Investigation of Nanodiamond Foil Product for H- Stripping to Support Spallation Neutron Source

Rachel Lim
Materials Science, Rice University

NNIN REU Site: Howard Nanoscale Science & Engineering Facility, Howard University, Washington, DC
NNIN REU Principal Investigator: Dr. Gary Harris, Electrical Engineering, Howard University
NNIN REU Mentor: Mr. James Griffin, Electrical Engineering, Howard University
Contact: rel3@rice.edu, gharris@msrce.howard.edu, griffin@msrce.howard.edu

Abstract:

Diamond is an ideal material as an H- stripper foil for spallation neutron source (SNS) applications due to its high thermal conductivity, low molecular weight, and strength. Polycrystalline diamond is characterized by a high degree of internal stress, which causes the foil to curl when not supported by the substrate. Hot filament chemical vapor deposition (HFCVD) was used to grow diamond on a silicon substrate. A 1.2 cm diameter window was etched in the silicon using a 1:1:3 solution of hydrofluoric, nitric, and acetic acids, so that the diamond foil would be suspended while being supported on all sides by the silicon. Wax and diamond were used as masks to protect the outer silicon from etching. Raman spectroscopy verified a high quality diamond foil. Atomic force microscopy (AFM) revealed that the foil originally against the substrate had an average roughness of 6.7 nm while the foil away from the substrate had an average roughness of 13.2 nm. Scanning electron microscopy (SEM) revealed no cracks in the suspended foil.

Introduction:

SNS is a process that produces intense neutron beams for research. An ion source produces H- ions, hydrogen atoms with two electrons, which are injected into a linear particle accelerator. The ions pass through a foil that strips them of their electrons, yielding protons. The protons are collected in accumulator ring, then released in high energy pulses towards a liquid mercury target. Neutrons are ejected from the mercury target upon impact, which can be used for different experiments.

Currently, a carbon stripper foil is being used, but diamond would be an ideal foil. It would be able to withstand the high energy radiation because of its high thermal conductivity, low molecular weight, and strength. The foil would not have to be changed as often because diamond is more durable than just carbon.

Experimental Procedure:

A piece of <100> silicon was seeded with a nanodiamond slurry solution for ten minutes in an ultrasonic bath, then the sample was loaded into the HFCVD reactor. The working distance between the filaments and the sample was set to 20 mm, the process pressure was set to 20 torr, and the diamond heater was set to 750°C. A flow of 80.0 sccm of H_2 and 1.0 sccm of CH_4 was introduced, and when the process pressure reached 20 torr and the diamond heater temperature reached 650°C, the filament temperature was raised to 2350°C. The nanodiamond film was left to grow for 6-8 hours.

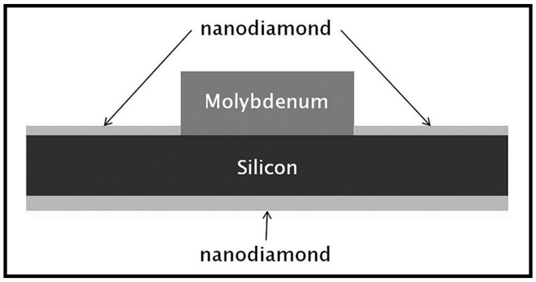

Figure 1: The circular piece of molybdenum placed in the center of the sample.

After growth of the top foil layer, the sample was flipped over, and a circular piece of molybdenum was placed in the center of the sample (see Figure 1). A diamond layer for etch masking was grown on the backside for two hours with conditions similar to the topside foil layer. The molybdenum prevented diamond from growing on part of the backside of the sample. Some of the samples were annealed at 600°C for 20 hours in a 5.0 sccm flow of N_2 at a process pressure of 20 torr in an attempt to reduce the internal stresses in the diamond foil.

The sample was placed in a 1:1:3 solution of 48% hydrofluoric, 70% nitric, and 100% acetic acids to etch a window in the silicon. The ratio of acids determined the etch rate. If the etch was too fast, the diamond came off in little pieces. If the etch was too slow, it did not etch all the way through the silicon. The backside layer of diamond protected the silicon outside of the circle from the etch, yielding a suspended diamond foil

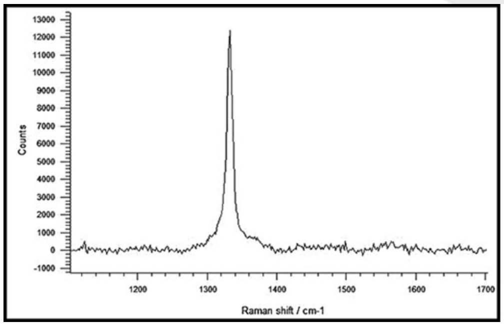

Figure 2: Raman spectroscopy verifying a high quality diamond foil with a strong diamond peak and a very weak graphite peak.

