, Sr, Sb, Cs, Th, Eu, Tb, Dy, Ho, Yb, Lu, Hf, Ta, W, Ir, Au.

Elements present in diamond but no absolute values available: Ar, Zn, Ga, Ge, Br, Zr, Sn, Nd, U, Sn, Gd, Er, Pt, Pb.

Data from Sellschop (1979, and pers. comm.)

then the wide range of colors in different diamonds from the same pipe can also be used, in part, to support the above argument (Robinson, 1978).

Isotopic studies

Geochemical information bearing on the genesis of diamond is available from studies on the isotopes of carbon, nitrogen and rare gases.

Carbon

On the basis of a few measurements, the $\delta^{13}\text{C}$ value for diamond was once considered to be generally in the range -4 to -9% (Craig, 1953; Wickman, 1956). More recently several authors (Galimov et al., 1978; Gurkina et al., 1979; Sobolev et al., 1979; Deines, 1980 and Milledge et al., 1983) have extended the range from $+3$ to -34% . Nevertheless, the majority of diamonds studied (e.g., Deines, 1980) have $\delta^{13}\text{C}$ values in the range -4 to -8% .

In a significant study, Milledge et al. (1983) have examined the $\delta^{13}\text{C}$ content of 18 Type II diamonds and obtained a range of values between 0 and -32% , although only two values were below -15% . It was concluded that Type II diamond is isotopically lighter than Type Ia diamond which presumably comprises the majority of diamonds, as noted above.

The existence of a range of $\delta^{13}\text{C}$ values for diamond has a bearing on the source of carbon and the chemical reactions that produce diamond. Recent models (Deines, 1980; Mitchell, 1975) as to the role of a vapor phase in diamond formation have proven ambiguous, although the presence of a vapor could help explain the range of $\delta^{13}\text{C}$. Deines (1980) concluded that this range was most likely inherited from the source carbon, suggesting isotopic heterogeneity in the mantle. Mitchell (1975) in contrast has pointed out that variation in the isotopic composition of diamond may be the result of re-equilibration (isotopic) of the solid phase with changing carbon-bearing gas compositions.

Discussed later is the fact that minerals included in diamond can be assigned to either an ultramafic or eclogitic suite. A careful study by Sobolev et al. (1979) involved examination of $\delta^{13}\text{C}$ values of diamonds in terms of whether they contained ultramafic or eclogitic suite inclusions. For diamonds with ultramafic inclusions the majority had a $\delta^{13}\text{C}$ value of -5% and a restricted range between -2 and -9% . Conversely, diamonds with eclogitic suite inclusions had $\delta^{13}\text{C}$ values ranging from 0 to -34% . As pointed out by Milledge et al. (1983), this latter range is similar to that of Type II diamond, whereas the peak at -5% for the ultramafic suite diamonds corresponds to that of Type I. A study of the type of diamond in which eclogitic inclusions occur has yet to be undertaken.

Nitrogen and rare gases

Attempts to determine the origin of diamond and the geochemical nature of the upper mantle have also involved studies of the isotopes of nitrogen as well as those of rare gases, notably helium. Data on nitrogen isotopes are sparse and consequently any conclusions are speculative. Wand et

al. (1980) obtained a mean value for $\delta^{15}\text{N}$ of $+1.5\%$, whereas Becker (1982) reported a mean value of $+2.9\%$ for five diamonds, with a range of 0 to $+5\%$. Both these mean values at present result in diamond being isotopically lighter in nitrogen than mantle rocks (Becker and Clayton, 1977) whose range is $+6$ to $+20\%$. Studies of rare gases in diamond (Ar, He, Ne) have been undertaken by Takaoka and Ozima (1978), Ozima and Zashu (1983), and Ozima et al. (1983). The latter authors reported $^3\text{He}/^4\text{He}$ values for three diamonds that were in excess of the primordial value for meteorites ($1.42 \pm 2 \times 10^{-4}$; Reynolds et al., 1978), and close to the solar value ($\sim 4 \times 10^{-4}$; Black, 1972). A second group of diamonds have much lower $^3\text{He}/^4\text{He}$ ratios. Ozima and coworkers propose that the diamonds with high $^3\text{He}/^4\text{He}$ ratios trapped primitive helium soon after the formation of the earth, whereas those with lower values may have formed later and included more evolved helium from the mantle.

