DIAMOND, NATURAL

1. Introduction

Naturally occurring diamond is a relatively rare polymorph of carbon characterized by a three-dimensional (3D) arrangement of tetrahedrally coordinated carbon atoms. On the Earth's surface, diamonds occur in several major kinds of deposits: primary and secondary (both alluvial and littoral). In primary deposits, they are enclosed in host rocks of kimberlite or lamproite that form "pipes" that are downward-tapering, cone-shaped structures of igneous (volcanic) origin. Subsequent erosion of these pipes and fluvial transport of the diamonds lead to the formation of alluvial deposits in river beds and river terraces. The final resting place of diamonds is in littoral or ocean floor deposits. Only a small number of

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pipes, alluvial, or littoral deposits contain a high enough concentration of diamonds to warrant mining. Separation of diamonds from their enclosing or associated rocks include crushing, screening and sieving, use of grease belts, suspension in heavy or dense media, and sorters that use the luminescence of diamonds when they are exposed to X-rays. Diamond crystals are sorted into many categories depending on size, color, clarity, and shape for valuation purposes. The classification of diamonds into four types (Ia, Ib, IIa, IIb) is based upon the presence or absence of nitrogen and boron distributed in the lattice. Both natural and synthesized diamond [7782-40-3] have the highest hardness of all known materials, the highest thermal conductivity at room temperature, a high refractive index and optical dispersion, a low thermal expansion, and a relatively high inertness to chemical attack. This unique combination of properties permits diamond to be foremost in certain applications: as a highly prized gemstone; industrially as an important abrasive unsurpassed in certain cutting, drilling, sawing, machining, grinding, and polishing operations for many materials; and in electronic and optical applications as a heat sink and window material, respectively.

2. Occurrence and Exploration

Diamonds were first discovered in ancient times in India and Borneo, and later in Brazil in the early 1700s, in alluvial deposits where flowing water had sorted minerals on the basis of density and toughness. These alluvial concentrations of diamonds are derived from primary source rocks by erosion and fluvial transport, where the tumbling action in the transported sediments often concentrates the better quality diamond crystals by preferentially destroying flawed ones. These processes have occurred over long periods of geologic time in all rivers draining primary sources, and particularly good quality alluvial diamonds are found in rivers in Angola, Brazil, India, and in areas of southern and western Africa. The Orange river system in South Africa has transported diamonds from inland primary sources to the Atlantic Ocean, where ocean currents have distributed the diamonds along both the littoral zone and the ocean floor off of South Africa and Namibia. Exploration of alluvial deposits can be done by panning of stream sediments, or by drilling, pitting, and trenching of streambed and terrace deposits, in conjunction with a search for the heavy mineral assemblages that accompany diamond. Recovery of diamonds from the ocean floor requires the use of sophisticated underwater equipment to collect the diamonds and return them to a processing ship. Alluvial deposits account for $\sim$20% by weight (but 41% by value) of the world's annual production of diamonds.

Upstream exploration has sometimes led to the discovery of the primary sources of alluvial diamonds, namely, kimberlite or lamproite pipes that occur in cratons (portions of the Earth's crust that have been geologically stable for very long periods of time) (1,2). These pipes are the principal sources of natural diamonds, and there are >6000 occurrences of them in the world (2). Only $\sim$60 of these occurrences (including dormant, active, and developing mines) contain a high enough concentration of diamonds to warrant commercial mining (3).

Even in successful mining operations, the diamond/gangue ratio that has to be removed and crushed is of the order of less than one part in a million (<1 ppm) (rarely >5 carats/tonne ($=1$ ppm), and often 1 carat/tonne ($=0.2$ ppm) or less), of which the proportion of gem quality crystals is $\sim 20\%$. The first key discovery of diamond-containing kimberlite pipes was in South Africa in 1871, and these structures are well known from the publicity given to the Kimberley, Premier, and other mines there. Beginning in the mid-1950s, a number of diamondiferous kimberlites were found in Yakutia (now the Sakha Republic) in the Siberian portion of the Russian Federation (more recently, kimberlites have been discovered in the Arkhangel'sk region on the eastern side of the White Sea). Additional successful discoveries were a producing pipe in Botswana in 1967 (the first of three commercial finds in that country), and the Argyle alluvial deposits and lamproite pipe in northwestern Australia in 1979. Discoveries in the Arctic parts of Canada in the 1990s marked the beginning of a new important and long-term economic source of diamonds. Additional major producing countries are Angola, Namibia, and the Democratic Republic of the Congo. There is a lamproite pipe in Arkansas in the United States where amateurs can dig with a reasonable probability of finding a diamond (4). Exploration methods to prospect for potentially commercial primary deposits include the tracing of certain heavy "indicator" minerals upstream, core drilling to sample the subsurface geology, the use of aerial photographs to try to find manifestations of circular pipe structures at the surface, and magnetic anomalies or other geophysical measurements by aerial or surface surveys.

