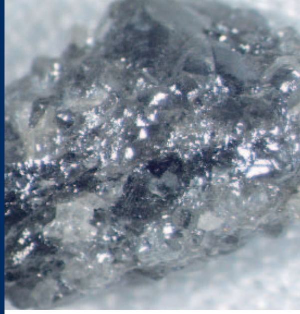

Strange Diamonds:
The Mysterious Origins of Carbonado and Framesite

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Polycrystalline aggregates of diamond called carbonado and framesite have excited the attention of scientists because their crystallization histories are thought to depart markedly from established modes of diamond genesis. In contrast to kimberlitic diamonds, the geochemical signatures of carbonados are systematically crustal. Since the apparent age of carbonados is Archean (~3.2 Ga), a number of exotic formation theories have been invoked, including metamorphism of the earliest subducted lithosphere, radioactive transformation of mantle hydrocarbon, and meteorite impact on concentrated biomass. Unlike carbonados, framesites are known to originate in the mantle. They appear to have crystallized very rapidly, shortly before the eruption of the kimberlites that brought them to Earth's surface, suggesting that old cratonic materials can be remobilized after long-term storage in the lithosphere.

KEYWORDS: carbonado, framesite, polycrystalline diamond

INTRODUCTION

Most people who like diamonds like them large. Nevertheless, a small cadre of geochemists and mineralogists has focused its efforts on diamondiferous grunge—the black multigranular masses of uncertain pedigree that fall below even the basest standards of the Gemological Institute of America grading scales. Generically called bort, these diamonds have earned some measure of respect not as gems but as the best abrasives for jobs that require superhardness and supertoughness (Feenstra 1985). Single-crystal diamonds cleave so easily that they wear rapidly during drilling applications, and inclusions can explode a single-diamond host when heated. In contrast, the high densities of grain boundaries in polycrystalline diamonds (PCD) impede fracture propagation, and the porosity in natural PCD allows for thermal expansion of included mineral grains without catastrophic failure of the entire compact.

The taxonomy for polycrystalline diamond tends to be based on qualitative external characteristics, as the names are of a cultural rather than scientific origin. Consequently, even the modern literature is inconsistent in its application of the terminology. In this review, "carbonado" refers specifically to multigranular diamond aggregates from the Central African Republic and Brazil with distinctive properties (summarized in TABLE 1 and FIG. 1). "Framesite" is used more broadly to describe clusters of randomly oriented microcrystalline diamonds found in association with kimberlites all over the world. The unusual physical features observed in carbonados and framesites have provoked a wide array of hypotheses regarding their origin. This article will focus on the chemical and structural properties that unite and divide these two puzzling diamond varieties.

CARBONADO

Mineralogical Characteristics

Carbonado nodules typically are pea-sized or greater. Indeed, the largest known diamond of any type is the Carbonado of Sergio (Brazil), weighing in at 3,167 carats. Named for the Portuguese word for "burned," carbonados are black polycrystalline masses. As with framesite, carbonado nodules often exhibit a "microporphyritic" texture in which euhedral crystals measuring hundreds of microns across are cemented by a matrix of micron-sized grains (Fettge and Sturges 1932). In contrast to framesites, however, X-ray microtomography reveals pervasive macroporosity with tunnel sizes in excess of 1 mm (Vicenzi et al. 2003).

Haggerty (1999) has described carbonados as "the most enigmatic of all diamonds," and the number of scenarios proposed for their formation would seem to support this claim. Unlike framesites, carbonados have never been found in direct association with kimberlite pipes, raising the possibility that the Earth's mantle was not their ultimate source. Although carbonados have been reported from Venezuela (Kerr et al. 1948) and various Russian localities (e.g., Gorshkov et al. 1996), carbonados sensu stricto are found in Mid-Proterozoic (1–1.5 Ga) metaconglomerates overlying the São Francisco and Congo-Kasai cratons in Brazil and the Central African Republic, respectively. The São Francisco craton contains the oldest rocks in South America, at least ~3.2 Ga (Martin et al. 1997; Magee 2001), and the great age of carbonado is part of its fascination. Both bulk and in situ analyses of radiogenic lead isotope compositions of carbonado diamond and its inclusions indicate that crystallization occurred between 2.6 and 3.8 Ga ago (Ozima and Tatsumoto 1997; Sano et al. 2002).

