

Multiple flame deposition of diamond films

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A multiple flame burner has been used to deposit diamond films of up to 20 mm in diameter from an oxygen-acetylene mixture. The burner consists of nine equally spaced linear holes each sustaining its own flame and rotates under a water-cooled substrate. The diamond film's quality is characterized as a function of its radial distance from the center of the film by scanning electron microscopy, Raman spectroscopy, and photoluminescence spectroscopy. Typical films exhibit variations in thickness and crystal structure with an increasing graphitic component present towards the edge of the film as evidenced by Raman analysis. Photoluminescence spectra exhibit defect bands at 1.95 eV present near the outer edge of the film and at 2.16 eV present near the center of the film. These luminescence bands are discussed and attributed to defects induced from the flame's chemistry.

Diamond exhibits several desirable properties that make it a candidate for applications ranging from coating optical windows to insulating power microelectronic devices. Among these properties are diamond's extreme hardness (10 000 kg/mm²), high thermal conductivity (20 W/cm K), high electrical resistivity (10¹⁶ Ω cm), chemical inertness, optical transparency in the visible to far-infrared spectrum, and low coefficient of friction (0.1). Diamond films can be deposited from several chemical vapor deposition (CVD) techniques¹⁻³ using relatively low temperatures (400–1200 °C) and pressures (below atmosphere) to obtain diamond in its metastable form.

The discovery of diamond synthesis from combustion flames in open air⁴ has provided an alternative to low-pressure diamond CVD techniques. Flame deposition of diamond is attractive due not only to the simplicity of the process but also to the outstanding quality of the diamond films obtained. Conventional single-tip torches limit the total area of diamond deposition to a few millimeters.^{5,6} Experiments conducted by linearly scanning an oxyacetylene flame over foreign substrates have yielded narrow but long continuous diamond films limited only by the scan speed and deposition time.⁷ Most applications involving large-scale diamond deposition require a slow scan speed to obtain the diamond film quality and thickness desired, thus lengthening the deposition time considerably. To overcome this drawback, we will report the results obtained from a nine-hole linear torch capable of depositing diamond films of 20 mm in diameter.

A torch was built consisting of nine holes linearly spaced 3 mm apart with each hole having a 1 mm diameter. Oxygen and acetylene gas flows were controlled by two separate flow tubes. The torch was oriented so that the flame would burn upwards mainly to keep the hot exhaust gases from heating the torch tip. Molybdenum disks used as substrates were attached with silver paint to a water-cooled copper block and the substrate temperature was controlled by adding thin stainless-steel disks between the substrate and the substrate holder, thus increasing the ther-

mal resistance. The substrate holder was rotated by a dc motor at around 60 rpm so that a circular area on the substrate was exposed to the flame, as can be seen in Fig. 1. Several safety devices to deal with the open flame hazard and the potential of flashback were also installed, including flashback arresters, a quick shut-off mechanism, and a nitrogen purge line used in lighting and extinguishing the flame.

The molybdenum substrates used were scratched with 1 μm diamond powder and then solvent cleaned with acetone and methanol. Typical operating parameters include an oxygen to acetylene flow ratio between 0.9 and 1.0. The temperature of the substrate was around 800 °C. Exact surface temperature could not be measured with an optical pyrometer due to the substrate's position and the intensity of the infrared emissions from the flame. Continuous circular diamond films with diameters of 20 mm were deposited in about 2 h. After the substrate cooled down from the deposition temperature, the films were left free-standing due to the mismatch between the coefficients of thermal expansion of the diamond film and the molybdenum substrate.

The characteristics of the films deposited by the nine-hole torch exhibit a radial dependence (see Figs. 2–5). The variation in film thickness and quality can be caused by several factors including temperature gradients, inward diffusion of surrounding air, and nonuniform flow rates through the nine holes. A more uniform multiple hole linear flame may be possible if the gas flow through each hole is controlled separately.

