

## DIAMOND STRIPPER FOIL EXPERIENCE AT SNS AND PSR

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### Abstract

The SNS is currently operating at about 35% of the 1.4 MW design power, and the diamond stripper foils developed at ORNL continue to perform well. Several corrugated, nanocrystalline diamond stripping foils have been tested at SNS. Beyond about 300 C of injected charge, significant distortion and darkening of the foils is observed. These foils are currently limited in freestanding area to about 17x25 mm due to stress-induced tears in larger foils. The PSR experience with these diamond foils has been promising, with the interesting observation that both the foil current due to secondary emission of electrons and the thermionic foil current are reduced for diamond foils relative to LANL/KEK foils. For comparable thickness foils, losses due to the  $H^0$  yield also appear to be higher for diamond. A recent development in our foil preparation has been a change to nano-seeded nucleation from the earlier microcrystal slurry ultrasonic abrasion technique. This has led to a more reproducible and uniform foil morphology with smaller crystallites. We now determine foil thickness using alpha particle ranging.

### INTRODUCTION

We first discussed the use of polycrystalline diamond foils for use at SNS at PAC03.[1-3] In order to stiffen the diamond foils, physical corrugations are incorporated by patterning the silicon growth substrate before microwave plasma enhanced chemical vapor deposition (CVD) growth. An acid etch of a masked sample after diamond growth releases a large, free-standing foil that is supported on only one edge. Since our last report at PAC07 [3], two developments have been undertaken. Nano-seeded nanocrystalline diamond preparation has replaced abrasion nucleation and our foil thickness determinations are accomplished using alpha particle ranging. Those two developments, along with the current diamond foil experience at both the SNS and the LANSCE PSR are summarized here.

### FOIL FABRICATION AND THICKNESS

#### *Nano-seeded Diamond Foils*

We have implemented a new method for diamond growth nucleation for stripper foils. In the past, we have abraded the silicon wafer surface by ultrasonic treatment in a suspension of diamond particles in methanol. We now seed the surface of the silicon with nano-sized (<10 nm) diamond particles (#636444, Sigma-Aldrich, St. Louis, MO). The goal is to create a dense bed of nuclei upon which to grow larger particles, and eventually a

continuous film, during the plasma CVD process. According to Ralchenko [4] and Xiao [5], seeding the surface of silicon by ultrasonic treatment with nanodiamond powder in an appropriate solvent provides high nucleation densities ( $>10^{10}$  cm<sup>-2</sup>). The foils were examined using scanning electron microscopy (SEM) and were found to be continuous and almost free of seed aggregates (potential points of physical weakness in free-standing foils) when dilute seeding slurries (63 µg/ml) were used. The growth conditions used to prepare SNS nanodiamond foils were: 150 Torr total pressure (2% CH<sub>4</sub>, 8% hydrogen, and 90% argon), at 100 SCCM flow rate, with 1000 W of microwave power, and 1 hr. Foils produced *via* the nano-seeding method exhibit approximately uniform 100 nm grains.

#### *Alpha Ranging for Foil Thickness Determination*

Typically we have observed a thickness gradient (20-30%) for our diamond samples along their long axis. This gradient results from a microwave plasma ball dimension in the growth process that is only comparable to the sample size. We determined average film thickness by weighing the substrate wafer before and after diamond growth; this, however, produces an average thickness, not the true thickness at the beam injection location. More recently we have implemented an alpha ranging method [6] in order to measure a more appropriate thickness. The energy of alpha particles is degraded as they pass through condensed matter, and the energy shift can be experimentally or theoretically related to the sample thickness, if the density is known.

A commercial alpha spectrometer (Model 256, Tennelec, Oak Ridge, TN; 10 keV channel width) was employed. The source was a thin oxide film of Am-241 (2200 Bq). An aperture plate was positioned in the spectrometer; the diameter was 7.5 mm, representing the  $2\sigma$  diameter of the SNS injected beam. The sample foil was positioned on the aperture plate. The convenience of this thickness measurement technique is due to the fact that a standard commercial spectrometer was used without modifications.