Carbonados are distinguished from other polycrystalline diamonds by the inclusions that line pore spaces (Trueb and Butterman 1969; Trueb and deWys 1969, 1971; Dismukes et al. 1988). The hydrated rare-earth phosphate florencite is the most abundant of these, but over 30 other

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TABLE 1

COMPARISON OF CARBONADO, FRAMESITE, AND ECLOGITIC SINGLE-CRYSTAL DIAMOND.

	Carbonado	Framesite	Single-Crystal Eclogitic Diamond
Color and Surface Properties	Black, dark gray, or brown with smooth exterior	Light gray or brown with irregular surface	Clear to yellow single crystals are most common
Grain Sizes	Euhedral grains (Typically up to 200 µm) set in microcrystalline matrix (<0.5–20 µm)	Shares bimodal texture with carbonado but euhedral grains are larger	Variable, but mm-sized and larger crystals are common
Porosity	High (10% void space)	Low (1% void space)	Zero
Common Inclusions	Florencite-goyazite-gorceixite, xenotime, kaolinite, quartz, orthoclase, zircon, Fe, Fe-Ni, SiC, Si, Sn	Pyrope and almandine-pyrope garnet, Cr-rich clinopyroxene, chromite	Pyrrhotite, omphacite, almandine-pyrope garnet
$\delta^{13}C$ Composition	–23 to –30‰ with a mode at –27‰	–1 to –24‰ with a mode at –19‰	+3 to –34‰ with modes at –5‰ and –12‰
$\delta^{15}N$ Composition	–17 to +8‰	–3 to +15‰	–12 to +5‰

minerals have been identified, including orthoclase, quartz, and kaolinite. These phases are distinctly crustal, and, conversely, the mantle-derived inclusions commonly found in kimberlitic diamond (e.g., pyrope-rich garnet, chromian clinopyroxene, pyrrhotite) have never been reported from carbonado. The TEM results of Gorshkov et al. (1996) and De et al. (1998) also have revealed inclusions of native metals and metal alloys, such as Fe, Fe-Ni, Ni-Pt, Si, Ti, Sn, Ag, Cu, and SiC. These metal phases are nanometers to hundreds of microns in size, and they can be either enveloped within the diamond itself or present as a coating of the pores. Gorshkov et al. (1999) describe similar metal assemblages in kimberlitic polycrystalline diamonds from the Udachnaya pipe in Yakutia.

Geochemical Indicators for Formation

The similarities in the inclusion assemblages of carbonados from the Central African Republic and Brazil provide strong evidence that these now geographically distant carbonados are genetically related. Equally compelling support for a common origin comes from the tight clustering of carbon isotope compositions of nodules from the two localities, with $\delta^{13}C$ values ranging from –21 to –32‰ (Vinogradov et al. 1966; Galimov et al. 1985; Ozima et al. 1991; Kamioka et al. 1996; Shelkov et al. 1997) (Fig. 1). In situ isotope analyses (De et al. 2001) have shown that within individual samples, $\delta^{13}C$ values are very uniform, with a slight bimodality between the larger euhedral crystals (–26‰) and the finer-grained matrix (–24‰). Further confirmation for a connection between Central African and Brazilian carbonados is provided by noble gas analyses (Ozima et al. 1991; Burgess et al. 1998), which show that carbonados from both provenances contain high concentrations of implanted, radiogenic noble gases arising from ^{238}U fission. They also exhibit high contents of tightly trapped atmospheric gases.