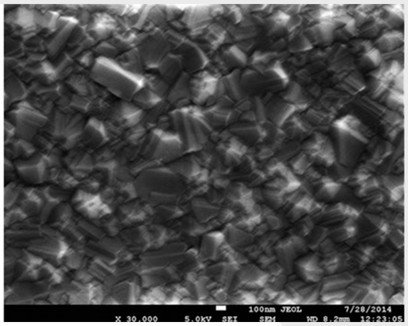

Figure 3: SEM image verifying the presence of polycrystalline diamond.

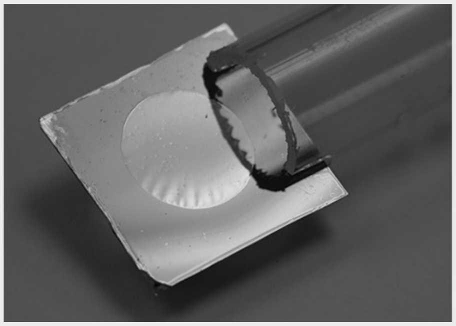

Figure 4: Optical imaging reveals no cracks in the suspended foil. (See full color version on page xxxvi.)

that was supported by the silicon. Once all of the silicon in the circle was etched away, the sample was rinsed in DI water, then allowed to air dry.

Results and Conclusions:

A high quality diamond foil was grown on a <100> piece of silicon by HFCVD. Raman spectroscopy verified a high quality diamond foil with a strong diamond peak at 1333 cm^{-1} and a very weak graphite peak as seen in Figure 2. Both SEM and AFM showed the presence of polycrystalline diamond as seen in Figure 3. A 1:1:3 acid ratio was found to be ideal because it was not too fast or too slow. Using diamond as a mask to protect the silicon during etching worked better than wax by providing better mask control and greater resistance to acid etching. AFM revealed that the foil originally against the substrate had an average roughness of 6.7 nm, while the foil away from the substrate had an average roughness of 13.2 nm.

SEM and optical imaging (see Figure 4) revealed no cracks in the suspended foil. Both 6-hour diamond film growths did not crack. The annealed sample appeared to be smoother, which means that it had less internal stress. The 8-hour unannealed diamond film growth cracked, while the 8-hour annealed film growth did not. It appears that annealing the diamond foil before etching away the silicon relieves some of the stresses in the film.

Future Work:

Currently, the diamond foils that have been grown are under much internal stress. Initial results suggested that pre-etching annealing of the diamond is useful for relieving internal stress. Future work should concentrate on various anneal temperature and times for stress reduction. Another area of investigation should involving introducing argon to the gas species during diamond growth. Depending on the argon concentration during growth, diamond grains can be reduced from poly to nanocrystalline. Smaller grains may also reduce the stress in the film. Lastly, foil testing by SNS should be performed to verify diamond foil performance.

Acknowledgements:

I would like to thank the NNIN REU Program, the NSF, and the Howard Nanoscale Facility (HNF). I would also like to thank my PI, Dr. Gary Harris, and my mentor, Mr. James Griffin, for their help and guidance, and Ms. Bokani Mtengi for training me at Howard University. Lastly, I would like to thank Dr. R.D. Vispute from Blue Wave Semiconductors for his expertise on this project.

References:

[1] V.Kh. Liechtenstein, T.M. Ivkova, E.D. Olshanski, R. Repnow, J. Levin, R. Hellborg, P. Persson, T. Schenkel. “Advances in targetry with thin diamond-like carbon foils”; Nuclear Instruments and Methods in Physics Research A, 480 (2002) 185-190.

[2] R.W. Shaw, D.P. Bontrager, L.L. Wilson, C.S. Feigerle, C.F. Luck, M.A. Plum. “An Electron Beam SNS Foil Test Stand”; White Paper.

[3] T. Spickermann, M.J. Borden, R.J. Macek, R.W. Shaw, C.S. Feigerle, I. Sugai. “Comparison of carbon and corrugated diamond stripper foils under operational conditions at the Los Alamos PSR”; Nuclear Instruments and Methods in Physics Research A, 590 (2008) 25-31.

[4] R.W. Shaw, V.A. Davis, R.N. Potter, L.L. Wilson, C.S. Feigerle, M.E. Peretich, C.J. Liaw. “Corrugated Thin Diamond Foils for SNS H-Injection Stripping”; White Paper.

[5] R.A. Campos, V.J. Trava-Airoldi, O.R. Bagnato, J.R. Moro, E.J. Corat. “Development of nanocrystalline diamond windows for application in synchrotron beamlines”; Vacuum 89 (2013) 21-25.

[6] RD Vispute, H.K. Ermer, P. Sinsky, A. Seiser, R.W. Shaw, G. Harris, F. Piazza. “Nanodiamond Foil Product for H- Stripping to Support Spallation Neutron Source (SNS) and Related Applications”; White Paper.