Most of the variations in isotopic contents of diamond can be explained by considering that the carbon has passed through a subduction cycle (Frank, 1966). In contrast, very high $^{40}\text{Ar}/^{36}\text{Ar}$ ratios obtained by Takaoka and Ozima (1978) are interpreted as indicating that the carbon from which diamond formed has never reached the earth's surface; a similar comment is pertinent for the high $^3\text{He}/^4\text{He}$ values of Ozima et al. (1983). Melton and Giardini (1980) obtained a $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of 190 for a diamond from the Prairie Creek diatreme, Arkansas, well below that of the atmosphere which is taken as 296. Interestingly, this diatreme has recently been reclassified as a lamproitic kimberlite (Scott Smith and Skinner, 1984), whereas Mitchell and Lewis (1983) regard part of the diatreme to be a madupite. The data of Melton and Giardini (1980) represent the first published results of rare gas contents in diamond from a non-kimberlitic primary source (see also Roedder, 1984, p. 508-511).

Geochemical and isotopic studies show that diamond contains many elements in trace amounts, but generally those occurring in the largest concentrations are similar to those present in silicate and sulfide magmas. This suggests that diamond grew in a similar environment to most silicate-bearing rocks. Isotopic data, particularly the $\delta^{13}\text{C}$ values, show that the carbon from which diamond formed was isotopically variable. Furthermore, the variation in $\delta^{13}\text{C}$ between Type I and Type II diamond can be interpreted as diamond having formed in more than one environment, and one possibility is that some diamonds represent recycled crustal carbon. It is important that future isotopic studies be carried out on well-documented specimens, including geographic source and type of diamond.

Mineral inclusions

Various studies of minerals included in natural diamond are documented by Sobolev (1974), Meyer and Tsai (1976a), Harris and Gurney (1979) and more recently by Meyer (1982a). The significance of mineral inclusions with regard to diamond genesis and the upper mantle characterization was appreciated early (e.g., Bauer, 1896),

but apart from Sutton (1921) and Williams (1932) little work was done until the 1950's (Gübelin, 1952; Mitchell and Giardini, 1953; Futergendler, 1956, 1958, 1960). These were mostly optical and X-ray diffraction studies, and it was not until 1967 that the first electron microprobe analyses were obtained (Meyer, 1968; Meyer and Boyd, 1969, 1972). Since then others have contributed significantly to the study of inclusions (see Meyer, 1982a for references).

A number of results from these studies bear on the genesis of diamond. For example, most inclusions can be assigned to one of two mineral suites—an ultramafic suite and an eclogitic suite (Table 3). Members of the two suites are mutually exclusive; that is, minerals of one suite do not coexist in the same diamond with minerals of the other suite. The discovery of this phenomenon (Meyer and Boyd, 1972; Prinz et al., 1975) was the first proof that diamonds grew in more than one geochemical environment.

It should be noted that inclusions can also be subdivided into protogenetic, syngenetic, and epigenetic. For purposes of this discussion only proto- and syngenetic inclusions are considered. Many syngenetic inclusions, particularly olivine, display a cubo-octahedral morphology that is imposed by the diamond host. In spite of diligent search by scientists through several tens of thousands of diamonds no macroscopic or microscopic fluid or gaseous inclusions have been found (Roedder, 1982; 1984).

The majority of mineral inclusions are small ($\sim 100 \mu\text{m}$) and monomineralic, although bi- and polymineralic inclusions do occur. Multiphase inclusions are important because the chemical and physical information they provide enables estimation of the pressure and temperature of equilibration of the inclusions, and by inference that of the host during diamond genesis. Estimated conditions of equilibration for co-existing ultramafic suite inclusions in diamond are between 900 and 1300°C and 45 to 65 kbar (Prinz et al., 1975; Meyer and Tsai, 1976b; Hervig et al., 1980; Boyd and Finnerty, 1980). These values lie within the region of diamond stability (Kennedy and Kennedy, 1976) and are similar to those obtained for diamond-bearing and diamond-free garnet lherzolite xenoliths (Shee et al., 1982). It is not possible to determine unequivocally pressures of equilibration for eclogite suite inclusions, but temperatures lie within the range calculated for the ultramafic suite.

