Editor: Evan M. Smith

The Liquids Lurking Inside Your Diamonds

Diamonds are dense, solid crystals of carbon. In top gem-quality form, a diamond appears to be a pristine, uniform, transparent material. It may be hard to believe, but even the highest-clarity natural diamonds are thought to contain nanometer-sized droplets of fluid. They are too small to see, even at high magnification with an optical microscope. And these are not the only kinds of fluid found in diamonds. Sometimes there are larger fluid inclusions, big enough to see with a microscope, that have either been trapped during diamond growth or have entered along small fractures that have healed and sealed themselves shut while the diamond was still deep within the earth's mantle. Some diamonds even contain colorful iridescent fluid mixtures of carbon dioxide and nitrogen. All of these fluids are reflections of the various natural processes that create and modify diamonds.

Fluids Trapped in Fibrous Diamonds

The discussion of liquid or fluid in diamond begins with something called *fibrous diamond*, which is generally not used as a gem because of its poor clarity. Fibrous diamonds contain abundant micrometer-sized inclusions (typically 0.1–1.0 μm) that render them cloudy or turbid in appearance. They often take the form of imperfect cube-shaped crystals or coatings atop octahedral diamonds (figure 1). Fibrous diamonds are thought to crystallize rapidly, and inclusions readily become trapped between their "fibers" or microscopic dendrites during growth (Kamiya and Lang, 1965; Navon et al., 1988; Sunagawa, 1990).

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Diamond growth in the mantle occurs at a depth of about 150–200 km and sometimes even deeper. Most forms of diamond growth, including fibrous diamond growth, occur by metasomatism, meaning the interaction between carbon-bearing fluids and preexisting host rocks (Haggerty, 1999; Stachel et al., 2005). As this fluid percolates along grain boundaries and cracks, it can undergo changes in temperature, pressure, or composition that cause its dissolved carbon to crystallize as diamond. A central aim of diamond research has been to understand what kinds of carbon-bearing fluids (or melts) contribute to diamond growth. In this respect, fibrous diamonds have offered considerable insight.

The abundant inclusions in fibrous diamonds are well-preserved samples of carbon-bearing, diamond-forming fluids (Navon et al., 1988). A range in fluid compositions has been observed, with four distinct "end-member" compositions: silicic, saline, high-magnesium carbonatitic, and low-magnesium carbonatitic (Navon et al., 1988; Izraeli et al., 2001; Tomlinson et al., 2006; Klein-BenDavid et al., 2007; Weiss et al., 2009). During the time of diamond growth, the fluid being trapped as inclusions was a single phase, without bubbles or crystals. Water makes up 10–25 wt.% of this fluid, with the remainder being dissolved material (Weiss et al., 2010; Elazar et al., 2019). Considering the high initial dissolved content, they are sometimes called high-density fluid (HDF) inclusions. However, when a kimberlite or related volcanic eruption carries a fibrous diamond up to the earth's surface, the cooler and lower-pressure conditions cause daughter crystals of silicate, carbonate, and other solids to form within the inclusions. The result is an inclusion containing multiple solid daughter phases and a residual fluid phase. These watery, carbon-bearing fluid inclusions represent snapshots of the parental diamond-forming fluid for fibrous diamonds. Interestingly, similar fluids may be involved in the growth of most transparent, gem-quality diamonds (Jablon and Navon, 2016; Krebs et al., 2019).