Evidence that would serve as a “smoking gun” for the origin of carbonado is frustratingly elusive, and the ambiguity surrounding the role of mantle versus crustal processes in the forging of carbonados is even more pronounced than in the case of framesites, as described below. It is possible that carbonados are merely composite diamond clusters that formed in the same way as kimberlitic diamond nodules but with enrichment in light isotopes of C and He. If so, carbonados may represent the end member of a chemical continuum in which framesites serve as a bridge to eclogitic diamonds. In this scenario, the strong crustal signature in carbonados is related to the subduction of cold slabs beneath continental margins, such that organic matter is imported to the mantle with virtually no mixing of carbon reservoirs (Robinson 1978). This idea could perhaps explain light rare-earth element (LREE) patterns suggestive of a crustal origin (Shibata et al. 1993; Kamioka et al. 1996), the high concentrations of polycyclic aromatic hydrocarbons in carbonado pores (Kaminsky et al. 1991), and the presence of aggregated N defects (Nadolinny et al. 2003).

If carbonados represent mineralized organic carbon, their measured age of ~3.2 Ga, when added to the time required for plate subduction, suggests that these diamonds are vestiges of some of the oldest biological material known. Nevertheless, it should be noted that mantle xenoliths are often depleted in ^{13}C, and efforts to model the range of $\delta^{13}C$ values in these samples by simple mixing models of carbon from organic and mantle reservoirs have not succeeded (Deines 2002). Deines’ study suggests that poorly understood kinetic fractionation effects may play a role in the generation of isotopically light diamonds.

The implications of the radiogenic gases in carbonados are especially controversial. Some authors have proposed that exposure of carbonados to radionuclides occurred post-eruption, so that the elevated levels of radiogenic gases are secondary (Kagi et al. 1994; Shelkov et al. 1998; Burgess et al. 1998). On the other hand, the recent observation of crustal nucleogenic Ne in framesites indicates that some parts of the mantle may contain significant quantities of crustal noble gases (Honda et al. 2004). It is possible that, in this fashion too, carbonados preserve evidence for early atmospheric chemistry and plate tectonic activity. Still another hypothesis is that the radiogenic gases reflect formation of carbonados by the transformation of a carbon-