Determination of the spectral energy shift was made using visual inspection. Any spectral broadening was beyond our resolution limit. To calibrate the alpha data against the actual sample thickness, the sample was sacrificed, and the cross-sectional thickness was determined using an SEM (Stereoscan 420, Leica, Thornwood, NY). The plot in Figure 1 shows the correlation between the SEM-determined thickness and

the alpha energy shift, as well as a linear least squares fit. Single-channel alpha spectral resolution corresponds to  $13 \mu\text{g}/\text{cm}^2$  thickness.

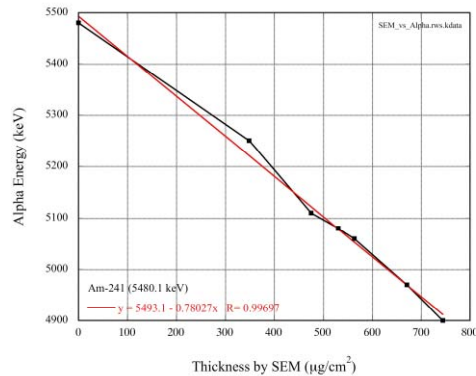


Figure 1: Correlation between diamond foil thickness as determined using SEM images and the energy shift of Am-241 alpha particles.

In addition to physical calibration using the SEM, which is inherently destructive, we also used the Monte Carlo Stopping and Range of Ions in Matter (SRIM) code (Ver. SRIM-2008.03) [7] to calculate energy shifts for the foils. A carbon target was chosen, but a  $3.51 \text{ g}/\text{cm}^3$  density (diamond) was selected rather than the default carbon value. SRIM agreement with the measured energy shifts was very good. For six samples ranging from 349 to  $744 \mu\text{g}/\text{cm}^2$  (by SEM), the mean error of the SRIM-determined thickness was  $17 \mu\text{g}/\text{cm}^2$  (4.2%). Thus, SRIM can be employed for calibration of foil thickness, and samples can subsequently be used at the accelerator.



Figure 2: SNS foil 601 after 300 C.

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### Primary SNS Foil

The diamond stripper foils presently in use at SNS are 17 mm wide, 45 mm tall (including the 20 mm tall silicon “handle”), and about  $300 \mu\text{g}/\text{cm}^2$  thick. This is wider and thicker than the design dimensions of 12 mm and  $270 \mu\text{g}/\text{cm}^2$  due to a beam loss issue in the injection dump beam line [8]. These foils have continued to perform nicely under all beam conditions as we have increased the SNS beam power up to the present production power of 500 kW, and during high intensity studies up to  $1.3 \times 10^{14}$  protons per pulse. Temperatures are now high enough that the  $\text{H}^-$  injected beam spot is easily visible using a video camera.

There have not been any catastrophic foil failures to date other than one foil that fractured in the early days of commissioning during a high intensity experiment. However, four foils have been removed from service due to a corner curling to about a  $30^\circ$  angle (Figure 2; foil curled away from the camera), or in order to insert a thinner foil to reduce beam loss in the ring. Corner curling at the beam location is a common occurrence for SNS diamond foils; the operational response to this curling is to insert the foil further into the beam. For larger insertions, an excessive amount of circulating beam passes through the foil, leading to unacceptable beam losses. In addition to curling, the foil darkens substantially indicating a conversion to graphitic character. That phase change is probably related to the observed curling. To date we have only observed foil curling toward the beam upstream direction, regardless of the diamond film orientation (front face/rear face) relative to the beam.