A major unsolved problem at present is the direct determination of the age of diamond from the diamond itself. Currently, it is necessary to obtain the age of syngenetic mineral inclusions and to assume this approximates the diamond age. Kramers (1979) examined batches of inclusion-bearing diamonds from kimberlites in South Africa, and using lead isotopic techniques demonstrated that in general the inclusions, and by inference the diamonds, were much older than the kimberlite eruption. In the case of the Finsch and Kimberley kimberlites, the age of the diamonds is greater than 2 b.y., whereas the kimberlites erupted about 90 m.y. ago. More recent work by Ozima et al. (1983) suggests that some diamonds may have ages comparable to the age of the earth.

Most ultramafic and eclogitic suite inclusions have

Table 3. Minerals occurring as inclusions in diamond

PROTOGENETIC AND SYNGENETIC	
Ultramafic Suite	Eclogitic Suite
Olivine	Omphacite
Enstatite	Pyrope-almandine
Diopside	Kyanite
Cr-pyrope	Coesite (Quartz)
Phlogopite	Rutile
Cr-spinel	Ruby
Mg-ilmenite	Ilmenite
Zircon	Chromite
Sulfides	Sulfides
Diamond	Diamond
EPIGENETIC	
Serpentine, Goethite, Graphite, Kaolinite Hematite, Sellaite, Xenotime	
UNCERTAIN PARAGENESIS	
Muscovite, Biotite, Sanidine, Magnetite, Amphibole	

chemistries that differ in detail from similar minerals that occur in kimberlite. This is not to say that one cannot find in kimberlite rare examples of minerals that are chemically equivalent to those occurring as inclusions (Gurney and Switzer, 1973) but these minerals are most probably xenocrysts unrelated to kimberlite crystallization.

Kimberlite and lamproite

Kimberlites are widely distributed throughout the continental regions of the world (Bardet, 1974, 1977; Wilson, 1982). Most are not diamond-bearing, and compared to the diamondiferous ones have been little studied. Diamondiferous kimberlites appear to be confined mainly to the interiors of stable cratons, such as Southern Africa and Siberia, although some do occur close to continental margins in Liberia and Sierra Leone (Bardet, 1974; Haggerty, 1982). Dawson (1980) has provided a review of kimberlites and their xenoliths, and recently Clement et al. (1984) have proposed a redefinition and classification of kimberlite (see also Skinner and Clement, 1979).

Kimberlite occurs in diatremes, dikes and rarely as sills; multiple intrusions within a single diatreme are common. Neighboring diatremes, or pipes, may contain similar or very different suites of xenoliths, or none at all. Some kimberlites contain almost entirely xenoliths of eclogite, for example the Roberts Victor kimberlite, whereas others, such as the Kimberley pipes, are rich in lherzolitic xenoliths. Multiple intrusions in a single pipe often contain their own suite of xenoliths, or xenocrysts and more importantly diamond. Type I and II diamonds can occur in the same pipe, and diamonds with ultramafic inclusions coexist with ones that contain eclogitic suite minerals. In spite of the economic importance of diamond, it is a trace mineral in kimberlite and ranges only up to 1 part in 2.5 million; a quoted figure is 1 in 20 million (e.g., Kennedy and Nordlie, 1968).

Confusion has arisen due to the discovery of diamond-bearing rocks in north-western Australia that unfortunately were referred to initially as kimberlite (Jacques et al., 1982; McCulloch et al., 1983). The so-called "kimberlites" in that

region are tuffaceous lamproites (i.e., Ellendale, Argyle)—some of which are rich in diamonds (Jacques et al., 1984).

Lamproites are ultrapotassic rocks that may be plutonic, hypabyssal, or volcanic. In the latter instance both lavas, tuffs and vent breccias may be present. Generally the various rocks occur in groups, or fields, but worldwide they are relatively restricted (e.g., Leucite Hills, Wyoming; West Kimberley, N.W. Australia). Mineralogically leucite (or sanidine) and Ti-rich minerals are almost ubiquitous. Ti-amphibole, Ti-phlogopite, priderite, etc. are usually present and depending upon the abundance and type of minerals several rock names are possible (e.g., orendite, wolgidite, wyomingite, fitzroyite, etc.—Hughes, 1972, p. 321; Mitchell, 1984).