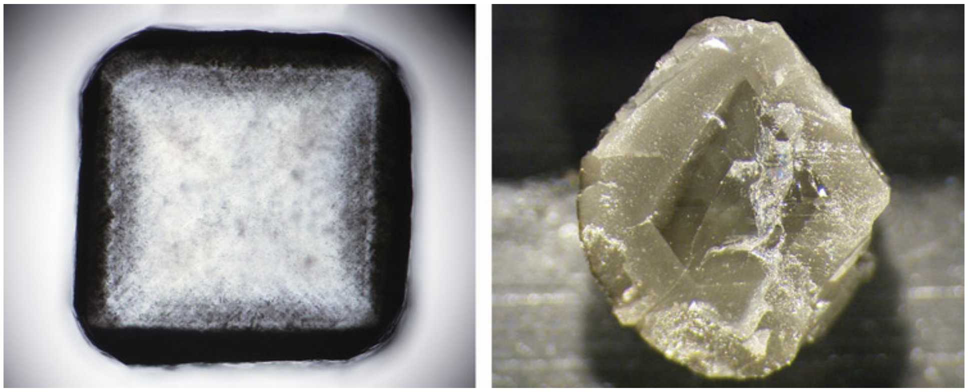

Figure 1. Examples of fibrous diamond. The polished slice (left) is roughly a {100} section through a fibrous cuboid. The broken diamond (right) is roughly a {110} section through a fibrous coat and a transparent, non-fibrous octahedral core. Photos by Evan M. Smith; fields of view 4.51 mm (left) and 5.38 mm (right).

Gem-Quality Diamonds

Why do scientists think there is a link between transparent gem-quality diamonds and the palpable cloudiness of fluid inclusions in fibrous diamonds? Part of the answer comes from macle twinned gem-quality diamonds. One study that searched for inclusions trapped along macle twin planes encountered fluid micro-inclusions in eight specimens from two localities, and the fluid compositions fell within the range of those seen in fibrous diamonds worldwide (Jablon and Navon, 2016). This finding was used to argue that fibrous and non-fibrous diamonds can form from the same kinds of parental fluids.

The second part of the answer lies in trace elements. Diamonds are made of a single major element, carbon, but also contain trace amounts of many other elements of the periodic table. The absolute and relative abundances of these elements represent a geochemical ledger that reflects the specific conditions and ingredients contributing to the growth of any given diamond. Comparing the trace element characteristics of transparent gem-quality diamonds with cloudy fibrous diamonds reveals striking similarities (Krebs et al., 2019). The similarity of trace element signatures provides an additional piece of evidence that many gem-quality diamonds crystallize from roughly the same kinds of fluids as those trapped in fibrous diamonds. Rather than occurring as substitutional or interstitial atoms in the diamond crystal lattice, the trace element signatures measured in gem-quality, transparent diamonds are proposed to come from sub-microscopic nano-inclusions of fluid like those of fibrous diamonds, but present in lower abundance. It implies that high-clarity, optically transparent diamonds that appear to be inclusion-free are actually laced with small droplets of fluid that are simply too small to see.

Exotic Metallic Liquid

There are multiple ways diamonds can form in nature, involving different host rocks, at different depths within the earth, and different kinds of diamond-forming fluids. We are still learning about all the different kinds of diamond-forming fluids, but we know there are multiple distinct groups. The range of saline-silicic-carbonatitic fluids found in fibrous diamond is just one of these groups. Some diamonds crystallize from completely different kinds of fluids. For example, we now have compelling evidence that some diamonds crystallize from methane-rich fluid (Smit et al., 2016) or even iron-rich metallic liquid (Smith et al., 2016). The latter are mixtures of iron, nickel, carbon, and sulfur that were molten at the time of diamond growth but now exist as solid inclusions (figure 2). Inclusions like these that are solid now at the ambient conditions of the earth's surface, but were molten liquid at the time of entrapment, are sometimes called melt inclusions.

Figure 2. Left: A type IIa diamond (4.12 ct, D color) with metallic inclusions. Photo by Jian Xin (Jae) Liao. Right: The large metallic inclusion has a localized round, graphitized fracture extending from its top end. Photomicrograph by Evan M. Smith; field of view 1.42 mm.

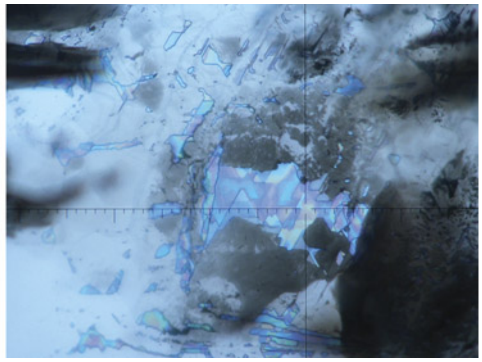

Figure 3. Left: A polished diamond slice (3.7 mm × 3.7 mm) with healed fractures and fluid inclusions. Right: The fluid inclusions display thin-film interference colors and have irregular polygonal boundaries governed by the diamond recrystallization as the fracture healed (field of view 245 µm). Photos by Evan M. Smith.