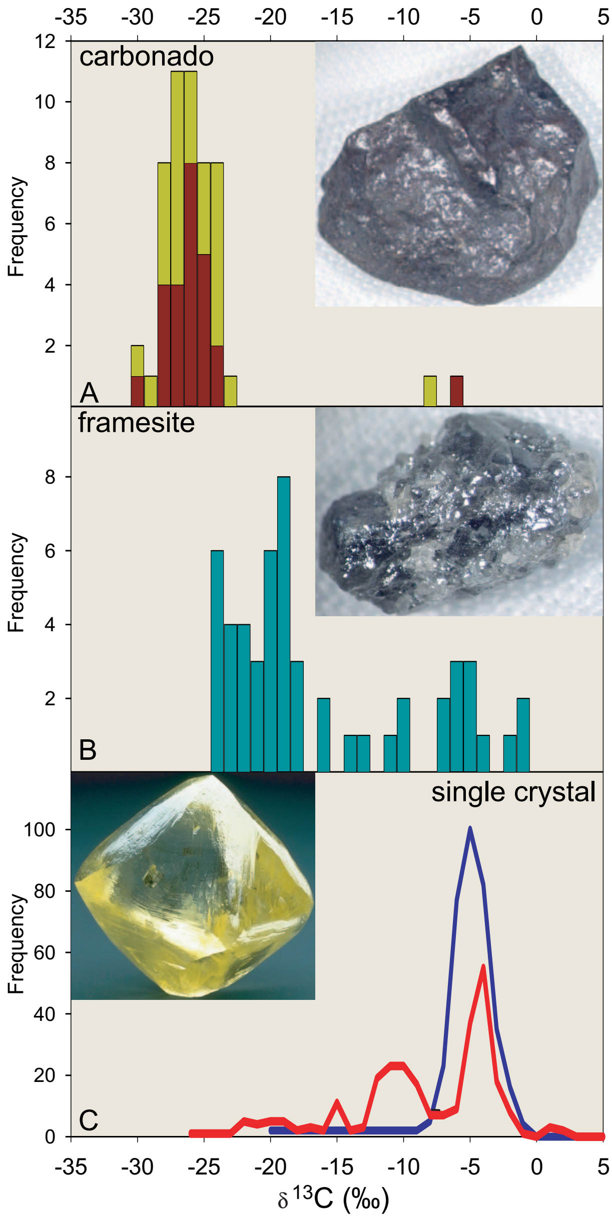

FIGURE 1 Compilation of published $\delta^{13}C$ values for (**A**) carbonado (avocado – Brazil; burgundy – Central African Republic); (**B**) framesite; and (**C**) monocrystalline diamonds (blue – harzburgitic; red – eclogitic). Insets: (**A**) carbonado specimen from Mambere River, Haute-Sangha Province, Central African Republic (15.9 mm across); (**B**) framesite specimen from Jwaneng, Botswana (9.0 mm across); (**C**) Oppenheimer diamond, Dutoitspan Mine, South Africa (38 mm across).

rich matrix to diamond through ^{238}U irradiation (Kaminsky 1987; Ozima et al. 1991; Ozima and Tatsumoto 1997). However, since natural examples of radiogenic diamonds (Daulton and Ozima 1996) do not exceed 500 nm in diameter, it seems that radiosynthesis might have provided, at most, the seeds for carbonado diamond growth.

Evidence for an Impact Origin

The nearly complete absence of a mantle fingerprint in carbonados has caused some researchers to seek a purely surficial crystallization process—a tricky proposition in light of the high pressures needed to stabilize the diamond structure. Smith and Dawson (1985) attacked this issue by invoking a meteorite impact within what was once a united landmass. The light $\delta^{13}C$ values for carbonado can then be attributed to shock metamorphism of organic matter, and the tightly trapped atmospheric gases, crustal inclusions, high polycyclic aromatic hydrocarbon levels, and magnetic inclusions all are logical results (Girdler et al. 1992; Kletetschka et al. 2000). Moreover, pervasive defect lamellae indicative of plastic deformation have been observed in TEM studies of both Central African and Brazilian carbonados (De et al. 1998).

Despite their undeniable appeal, however, impact scenarios for carbonado formation are troublesome. Deformation lamellae have been documented in framesites (DeVries 1973) and reproduced experimentally at mantle temperatures and pressures (De et al. 2004); thus, they may not be analogs of planar deformation features in shocked quartz. Unlike the impact-generated variety of PCD known as yakutite (Kaminsky 1991; Titkov et al. 2004), carbonados have never been shown to contain the hexagonal C polymorph known as lonsdaleite or any other high-pressure phase. Shock wave calculations (DeCarli 1998) indicate that the maximum size for a diamond formed by impact is ~1 cm, and impact-generated diamonds typically are submillimeter in dimension (e.g., Hough et al. 1995; Pratesi et al. 2003; Ding and Veblen 2004). As their large size is one of the hallmarks of carbonados, the impact scenario is difficult to reconcile with the physical attributes of this PCD variety.

FRAMESITE

Mineralogical Characteristics

In contrast to carbonados, there is no doubt that framesites are derived from kimberlite pipes that sample Earth's deep interior. Named for P. Ross Frames, chairman of the De Beers and Premier companies in the 1920s, framesite is produced from the Premier and Venetia mines in South Africa and the Orapa and Jwaneng mines in Botswana, where it can represent several weight percent of the total diamond output. Framesites from Russian localities (e.g., the Mir pipe) are known as well (Sobolev et al. 1975). A rigorous study of the range of diamond crystal sizes in framesite is lacking, but nodules comprising black or brown, euhedral, mm-scale diamonds cemented by randomly oriented, micron-sized crystals are not uncommon.