The significance of the diamond-bearing lamproites is that kimberlite is now not the only primary crustal source of diamond. It has been reported that some non-kimberlitic rocks in the Soviet Union contain diamonds (Kaminsky and Gevorkin, 1976; Kaminsky, 1980; Dawson, 1980). In several instances re-evaluation of what were once considered "odd" kimberlites has shown them to be lamproitic. The diamond-bearing Prairie Creek diatreme in Arkansas is one such example, and is now referred to as lamproite (Scott Smith and Skinner, 1982, 1984), and most likely the metakimberlites (Bardet, 1974) of the Ivory Coast (Knopf, 1970) and Gabon are lamproites.

The question of whether or not diamonds from lamproites have different inclusions than those from kimberlite has been answered in part from evidence at Prairie Creek, Arkansas. Diamonds from the Prairie Creek "lamproite" contain similar inclusions to those in diamonds from kimberlite—namely, olivine, enstatite, diopside, Cr-pyrope, pyrope-almandine, chromite and sulfides (Newton et al., 1977; Pantaleo et al., 1979). It is anticipated that similar mineral inclusions will be present in diamonds from the lamproites in N.W. Australia.

Discussion

Relation of diamond to kimberlite and lamproite

The presence of diamonds in both kimberlite and lamproite, the presence of similar inclusions in diamonds from kimberlite and lamproite, the chemical differences between these inclusions and cognate minerals in the host kimberlite and lamproite, plus the large disparity between the ages of diamond and kimberlite intrusions lead to the conclusion that diamond is not genetically related to either kimberlite or lamproite. The relationship of these two rock types to diamond is that they are the transporting medium by which diamond ascends to the earth's surface. Diamond is best described as a xenocryst in kimberlite and lamproite.

Transport of diamond

The process of kimberlite ascent through the mantle is unknown although several authors have suggested various models which include zone refining (Harris and Middlemost, 1969), diapirism (Green and Guegen, 1974; Wyllie,

1980; Anderson, 1982; Allegre, 1982) and conduit formation (Mercier, 1979). McGetchin and Ullrich (1973) calculated a speed of ascent for kimberlite of approximately 70 km/hr on the basis of xenolith size and density. A similar value was computed by Mercier (1979) from olivine annealing data. Both these rates of ascent are consistent with the cooling rates calculated by McCallister et al. (1979) for exsolution phenomena in pyroxenes from kimberlite. The evidence at present thus favors a rapid ascent, possibly hours, for kimberlite (and diamond) from depths in the upper mantle. Although geochemical processes in a diapir may have contributed to the formation of kimberlite, the route through the upper mantle was probably by crack propagation (D. H. Eggler, pers. comm., 1984).

It can be argued that the speed of ascent has served to preserve diamonds that would have been absorbed into the kimberlite magma with slow ascent. Other factors affecting the stability of diamond would be temperature, particularly the length of residence time at high temperatures and pressures outside the diamond stability field, and oxygen fugacity. Preliminary experiments (Meyer, unpub.) on the effect of temperature and f_{O_2} on the oxidation rate of diamond at room pressure have provided interesting data. At an f_{O_2} equivalent to 10^{-12} atm. and a temperature of 1000°C, roughly between the FMQ and MW (fayalite-magnetite-quartz, magnetite-wustite) buffers, diamond would disappear in about 21 days. In contrast under the same f_{O_2} but at 800°C diamond would remain for about 110 years. At present the f_{O_2} -temperature conditions for generation of kimberlitic, or lamproitic, magmas at depth are unknown. Arculus et al. (1982), Haggerty and Tompkins (1983) and Eggler (1983) suggest conditions in the region of the FMQ-MW buffers.

Mantle rocks and diamond

A large number of eclogite xenoliths that contain diamond are known (Hatton and Gurney, 1979) but only a small number of diamondiferous ultramafic xenoliths have been found. In contrast, the majority of diamonds studied contain ultramafic suite inclusions (Boyd and Finnerty, 1980).