The solidified metallic liquid in figure 2 is a kind of inclusion that has been observed repeatedly in what can be called "CLIPPIR" diamonds (Smith et al., 2016, 2017). This is a variety of sublithospheric or super-deep diamond that originates from an estimated 360–750 km deep in the earth. Large, colorless (D color), type IIa diamonds often belong to this family. In fact, the initial recognition of this diamond variety stems from recurrent observations in the gem trade that larger, higher-quality diamonds have a greater tendency to be type IIa. The CLIPPIR acronym captures their distinguishing physical properties: Cullinan-like, Large, Inclusion Poor, Pure, Irregular, and Resorbed.

Out of 83 inclusion-containing CLIPPIR diamonds in one study, 67 had inclusions with metallic melt (Smith et al., 2017). The relative abundance of metallic melt inclusions is one of the features that makes these diamonds so interesting. The origin of this metal has been illuminated by measuring the isotopic composition of iron (Smith et al., 2021). The iron can be traced back to a process called serpentinization, where water chemically reacts with the rocks in oceanic tectonic plates. Iron from seafloor serpentinization was carried deep into the earth by the action of oceanic tectonic plates sinking down into the mantle, a process called subduction. After sinking down to 360–750 km, the serpentinized rocks contribute to the growth of CLIPPIR diamonds and are responsible for supplying the exotic metallic liquid sometimes trapped as inclusions. Tracing out this deep subduction pathway for serpentinized oceanic rocks is important for our understanding of how water, carbon, and other materials get transported from the earth's surface down into the convecting mantle over millions of years.

Healed Fractures

Fluid micro-inclusions in fibrous diamonds, invisible nano-inclusions in some gem-quality diamonds, and metallic melt inclusions in CLIPPIR diamonds are all examples of primary inclusions. This means they were trapped during diamond growth. It is also possible to trap material post-growth in fractures. If a fracture allows fluid to penetrate into a diamond and conditions allow the fracture to heal, it can trap some of the fluid as an inclusion. So-called fingerprint inclusions in corundum and other minerals also arise from secondary, post-growth fluid trapping along fractures.

A colorful instance of fluid trapped in a healed fracture in diamond is shown in figure 3. This is an alluvial diamond from the Ebelyakh River, Siberia. Alluvial diamonds from this region sometimes have discontinuous, black graphitic features inside them that trace out healed fracture networks. Under closer examination, these healed fractures regularly host fluid inclusions (Tomilenko et al.,

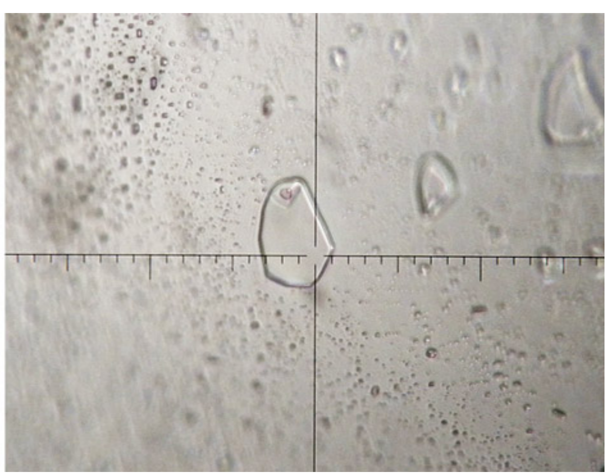

Figure 4. Left: A 3.66 ct octahedral diamond crystal (shown beside a millimeter scale) with silicate melt inclusions in healed fractures. The black and red ink marks are for planning prior to polishing for research purposes. Right: The largest inclusion (under the crosshairs) has a 4 micrometer wide pinkish bubble of nitrogen visible at the top end (field of view 180 µm). Photos by Evan M. Smith.