Both the origins of kimberlite (or lamproite) and the enclosed diamonds are still a source of controversy and continued geological study (2,5–10). The ages of some diamonds have been measured on the basis of certain radioactive elements in mineral inclusions that date these inclusions to be >3 billion years old. This is in great contrast to the age of the volcanic-like upward intrusions of the host kimberlite or lamproite magmas that took place mainly between ~ 80 and 600 million years ago (there are a few intrusions with ages of ~ 1.2 billion years). Moreover, all commercial primary deposits occur in cratons, portions of the crust that are older than 1.6 billion years (2,10). It seems somewhat anomalous compared to other mineral deposits that in spite of the large size of these natural "pressure vessels", the largest diamond ever found, the Cullinan, weighed 3106 carats (~ 176 cm^3 or 620 g). Only a limited number of rough diamonds weighing >300 carats (60 g) have ever been found (11). The fourth largest rough diamond (890 carats) emerged in the market as a polished gemstone of 407.48 carats (12). The growth history of each diamond crystal is complex, often involving intermittent periods of growth and dissolution as revealed by studies of the internal structure (13–15). There may also have been a problem in maintaining a supply of carbon to the growing crystal over long periods of time, and dissolution of material during the strenuous trip upward from the stability region of diamond at depth likely contributed to decreasing the size of some crystals. Overpressure is known to increase the nucleation frequency in synthesized diamond, and this might translate to many small crystals at great depths. It is quite certain that nature did not use a metal–carbon solution system as in the commercial process for the growth of diamond at high pressures and temperatures, but the pressure–temperature ranges for both processes may have been comparable, $>1000^\circ$C and 4.5 GPa (45 kbar) (7,8,10). This translates to depths of $\sim$150–200 km into the Earth.

Diamonds, usually in the size of microcrystals <0.5 mm in diameter, have also been found in non-kimberlitic and non-lamproitic host rocks. An extensive deposit of very small diamonds (<0.1 mm in size) occurs in certain metamorphic rocks in the Kokchetav region in northern Kazakhstan; these are thought to have formed when crustal rocks were subducted to surprising depths of 120 km (16). Diamonds also occur in meteorites as well as meteorite impact structures (astroblemes), probably as a result of high pressures produced dynamically by impact (17,18). The shock or explosive mode of synthesis is a viable process for producing fine diamond powders of both the cubic and hexagonal (lonsdaleite) polymorphs (19) naturally or otherwise. Some diamonds in space appear to have formed in supernovas by processes more closely related to the low pressure chemical vapor deposition processes (see CARBONS, DIAMOND, SYNTHETIC) (20).

3. Recovery

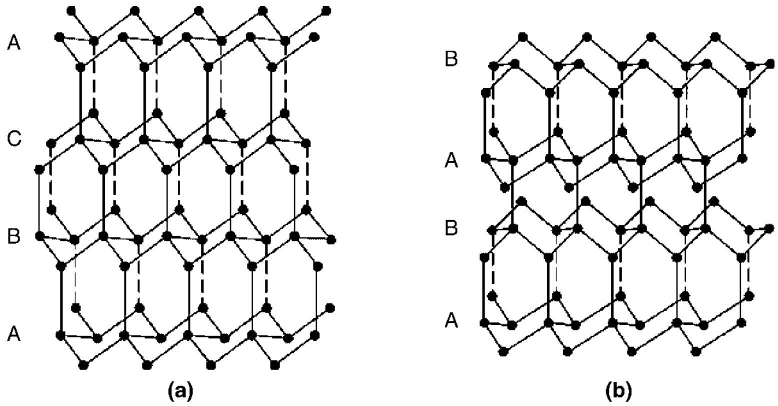
Alluvial diamonds are recovered from gravel-rich layers of sediments deposited by streams in terraces or along channels. Often the removal of the contents from depressions, potholes, or “pockets” in the streambed where fluvial concentrations are very great has produced spectacularly large amounts of diamonds. High quality diamonds (95% gemstones) are recovered from ocean deposits in the littoral zone off the western coast of southern Africa. This is accomplished by holding back the water with seawalls long enough to remove a considerable overburden of sand to reach the gravel-rich layers and underlying furrowed bedrock. The furrows in the bedrock have to be cleaned out completely as these are the sites where the highest concentrations of diamonds occur. Underwater recovery of diamonds from both the littoral zone and the ocean floor in this same region requires the use of sophisticated mining ships that anchor over productive areas. A remote-controlled underwater vehicle is lowered to the bottom, which suctions material back up to the ship for processing to separate the diamonds from rocks, sand and other debris (1).