Diamonds that contain eclogitic suite inclusions, and diamonds that occur in eclogite are obviously easily assigned to an eclogite source rock. Sobolev et al. (1972) documented this relationship by examining inclusions in diamond that itself was contained in an eclogite. The inclusions and the constituent minerals of the eclogite (clinopyroxene and garnet) were chemically equivalent, suggesting that the diamond is a constituent of the eclogite and thus formed contemporaneously with the eclogite host.

In contrast, it is less easy to determine the source rock in which diamonds with ultramafic suite inclusions formed. One reason for this is that few rocks are known that have mineral phases of comparable chemistry to the inclusions. Furthermore, the ultramafic xenoliths that do contain diamond are chemically and mineralogically varied and include dunites (garnet + olivine—Pokhilenko et al., 1977), harzburgites (garnet + olivine + orthopyroxene—

Sobolev, 1974; McCallum and Eggler, 1976), and lherzolites (garnet + olivine + clinopyroxene + orthopyroxene—Dawson and Smith, 1975; Shee et al., 1982). Diamond-bearing dunites consist of olivine and garnet whose chemistries are somewhat similar to those of the ultramafic suite inclusions from Siberian diamonds (Pokhilenko et al., 1977). Thus some Siberian diamonds may have been derived from the disaggregation of dunitic rocks, but such dunites have not been recognized elsewhere in the world. In contrast Shee et al. (1982), with specific reference to the Finsch kimberlite, South Africa, report no similarity in chemistry between the constituent minerals of a diamond-bearing garnet lherzolite and inclusions in loose diamonds from the same kimberlite.

Any model of diamond genesis for the ultramafic suite diamonds also must explain the chemical variation, on a worldwide basis, of some of the mineral inclusion types. For example, clinopyroxene inclusions show a range in Ca/(Ca + Mg) values from sub-calcic (<40 Ca/(Ca + Mg)) to normal diopsides, while some are Cr-rich (Sobolev et al., 1975). It is unlikely that the sub-calcic diopside inclusions would have formed in the same environment as the Cr-rich ones, and thus it seems probable that diamond forms in more than one rock type of ultramafic character.

The aforementioned differences in chemistry and mineralogy of the ultramafic xenoliths and inclusions, as well as the variety of xenolith types (dunite, harzburgite, lherzolite, etc.) that contain diamond are also paralleled by the mineralogical variety of eclogite xenoliths. Diamonds with coesite inclusions (Milledge, 1961; Sobolev et al., 1976) and coesite eclogite (Smyth and Hatton, 1977) most likely have formed under conditions chemically distinct from those of ruby-bearing diamond (Meyer and Gubelin, 1981) or corundum eclogite.

Further evidence in support of diamond having grown in several rock types within the broad classification of eclogite and ultramafic rock is the variation of $\delta^{13}\text{C}$ values, noted earlier, for eclogitic and ultramafic suite diamonds. The view that diamond is a constituent of several mantle rocks has also been expressed by Sobolev (1974).

Pressures and temperatures of diamond growth

The proto- and syngenetic inclusions in diamond are all phases that are stable at high pressures and temperatures, and are compatible with formation in the stability field for diamond. Estimates of equilibration conditions for diamonds are based on the chemistries of the inclusions and utilize the various geothermometers and barometers that are also used in determining pressure and temperature regimes for xenoliths (e.g., Boyd, 1973; MacGregor, 1974; Lindsley and Dixon, 1975; Wells, 1977; O'Neil and Wood, 1979). Ideally for thermobarometric calculations three coexisting and touching inclusions should be present in the diamond. This is seldom the case and accordingly some assumptions have to be made. In spite of probable errors in the use of these geothermobarometers, the equilibration temperatures and pressures obtained for diamonds are

within the same general range (900 to 1300°C; 45 to 65 kbar) as those obtained for xenoliths and megacrysts, including those which contain diamond. Thus it appears that diamonds with inclusions have equilibrated in the mantle at depths between 140 and 200 km; within the bounds of the asthenosphere and roughly coinciding with the thermal maximum suggested by Anderson (1980) to occur in the temperature profile of the upper mantle.