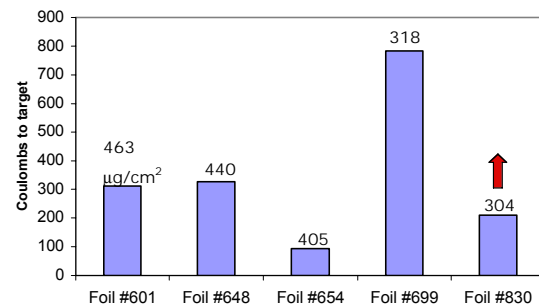


Figure 3: Integrated SNS charge delivered to target for 5 nanocrystalline diamond foils before removal from service. #830 is still in use as of 28/May/08.

A plot of the integrated beam charge delivered to the neutron spallation target for each of the foils that have experienced significant injected beam charge and have been removed for visual examination is shown in Figure 3. The chart bars are ordered in time sequence of foil use, during which operation increased from nominally 10 kW at 5 Hz to 500 kW at 60 Hz. Foil #699 injected almost 800 C of  $\text{H}^-$  before it was retired. Note in Figure 3 that foil #699 is actually  $17 \times 20$  mm unsupported diamond. The only nano-seeded foil plotted is # 830, which is still in use as of May 28, 2008. Currently five nano-seeded foils are loaded in the SNS ring. The thicknesses cited in Figure 3 are wafer-averaged values.

### Secondary SNS Foil

In addition to the primary diamond stripper foil, the SNS ring also requires a secondary stripper foil. Any  $\text{H}^-$  beam that misses the primary foil (2% by design) and the partially stripped  $\text{H}^0$  beam (3% by design) are intercepted at a secondary foil to fully strip them to  $\text{H}^+$  so they can be transported to a beam dump. The ideal secondary foil would be made of some form of carbon, and would be about 12 cm wide, 10 cm tall, and  $1 \text{ mg}/\text{cm}^2$  thick. We presently have a two-piece secondary foil -- one piece is 6.1 cm wide and the other is 5.6 cm wide, and both are

$\sim 19 \text{ mg/cm}^2$  thick -- carbon-carbon material from Allcomp (City of Industry, CA). This is the thinnest pinhole-free carbon-carbon material that we could find.

Obviously the Allcomp foils are much thicker than desired, which leads to excessive beam loss due to scattering. Also, after a 3-month run cycle the Allcomp foils are seriously degraded. As shown in Figure 4, a lamp positioned behind a foil shows many pinholes where the beam has passed through the foil. We are now pursuing alternative foils.



Figure 4. SNS secondary carbon/carbon foil after a 3-month run cycle. The foil halves (left,  $\text{H}^0$ ; right,  $\text{H}^+$ ) have been pieced together for this image.

### PSR Foils

Our polycrystalline diamond foils have also been evaluated at the LANSCE PSR. In general, the foil current due to secondary emission of electrons and the thermionic foil current are reduced for diamond foils relative to LANL/KEK foils. [9]. The conductivity of the nanocrystalline diamond foils is probably dominated by graphitic character at grain boundaries. For comparable thickness foils, losses due to the  $\text{H}^0$  yield also appear to be higher for diamond, a phenomenon that is not currently understood. Most recently a  $512 \mu\text{g/cm}^2$  (average) foil was tested at PSR. It survived 239 C of injected charge with increasing beam loss over time until it was removed from service. That charge represents about 70% of the typical service life for a LANL/KEK foil, and is somewhat less than for earlier diamond foils. Foil curling and darkening similar to that observed at SNS occurred for irradiated PSR foils, with the same curling directionality.

### SUMMARY

Nanocrystalline diamond stripping foils continue to perform well at the SNS accumulation ring as the beam power has been ramped up. Current operation is at approximately 35% of the 1.4 MW design power. Foils graphitize and curl upon experiencing more than about 300 C of injected charge. Similar results have been

obtained for diamond stripper foils at PSR. A large-area carbon-carbon composite foil is used as a secondary stripper foil at SNS, but a substantially thinner material that is resistant to pinhole formation is needed. Currently nano-seeding is used for diamond film nucleation. This has replaced the ultrasonic abrasion method and results in more uniform nano-textured films. In addition, we have developed a nondestructive capability for determining foil thickness at the beam injection location.

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