The original mining of the kimberlite pipes begins at the surface in the softer weathered “yellow ground”, but most of the subsequent recovery is by removal from depth of the harder, unweathered “blue” kimberlite that is accomplished by open-pit and usually followed by underground mining methods. The removed rock is first crushed, screened, and sieved into several size fractions. Recovery of diamond from its gangue is carried out by a variety of techniques depending on the size of the deposit and physical properties of the host rock. One of the more interesting sorting methods is the grease table or belt that takes advantage of the fact that most diamonds (but not those from littoral deposits) will stick to grease but not to water that washes away the gangue. Non-stick diamonds can be recovered by electrostatic methods. The method most often used to concentrate heavy minerals (including diamonds) from the crushed rock is by suspension in a heavy or a dense medium [hence the acronyms HMS (heavy media separation) and DMS (dense media separation) plants at the major diamond mines]. The more modern method for the separation of diamonds from the heavy mineral concentrate is by an X-ray sorter that uses the emission of light by luminescence of diamond when exposed to X-rays. The concentrate fragments are passed on a vibrating conveyor belt (the vibration ensures the mineral grains are not lying on top of each other, but form a layer that is one grain thick) beneath an X-ray source. The luminescing diamonds are detected by an optical sensor, and are removed from the moving belt by a finely directed blast of air. Final sorting is done by hand to separate the diamonds into many categories according to weight, color, clarity, and crystal shape. Recently installed separation processes at very large mines, such as Orapa and Jwaneng in Botswana, are fully automated, and do not involve any sorting by hand. At a large mining operation, thousands of tons of ore are processed per day (1).

4. Properties

Not all of the properties of diamond are considered here, and additional information and qualifications are available (13–15,21–27).

The lattice constant of the unit cell of the face-centered cubic (fcc) form of diamond is 0.3567 nm with a density of 3.52 g/cm^3. For the hexagonal form lonsdaleite, $a = 0.252$ nm and $c = 0.412$ nm with the same density as the cubic form (Fig. 1). In this allotropic form, the stacking of successive layers of carbon atoms is ABABAB ... as compared to the ABCABC ... type of stacking of the normal cubic form. Diamond twins readily during growth on the (111) plane resulting in both interpenetration and flat twins called macles (other more complex twin relationships are also possible) (14). The growth morphology of natural diamond is typically octahedral, but dodecahedral crystals are common, perhaps because of subsequent solution after growth. Many crystals exhibit evidence of uneven development of crystal faces leading to less regular shapes, and of modification by etching and dissolution to produce rounded surfaces and edges. A few diamond crystals exhibit the approximate form of a cube, but the nonplanar irregular surfaces are the result of curved, noncrystallographic forms of growth.



(a) (b)

Fig. 1. Crystal structures of (**a**) cubic diamond and (**b**) hexagonal lonsdaleite. A, B, and C indicate the stacking sequence of sheets of carbon atoms.

"Coated" diamonds are dull and opaque due to the presence of an outer layer (formed by a fibrous, columnar growth) that contains a high level of impurities (14). Only very small grains (<30 μm) of the hexagonal lonsdaleite have ever been seen, so not much can be said of their morphology.