Crystallization of diamond

The manner in which diamond grows is a subject of controversy. It has been suggested that diamond forms in a solid state i.e., metamorphic reaction (Meyer and Boyd, 1969; Boyd and Finnerty, 1980), or is the product of some form of metasomatism (Shee et al., 1982) or is of igneous origin and crystallized from a magma (Meyer and Boyd, 1972; Harte et al., 1980; Meyer, 1982a,b).

The evidence is conflicting. The small size of the inclusions plus the fact that most are monomineralic led Meyer and Boyd (1972) to suggest that diamonds crystallized from a melt. It is possible that the chemical dissimilarity (particularly with regard to the ultramafic suite) of inclusions in diamonds and minerals from the host xenoliths is a result of the inclusions having formed early on the liquidus and having been removed from further chemical reactions with the liquid by the armoring effect of the diamond. Similar views have been expressed by Fesq et al. (1975) and Robinson (1978).

Although Gurney et al. (1979) and Harte et al. (1980) consider diamond to be an igneous phase they relate its genesis directly to the kimberlite magma. In view of the disparity between kimberlite ages and the age of diamond, growth from a kimberlite magma is untenable.

The presence of alternating stratigraphy of Type I and II diamond suggests discontinuous growth and minor chemical variation in the environment in which growth occurred. Clues as to whether the growth process was either igneous or metamorphic may be found through a detailed study of the relationship between mineral inclusions and the host diamond stratigraphy.

On the basis of various geothermometers and barometers, particularly those of O'Neil and Wood (1979), Boyd and Finnerty (1980) cautiously suggest that the majority of diamonds with ultramafic suite inclusions have formed in subsolidus events. The main reason for this conclusion is the fact that most inclusions in diamonds have equilibration temperatures below 1150°C.

Metasomatic processes in the mantle are currently widely discussed (Bailey, 1984; Boettcher et al., 1979; Wyllie, 1980) although recently Walker (1983) has cautioned, quite correctly, against solving all geochemical problems of the mantle by invoking the "deus ex machina" of metasomatism. Metasomatizing agents relevant to diamond formation are various combinations of C, H and O.

Experimental studies of $\text{CO}_2\text{-H}_2\text{O}$ -peridotite systems (Eggler, 1977; 1978; Wyllie, 1977, 1978; Eggler and Wendlandt, 1979; Ellis and Wyllie, 1980) have provided significant data and models for the genesis and eruption of kim-

berlite magma (e.g., Wyllie, 1980). However, the formation of kimberlite with or without CO₂ has little to do with the much earlier genesis of diamond. Furthermore, if most diamonds are Archaean or Proterozoic, it could be questioned as to whether present thermal models of the mantle (Sleep, 1979; Anderson, 1980) are valid for discussions of the genesis of diamond.

Carbon in a melt is unlikely to be in elemental form (Olafsson and Eggler, 1983) but rather as some dissolved species such as CO₂ or CH₄. Whether CO₂ or CH₄ is present in the melt depends on the redox conditions. Studies of ilmenite (Haggerty and Tompkins, 1983; Arculus et al., 1982) and megacrysts (Eggler, 1983) suggest *f*_{O₂} values in the mantle to range from fayalite–magnetite–quartz to magnetite–wustite. These conditions possibly do not reflect those of the early mantle which may have been more reduced (Haggerty and Tompkins, 1982). Sobolev et al. (1981) have reported metallic Fe inclusions in Siberian diamonds. In the event of a more reduced mantle, perhaps equivalent to iron–wustite buffer, the predominant dissolved species in a magma would be CH₄ (Woerman and Rosenhauer, 1982; Eggler and Baker, 1982; Eggler et al., 1980; Ryabchikov et al., 1981). The occurrence of CH₄ in the mantle and its role in diamond formation has been the subject of modelling by Deines (1980).