Figures 2–5 show the results obtained from analyzing the diamond films deposited. For each analytical technique used [thickness measurements, scanning electron microscopy (SEM), Raman spectroscopy, and photoluminescence spectroscopy (PL)], data were collected at five different locations along the radius of the film so as to detect any nonuniformity in the film's structure and composition. The thickness of the film tested varied from 18 μm at the center of the film to 40 μm closer to the outer edge (Fig.

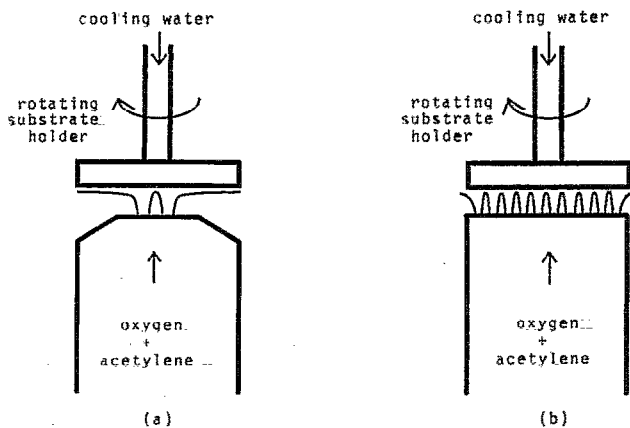


FIG. 1. Schematic illustration of the nine-hole linear torch. (a) Side view showing the acetylene feather covering the entire substrate; (b) side view showing the nine inner flames.

2). Figures 3 and 4 show the Raman spectra and SEM micrographs of the sample. The ratio of the intensity of the diamond Raman peak to that caused by imperfections in diamond crystallites deteriorates with movement away from the center. The corresponding SEM micrographs show the diamond particle size approximately in proportion to the radial dependence of the growth rate as shown in Fig. 2.

The PL spectra in Fig. 5 show that the zero phonon line (ZPL) PL band at 2.16 eV dominates near the center of the film and the ZPL PL band at 1.95 eV and its phonon replicas are more pronounced closer to the edge of the film. The 2.16 eV band has been attributed by researchers to a divacancy⁸ and to a complex center related to nitrogen.^{9,10} The 1.95 eV band has been assigned to a nitrogen-vacancy pair⁸ and is predominant at the outer edges of the film due to the inward diffusion of nitrogen from the surrounding air.¹¹ The high-pressure area at the center of the film does

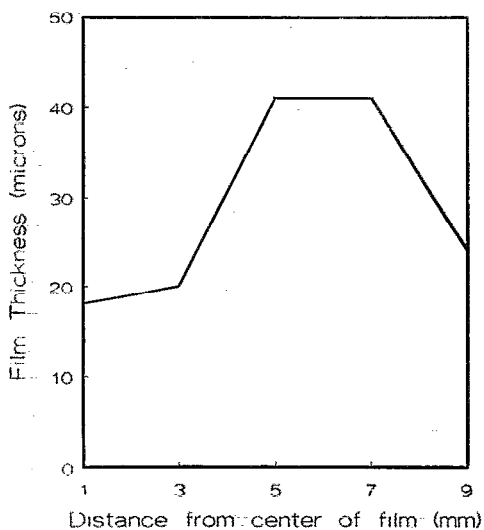


FIG. 2. Radial dependence of film thickness from the center of a circular film to the edge.