Besides the single crystals usually implied in a discussion of diamond, it also occurs in nature in the form of polycrystalline aggregates (14). These can be classified into two types based on origin: carbonado and ballas. The former is a dark-colored aggregate of previously formed, submicron-sized grains that have become cemented to each other to varying degrees of bonding. Some varieties such as framesite show evidence of mechanical deformation during the aggregation process. Ballas refers to a grey-colored polycrystalline aggregate of diamond grains that grew simultaneously as opposed to an assemblage of previously grown crystals. Specifically the term refers to a ball-shaped polycrystalline diamond with a radial structure (like some hailstones). Contrary to carbonado, ballas is free of secondary phases. Most synthesized compacts have a structure that resembles carbonado rather than ballas. Bort is a general term used to describe low grade industrial diamond suitable only for use in a fragmented form.

Diamond is nominally pure carbon with a $^{12}C/^{13}C$ ratio of $\sim$99:1. Although other elements are reported in chemical analyses, most are considered to be present in oxide, silicate, and sulfide mineral phases as inclusions in the diamond. Only boron and nitrogen are considered to be truly substitutional in the lattice. Hydrogen and oxygen, possibly as OH, may also be important structural contaminants though they may also be present as second-phase gases along with CO, CO_2, H_2O, CH_4, and other species (28). These impurity elements can be present in concentrations from $\sim$1 to $>$1000 ppm; a variety of additional elements can occur at even lower concentrations (14,15).

The classification of diamond into four principal types is based on the presence of nitrogen (type Ia and Ib) and boron (type IIb), and the effects of these impurities on the infrared (ir) and ultraviolet (uv) transmission (14,15,22–27,29). Nominally pure diamond without detectable amounts of these impurities is referred to as type IIa. It has the highest thermal conductivity and is transparent out to 10.6 μm. The concentration of boron in natural type IIb diamonds is $\sim$0.25 ppm and as much as 270 ppm in synthesized crystals; a crystal with $\sim$10 ppm boron is deep blue. The most famous natural type IIb diamond is the blue 45.52-carat Hope diamond. Dissolved nitrogen can range up to 2500 ppm in type Ia natural diamonds and to $\sim$500 ppm for synthesized type Ib. Most natural diamonds are type Ia, which is characterized by the presence of platelets, made visible by transmission electron microscopy on the (100) planes. The exact nature of these platelets is still controversial, but they are often called nitrogen platelets (possibly N−N or C−N clusters). An increase in the high temperature bending strength of type Ia over type IIa diamonds has been attributed to the platelets interfering with dislocation motion (30). When the nitrogen is in this aggregated atomic form, the crystals can be essentially colorless to light yellow, in contrast to the darker yellow type Ib, which has nitrogen as isolated atoms in substitutional positions in the lattice. Most synthesized diamond is type Ib unless special precautions are taken to remove nitrogen from the growth environment. A type Ib diamond can be converted to a type Ia and vice versa by annealing at high pressures and temperatures (31,32). This was first established in the laboratory and is now presumed to be the way nature made type Ia diamonds. If this conversion could be carried out in a practical manner, it would be of some interest in improving the value of slightly yellow gemstones (to date, there is no evidence that such treated type Ia diamonds exist in the jewelry marketplace). The latest development in the jewelry trade is treatment of brown type IIa diamonds to lighten or remove their brown color to make them colorless by annealing at very high temperatures and pressures (33).

The main inclusions in natural diamond are pyroxene, garnet, spinel and olivine, and the understanding of the conditions of their formation from laboratory studies is the basis for the determination of the $P-T$ conditions where diamond was formed (5–9). The compounds CO, CO_2, H_2, H_2O are also found in diamond (28), and it is possible that diamond nucleated and grew in a C–H–O system in equilibrium with the silicate matrix (7). Graphite [7440-44-0] is also a common inclusion in natural diamond. Additional minerals identified as inclusions include oxides (chromite and rutile), sulfides (pyrrhotite), and minerals such as zircon, coesite kyanite, and metallic iron.

Diamond is thermodynamically unstable at ambient conditions, but unless heated to 650°C in air (oxidation to CO_2) or 1700°C in vacuum or an inert atmosphere (graphitization), it will remain as diamond indefinitely. To convert graphite directly to diamond requires very high pressures, ~12 GPa (120 kbar) and temperatures ~1800°C (see Fig. 1 in CARBON, DIAMOND SYNTHETIC). For synthesis of industrial diamond these conditions can be lowered by the presence of metal solvent/catalysts. More recent work on the thermodynamically stable region for diamond indicates a positive slope for the melting curve instead of the negative slope deduced previously on the basis of analogy to the $P-T$ diagrams for Si and Ge (34). This has important implications for the origin and extent of diamond in the earth. A "... diamond as big as the Ritz ..." may indeed exist at great depths in the Earth (35).