It has been hypothesized (Gold, 1979; Gold and Soter, 1979) that the deep mantle is being degassed of methane. Melton and coworkers (1973, 1974) reported the presence of CO₂ and CH₄ among gases that were evolved from diamonds they crushed between steel anvils in a mass spectrometer. They found indications that other organic complexes of C and H were present, but their results require independent substantiation. As noted earlier, no macroscopic fluid or gaseous inclusions have been observed in diamond, although CO₂ has been recorded in fluid inclusions in olivines from xenoliths in kimberlite (Roedder, 1963, 1982, 1984).

In summary the evidence generally favors the formation of diamond either directly from an igneous melt or from some type of metasomatising fluid that has pervaded various types of mantle rock, perhaps at different times. It is not clear whether the carbon was derived from CO₂ or CH₄ but the presence of CH₄ in diamond, metallic iron as inclusions, and Cr²⁺ in olivines included in diamond indicate reduced conditions which would favor CH₄.

Source of mantle carbon

The source of the carbon that forms diamond is an enigma. Variations in δ¹³C values for diamonds may reflect either an isotopically inhomogeneous mantle or the presence of recycled crustal material, or both. A similar comment is valid for the ³He/⁴He and ⁴⁰Ar/³⁶Ar values, although more data are obviously needed in order to remove ambiguities. Meteorites, particularly the carbonaceous variety, have a range of δ¹³C values that is similar to that for diamond (Robert and Epstein, 1982).

In addition to continual degassing of the mantle in CH₄ and CO₂ as suggested by Gold (1979), the intriguing and

speculative model of Dickey et al. (1983) provides a ready source of carbon. In this model elemental carbon in the lower mantle is in the form of a metallic liquid. Subsequent perturbations cause upward migration of the carbon resulting in the formation of mantle plumes. Oxidation of the carbon produces CO₂ which causes partial melting and carbonation of the mantle silicate rocks. Theoretically, if elemental carbon ascended from the lower mantle it should pass through the hexagonal diamond phase—lonsdaleite (Bundy and Kasper, 1967; Frondel and Marvin, 1967) before inverting to cubic diamond at lower pressures. However, the absence of lower mantle phases as mineral inclusions suggests it is unlikely that the carbon of Dickey et al. (1983) directly forms diamond.

Recycled crustal carbon in the form of carbonate in the mantle may be present within regions of subduction. It is stable to appropriate depths (Wyllie, 1978, 1980) and under the correct conditions diamond should be produced. The influence of such carbon is unknown, but generation of some diamonds from recycled carbon (say eclogitic types), and other diamond from primitive carbon (mostly ultramafic types) would account for various chemical and isotopic features. If this is the case and if eclogitic diamonds are also very old (i.e. Proterozoic or older) then a corollary is that subduction must have been operative during the early history of the earth.

Summary

Various partial melting or metasomatic events in which dissolved CH₄ or CO₂ were present, resulted in crystallization of diamond at depths of approximately 180 km in the upper mantle. Isotopic data, especially that of carbon, are compatible with the possibility that diamonds formed from carbon whose sources were isotopically different. Variation in isotope content could have been produced by inhomogeneity in primitive mantle material, the presence of recycled crustal carbon, or both. The growth of diamond was not unique to a single rock but took place in any mantle material in which chemical interactions produced elemental carbon. Consequently several types of rock broadly grouped under eclogite and ultramafic (peridotite) are the original cognate hosts of diamond. Evidence suggests that this crystallization of diamond occurred in the Archaean or Proterozoic. Subsequent mobilization of mantle diapirs and associated physical and thermal perturbation of the proximal mantle rocks resulted in the incorporation of diamond and xenoliths. Depending upon the location of the diapirism relative to the lithosphere, various magmatic end products could have been produced. Kimberlite or lamproite magmas, formed as a consequence of diapiric processes, conveyed diamond rapidly to the crust. Rapid ascent was possibly through crack propagation in the mantle, and through a thick cool lithosphere whose thermal regime resulted in little loss of diamond.

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Note added in press:

After the present manuscript was written and accepted for publication, Richardson et al. (Science, 310, 198–202, 1984) have presented isotopic evidence that chrome-pyrope garnet inclusions (ultramafic suite inclusions) in diamonds have model ages of 3.2 to 3.5 b.y., whereas the host South Africa kimberlites were intruded roughly 90 m.y. B.P.

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