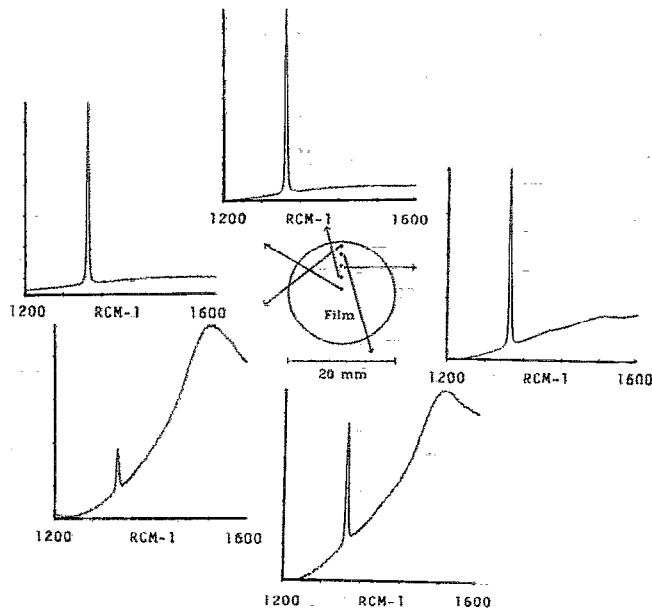


FIG. 3. Raman spectrometer measurements taken at different locations along the radius of a circular film. Clockwise from upper left-hand corner: Raman plots taken from center outward.

not allow the nitrogen to enter and thus no observable bands at 1.95 eV are present. The PL spectrum at half the radial distance from the center of the film produces phonon replicas that are easier to observe. The signal to noise ratio in this region is better due to the larger crystallites present, as evidenced by the SEM micrographs of the same location.

In summary, diamond films with well-faceted crystallites covering the entire area of 20 mm in diameter have been deposited using a nine-flame linear oxy-acetylene torch with a rotating substrate. Further improvement in the uniformity of both film thickness and quality is being conducted.

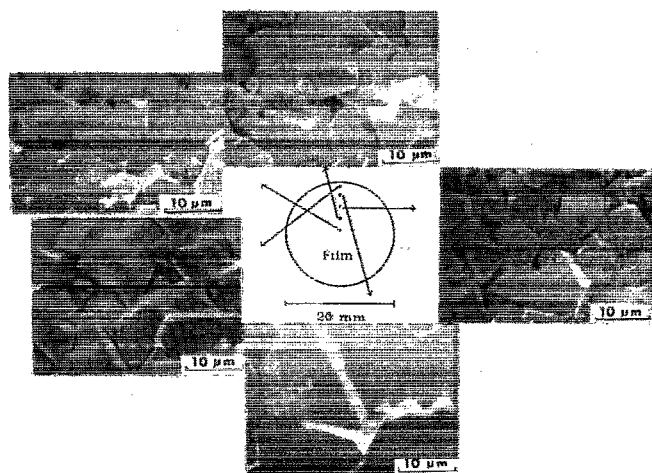


FIG. 4. Corresponding SEM micrographs taken at different locations along the radius of a circular film. Clockwise from upper left-hand corner: SEM pictures taken from center outward.

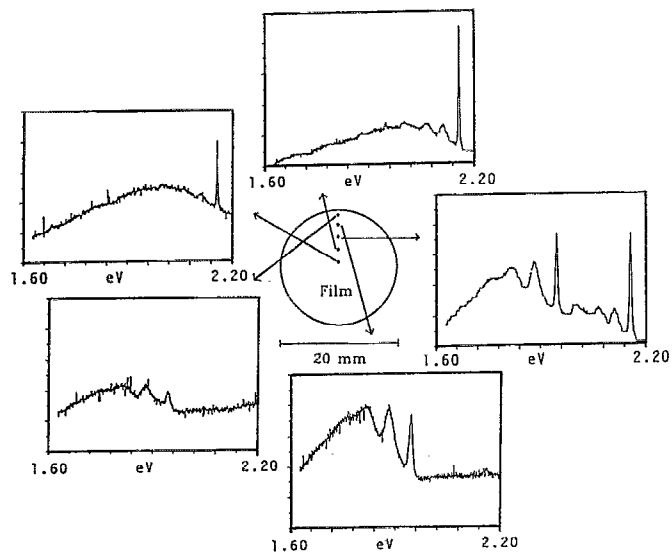


FIG. 5. Photoluminescence spectra taken at different locations along the radius of a circular film. Clockwise from upper left-hand corner: PL spectra taken from center outward.

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