Diamond is chemically inert to inorganic acids, but can be etched in oxidizing molten salts such as KNO_3 at 600°C. Carbon as diamond or graphite is soluble in several metals, particularly Fe, Ni, Co, and other Groups 8–10 (VIII) elements. This imposes a serious limitation on the use of diamond for machining alloys based on these materials in spite of the high hardness of diamond. At the temperatures developed at a cutting tool tip, diamond reacts with these metals. However, similar reactions facilitate the bonding of diamond to metal in diamond tool making because of the wetting of diamond by carbide-forming metals such as W, Ta, Ti and Zr (14,15,36,37).

Diamond is the hardest material known because of its combination of a three-dimensional (3 D) arrangement of tetrahedrally coordinated C–C bonds with a bond distance of 0.154 nm. The Knoop hardness (K) is in the range of 68.7–98.1 kN/m^2 (7000–10,000 kgf/mm^2). Harder materials have been postulated on the basis of bond distance, but so far have not been found or made in a useful form (38,39). Diamond is twice as hard as cubic BN ($K = \sim 44$ kN/m^2), which in turn is twice as hard as SiC ($K = \sim 23.5$ kN/m^2). The hardness of diamond varies as a function of crystallographic orientation; this is important when mounting diamonds for drill bits or in polishing gemstones (14). The "table", the largest facet of a brilliant cut diamond, is generally the (001) face because it is easier to saw and polish than the (111) face using traditional techniques. The use of lasers to cut diamond diminishes the importance of orientation dependence of hardness in some industrial operations.

Although hard, diamond is also very brittle and cleaves readily on the (111) plane and also on other planes under certain conditions. The ability of diamond to fracture is useful in cutting and grinding applications because the crystals are self-sharpening. For other procedures such as sawing rocks or concrete, tougher diamonds are desirable (14). The uncontrolled fracture of a gemstone is disastrous, but in the cutting and polishing industry large diamonds are first reduced to desired sizes by controlled cleavage or laser cutting to avoid the time-consuming process of sawing (11,40–42).

Measurement of the mechanical properties of diamond is complicated, and references should be consulted for the various qualifications (11,14,43). Table 1 compares the theoretical and experimental bulk modulus of diamond to that for cubic BN and for SiC (38), and compares the compressive strength of diamond to that for cemented WC, and the values for the modulus of elasticity E to those for cemented WC and cubic BN.

The thermal conductivity value of 2000 W/(m · K) at room temperature for type IIa natural diamond is about five times that of Cu, and recent data on 99.9% isotopically pure ^{12}C type IIa synthesized crystals are in the range of 3300–3500 W/(m · K) (44). This property combined with the high electrical resistance makes diamond an attractive material for heat sinks for electronic devices. More impure forms of diamond (type Ia) have lower thermal conductivities [600–1000 W/(m · K)].

The averaged value of the coefficient of linear thermal expansion of diamond over the range 20–100°C is 1.34×10^{-6}cm/cm/°C and 3.14×10^{-6} from 20 to 800°C. At room temperature, the values for silica glass and diamond are 0.5×10^{-6} and 0.8×10^{-6}, respectively. The relatively low expansion combined with the low reactivity of diamonds, except for carbide formation, leads to some challenges in making strong bonds between diamond and other materials.

The high refractive index (2.42 at 589.3 nm) and dispersion (0.044) are the basis for the "brilliance" (white light return) and "fire" (flashes of colored light return) of a properly cut diamond gemstone. The optical transmission out to 10.6 μm for type IIa diamonds makes possible windows for CO_2 lasers and for devices such as were in the Venus interplanetary probe. References should be consulted for the details of the optical transparency of the different type diamonds (13–15,22,24–27). Pure diamond produces no absorption of visible light, but it exhibits considerable absorption in both the uv and ir regions.

Table 1. **Mechanical Properties of Diamond and Other Hard Materials, (GPa)**[a]

	Diamond	Cubic BN	WC[b]	SiC
bulk modulus	440	370		211
compressive strength	8.69		4–6	
modulus of elasticity, E	950–1100	890	460–675	

[a] To convert GPa to psi, multily by 145,000.
[b] Cemented.