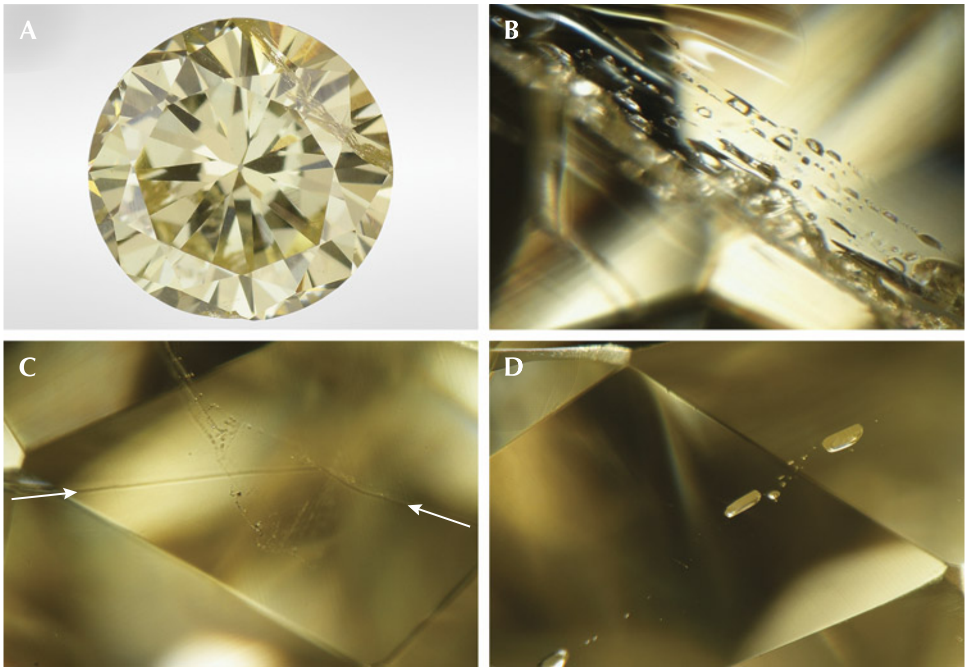

Figure 5. A 1.06 ct yellow diamond (A) containing a series of healed fractures with silicate melt inclusions (B–D). At bottom left (C), the healed fracture has interrupted the quality of polishing and appears as a surface grain line, indicated by arrows. Top left photo by Jian Xin (Jae) Liao. Photomicrographs by Evan M. Smith; fields of view 1.15 mm (B) and 1.42 mm (C and D).

1997, 2018). The inclusions in figure 3 contain a mixture of carbon dioxide (CO_2) and nitrogen (N_2), identified using Raman spectroscopy and microthermometry (Smith et al., 2015). These inclusions are often only a few micrometers thick, and thin-film interference gives them a colorful, iridescent appearance (like the colorful swirls seen in soap bubbles). The CO_2-N_2 inclusions are a snapshot of a free-flowing fluid that could have been percolating along crevices and between grains of the solid surrounding eclogitic host rock when this diamond sat in the mantle at a depth of 150–200 km (Ragozin et al., 2002; Smith et al., 2015).

Figure 4 shows another example of fluid trapped in a healed fracture in a diamond, this one from the Democratic Republic of the Congo. In this case, the fluid is best described as an ultramafic (magnesium-rich and silica-poor) silicate melt (Smith et al., 2014). When this melt solidified, it exsolved a nitrogen-rich (N_2) fluid phase that is visible as a pinkish-colored bubble within the largest melt inclusion (figure 4, right). Similar solidified melt inclusions were observed in a diamond from the Roberts Victor mine in South Africa (Smith et al., 2014) as well as in a yellow faceted diamond of unknown origin, shown in figure 5. The yellow diamond contains numerous transparent inclusions that lie together in an irregular planar configuration defining a healed crack. Individual inclusions are spatially confined to the plane of the crack, with larger inclusions having flattened, tabular shapes. Some inclusions are rodlike in shape or are grouped in lineaments that define the receding edge of the crack during healing. Raman spectroscopy shows that these solidified melt inclusions are dominated by olivine. The process responsible for making secondary silicate melt inclusions like these remains uncertain and requires further study.

Concluding Remarks

The fluid inclusions shown here—whether liquid, gaseous, or solidified melts—are only some of what have been encountered in natural diamonds, and future research is likely to uncover more. In the geological world, it is common to encounter fluid inclusions in minerals, but those found in diamonds have exceptional scientific value because of their deep mantle origin. Diamond is the only mineral that routinely captures and preserves fluids from great depths inside our planet and gets transported up to the earth's surface. Thanks to their gem properties, we can both admire and learn from these crystals.

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