Framesites from different pipes exhibit subtle variations in diamond textures and included assemblages in the polycrystalline mass, but several studies have revealed evidence for a mixed eclogitic and peridotitic paragenesis. Two varieties of pyrope-rich garnet inclusions typically occur. Orange garnets with low Cr and moderate Ca contents have chemistries characteristic of eclogites, and pink garnets with higher Cr and lower Ca contents are representative of harzburgite (Gurney and Boyd 1982; McCandless et al. 1989; Kirkley et al. 1991). Olivine and eclogitic clinopyroxene or omphacite are curiously absent, but garnet, orthopyroxene, and peridotitic clinopyroxene commonly intergrow with the diamond and sometimes envelop diamond crystals, suggesting that diamond and silicate crystallization were contemporaneous (Kurat and Dobosi 2000). Chromite also can be a major accessory phase in framesites, and Cr concentrations in clinopyroxenes associated with framesites from Orapa, Jwaneng, and Mir are extraordinarily high (Sobolev et al. 1975; Gurney and Boyd 1982; Kirkley et al. 1991). Where present, magnetite imparts a strong natural remanent magnetization (Collinson 1998), giving rise to a subvariety of framesite called stewartite.

Carbon isotope studies of framesites from the Venetia, Orapa, and Jwaneng pipes (Kirkley et al. 1991; Shelkov et al. 1997; Burgess et al. 1998; Jacob et al. 2000) reveal a broad distribution of $\delta^{13}C$ values ranging from –2‰ to –25‰, with a major mode at –19‰ (Fig. 1). Moreover, noble gas analyses of framesite have yielded low $^3He/^4He$ ratios, indicating little contribution from primordial mantle-derived ^3He, and they also show high concentrations of crustal nucleogenic Ne (Burgess et al. 1998; Honda et al. 2004).

Theories of Formation

So how did framesites form? Virtually all authors attribute the fine grain size of framesites to rapid crystallization in localized areas of the mantle containing high concentrations of C and incompatible and volatile elements. Many scientists invoke subduction of ocean floor and subsequent mixing with upper mantle lithosphere as a mechanism for framesite crystallization (Kirkley et al. 1991; Burgess et al. 1998; Honda et al. 2004). This process explains the mixed eclogitic and peridotitic chemical signatures imprinted on many framesite samples, the unusual noble gas compositions measured for framesite diamonds, and possibly the broad range of $^{13}C/^{12}C$ ratios. Kirkley et al. (1991) propose that metamorphism of C from two crustal environments—one that was carbonate-rich and another that contained organics—could have generated populations of diamonds with distinct C isotope compositions.

Other researchers are less convinced of a crustal role. Kurat and Dobosi (2000) discount the importance of an eclogitic precursor and argue that framesite assemblages crystallized from upper mantle fluids containing a carbonatitic component, as suggested by trace-element profiles of garnets and of fluid inclusions in silicates. Jacob et al. (2000) also invoke a carbonatitic melt to explain trace-element, radiogenic, and stable isotope variations, but they argue that eclogitic material reacted with the melt. Moreover, these authors propose that framesites in the Venetia pipe crystallized very shortly before the kimberlite erupted. If true, framesites provide surprising evidence that remobilization of material stored for long periods in the cratonic lithosphere can lead to the formation of relatively young diamonds, that is, close to the emplacement age of 533 Ma in the case of Venetia.

SUMMARY

Polycrystalline diamond varieties once were regarded as eccentric expressions of the processes that generate kimberlitic diamonds, but state-of-the-art characterization studies have rendered the origins of carbonados and framesites more ambiguous. To the extent that these different varieties of PCD formed by unrelated mechanisms, their proper classification becomes an interpretive rather than merely a taxonomical exercise. As this review illustrates, widely accepted genetic models for these diamond aggregates remain an unfulfilled challenge, particularly with regard to the roles played by mantle and crustal components. Nevertheless, enough about these diamond varieties has been discovered to be sure that they will offer some startling insights about our planet and its history.

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