Diamond displays some intrinsic absorption features in the ir spectrum between 1600 and 3000 cm^{-1}; the region between 800 and 1400 cm^{-1} contains features characteristic of the state of nitrogen aggregation and as such is a means of distinguishing the different diamond types. The direct band gap for diamond is 5.47 eV. The visible spectrum of diamond can display both sharp and broad absorption features resulting from a variety of optical centers (due to nitrogen or boron impurities, radiation damage, plastic deformation, or annealing). Natural diamond exhibits many colors (yellow, brown, blue, pink, and green), and color modification by irradiation and annealing, and by heating at very high pressures and high temperatues (HPHT), is used for diamonds for jewelry purposes (33,45). Though cubic, most natural diamonds show anomalous strain birefringence when viewed under crossed polaroids.

Diamond is an electrical insulator ($\sim 10^{16}$ Ω/cm) unless doped with boron when it becomes a *p*-type semiconductor with a resistivity in the range of 10^{-1}–100 Ω/cm. *n*-Type doping has been claimed but is less certainly established. The dielectric constant of diamond is 5.58.

5. Applications

The selected applications of diamond are based on its high hardness, wear resistance, ability to self-sharpen as it cleaves, thermal conductivity, and chemical inertness. Loose abrasive grain is used in lapping and polishing operations, eg, on the scaife used for facetting gem crystals and polishing rock materials for monuments and buildings. Single-point tools for engraving, turning , or cutting applications are made by mounting an individual diamond in metal holders (14,36,37,46). Other uses for single diamonds are as phonograph needles, bearings, surgical knives, and wire dies. The differential hardness of diamond is the cause of the nonuniform wear, or loss of roundness, of a wire die. If sintered (polycrystalline) diamond is used for wire dies, the anisotropic wear problem disappears.

For many applications such as grinding, cutting, drilling, and sawing, abrasive grains are mounted in resin, metallic, or vitreous bonding materials on wheels, disks, or saw blade sectors. Metallic bonding can be accomplished by electroplating as well. The grooving of concrete runways and roads is a well-known example of the use of impregnated diamond saw blades. An interesting new development is as beads, which are strung on long wires for sawing blocks of stone from quarries. About 90% of industrial diamond is synthesized.

Worldwide production in 2000 of natural diamonds amounted to 110 million carats (22,000 kg) worth ~\$7.5 billion, of which ~20 million carats (4000 kg) of gem quality were used in the jewelry industry. The wholesale value of polished diamond sales in that year was ~\$13 billion, and \$58 billion for retail jewelry sales (47).

Diamonds have been used as thermistors and radiation detectors, but inhomogeneities within the crystals have seriously limited these applications where diamond is an active device. This situation is rapidly changing with the availability of more perfect diamonds of controlled chemistry obtained by modern synthesis methods. The defect structure also affects thermal conductivity, but cost and size are more serious limitations on the use of diamond as a heat sink material for electronic devices.

Besides the gem qualities dependent on optical properties, diamond is very useful as a light-transmitting window for lasers, and for simple windows for monitoring chemical processes in corrosive and otherwise hostile environments.

From standards set by the gem trade, a small amount of yellow lowers the value of a diamond gemstone, but when the color is strong and uniformily distributed, the value increases in what is called the "fancy" class. Pink, blue, and green diamonds are very rare and highly prized. Most of the demand is for colorless diamonds, of which only a small percentage are totally free of flaws. The selection of rough diamonds and the cutting to maximize the final value is a mix of science and art based on experience. For the procedures for transforming a natural diamond to a gem, see (41). Besides the traditional cutting centers such as in Belgium and Israel, India has recently become a primary supplier of the smaller sizes of polished diamonds (48). The supply and price of diamonds for jewelry are rigidly controlled (40,49).

A significant impact on research at high pressure has come about with the use of gem quality diamonds as Bridgman-type anvils in a small compact high-pressure device (50–52) . With this type of apparatus, pressures greater than those at the center of the Earth (360 GPa = 3.6 Mbars) have been reached, and phase transformations of many materials have been studied. Because of the X-ray transparency of diamond, it is possible to determine the structure of the phases under pressure. Because of the strenuous environment, crystals selected for this application have to be of very high quality.

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