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HAADF-STEM Study of Filament Material in Hot Filament CVD Diamond Films

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The extremely high thermal conductivity of diamond thin films grown by chemical vapor deposition (CVD) offers a novel solution to thermal management issues for electronic devices [1]. Hot filament CVD (HFCVD) is a particularly attractive approach for growing diamond films because of the lower equipment costs compared to other methods. Previous work has detected filament material in diamond films using SIMS [2] and RBS [3] but these techniques lack the spatial resolution to understand the distribution of the material in the diamond. A detailed evaluation of any contamination in the diamond is vital to understanding its thermal transport properties because even parts-per-million levels of impurities have been shown to significantly reduce the thermal conductivity [4].

A ~1.5 µm thick diamond film was deposited on a 100 mm [100] Si wafer seeded with diamond nanoparticles using a mixture of 1.5 % methane balanced with hydrogen. The heater assembly consists of an array of 9 commercial grade W filaments measuring 0.25 mm in diameter spaced ~ 1 cm apart and heated using a DC power supply set to 6 kW. The wafer was placed on a rotating, water-cooled stage 6 mm below the heater wires. A lamella was prepared by focused ion beam milling using a FEI Helios Nanolab 400 operated at 5-30 kV and then heated in air to remove surface damage on the diamond. [5] High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) images and energy dispersive x-ray spectroscopy (EDS) spectra were collected using a JEOL ARM 200F operating at 200 kV. Additional EDS spectra were collected from an unused filament wire by SEM using a JEOL 6010 PLUS/LA operating at 8 kV.

Figure 1(a) is a HAADF-STEM image collected from the diamond. In this image we can see bright dots measuring ~1.5 nm in diameter distributed throughout the field of view. In addition to these dots, on a larger scale, more diffuse regions of lower intensity are also observed. An EDS spectrum collected from the diamond is shown in figure 1(b), and identifies Al, Cr, Fe, and W (the Ga and Cu peaks are attributed to the sample preparation and the support grid, respectively) in the diamond.

A larger concentration of the deposited material is found to accumulate at the diamond-Si interface as shown in the HAADF-STEM image in figure 2(a) that indicates a bright region 10 nm thick layer between the two. Analysis of this region by EDS in figure 2(b) shows a distinct composition compared to the diamond. The Fe and W signals have increased while the Cr has decreased. Al can still be detected but now overlaps with a W peak, but a distinct splitting can be seen upon close inspection. EDS scans from the filament wire confirm the origin of the impurities observed in the film. Scanning individual points indicated that the wires were mostly free from impurities, however, localized regions showed high impurity concentrations. The spectrum from one such region is shown in figure 2(c). We see clear peaks for Al and Ca which were identified in the interfacial region and a Fe peak could also be detected. Na was also identified as being present on the wire but was not observed in the film. No Cr was identified in these scans. However, due to the sporadic distribution of the impurities it is possible that none was present in any of the regions scanned.

An understanding of the impurities in the diamond is critical to understanding the thermal and electrical properties of these films, especially when integrated with high-frequency, high-power devices.

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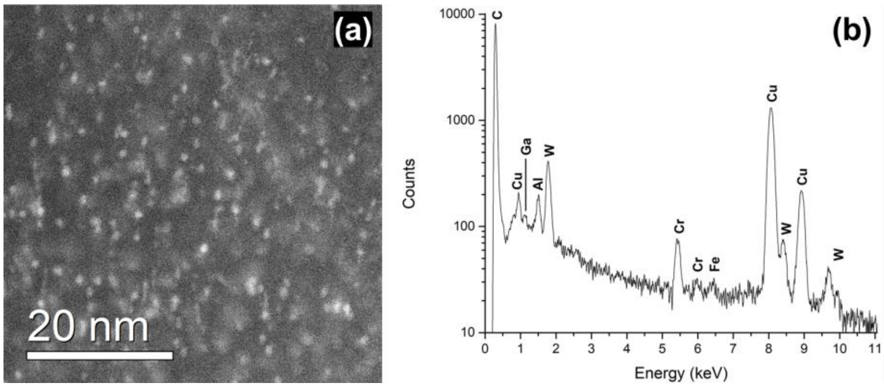

Figure 1. (a) HAADF-STEM image showing incorporation of filament material as bright dots and a diffuse haze. (b) EDS analysis of the region identifies the presence of W, Al, Cr, and Fe.

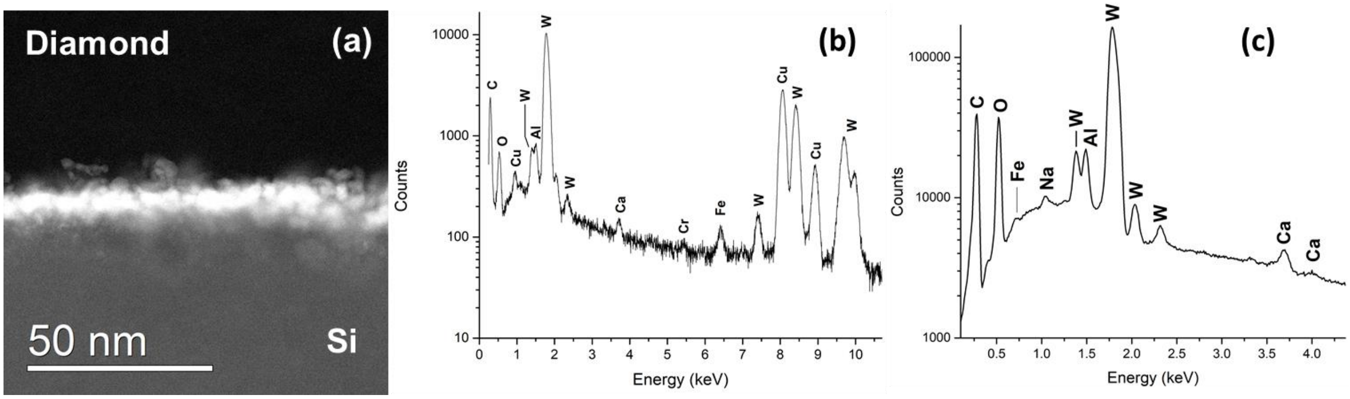

Figure 2. (a) HAADF-STEM image showing a layer of filament material at the diamond-Si interface. (b) EDS analysis of the interface shows a different composition from that observed in the diamond film. (c) EDS analysis of an unused filament wire by SEM confirming the source of the impurities.