Synthesizing Diamond From Liquid Feedstock

Precise proportioning of feedstock gases is not necessary.

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A relatively economical method of chemical vapor deposition (CVD) has been developed for synthesizing diamond crystals and films. Unlike prior CVD methods for synthesizing diamond, this method does not require precisely proportioned flows of compressed gas feedstocks or the use of electrical discharges to decompose the feedstocks to obtain free radicals needed for deposition chemical reactions. Instead, the feedstocks used in this method are mixtures of common organic liquids that can be prepared in advance, and decomposition of feedstock vapors is effected simply by heating.

The feedstock used in this method is a solution comprising between 90 and 99 weight percent of methanol and the balance of one or more other oxyhydrocarbons that could include ethanol, isopropanol, and/or acetone. This mixture of compounds is chosen so that dissociation of molecules results in the desired proportions of carbon-containing radicals (principally, CH_3) and of OH, H, and O radicals. Undesirably, the CVD temperature and pressure conditions thermodynamically favor the growth of graphite over the growth of diamond. The H radicals are desirable because they help to stabilize the growing surface of diamond by shifting the thermodynamic balance toward favoring the growth of diamond. The OH and O radicals are desirable because they preferentially etch graphite and other non-diamond carbon, thereby helping to ensure the net deposition of pure diamond. The non-methanol compounds are included in the solution because (1) methanol contains equal numbers of C and O atoms; (2) an excess of C over O is needed to obtain net deposition of diamond; and (3) the non-methanol molecules contain multiple carbon atoms for each oxygen atom and thus supply the needed excess carbon.

A typical apparatus used in this method includes a reservoir containing the feedstock liquid and a partially evacuated stainless-steel reaction chamber. The reservoir is connected to the chamber via tubing and a needle valve or other suitable flow controller. When the liquid enters the low-pressure environment inside the chamber, it evaporates to form a vapor mixture of the same chemical composition. In addition to the inlet for the feedstock liquid, the chamber is fitted with an outlet connected to a vacuum pump (not shown) through a throttle valve (also not shown) that is automatically controlled to keep the pressure at or near the required value throughout the deposition process.

Inside the chamber, a spiral filament made of tungsten, tantalum, graphite, or other high-melting-temperature material is electrically heated to a temperature >2,000 °C — high enough to cause dissociation of vapor molecules into the aforementioned radicals. A deposition substrate — typically, a diamond-polished silicon wafer about 2.5 cm square — is positioned about 2 cm away from the filament. The exact location of the substrate is chosen so that the substrate becomes heated by the filament to a deposition temperature in the approximate range of 800 to 1,000 °C.

The composition of the feedstock must be chosen in conjunction with other operational param-eters to obtain a high-quality diamond deposit. When the feedstock comprises methanol alone, no diamond is deposited on the substrate and there is too much oxidation of the hot filament and consequent gradual reduction in the diameter of the filament. When the liquid contains too much ethanol, isopropanol, or acetone, the filament becomes coated with graphite and thus swollen. When the substrate is placed too close to the filament, the concentration of the etchant radicals is too high, preventing net deposition. In experiments, it was found that choice of an optimum combination of composition of the feedstock, filament temperature, filament-to-substrate distance, and vapor pressure results in the deposition of high-quality diamond (see figure) on the substrate. Moreover, it was found that if the optimum vapor pressure is established in the chamber before heating the filament, then the filament becomes coated with a thin layer of carbon that prevents erosion of the filament by the etchant radicals during the deposition process.

This **Optical Micrograph** at a magnification of about 500 shows diamond grains in a layer ≈10 μm thick on a silicon substrate. The diamond was deposited using a feedstock solution of about 4 weight percent ethanol in methanol, a vapor pressure of 40 torr (≈5 kPa), a 1.5-mm-diameter tungsten filament wound in a spiral of ≈2.5 cm diameter and heated by a current of 110 A, and other deposition conditions as described above.

This work was done by Yonhua Tzeng of Auburn University for **Goddard Space Flight Center**.

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