Preparation and Lifetime Study of Diamond-like Carbon Release Films

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Abstract

Self-supporting diamond-like carbon (DLC) stripping films with a thickness of ~5 g/cm2 were prepared using filtered cathodic vacuum arc (FCVA) technology, combined with alternating current carbon arc (ACCA) technology and a relaxation technique. The uniformity and microstructure of the DLC films were characterized and analyzed using an XP2U electronic balance, scanning electron microscope (SEM), Raman spectrometer, and X-ray photoelectron spectroscopy (XPS) instrument. The results show that the prepared DLC films have smooth surfaces, are essentially droplet-free, are typical amorphous DLC films with sp3 hybridized bonds exceeding 70%, and exhibit a maximum non-uniformity of 8.82% within a Φ100 mm range. The lifetime of the DLC stripping films was investigated through testing with a series of heavy ion beams provided by the Beijing HI-13 tandem accelerator. The results demonstrate that the lifetime of relaxed DLC stripping films is approximately 3 times that before relaxation; for 63Cu- and 197Au- ion beams (acceleration voltage ~9 Mv, low-energy end beam current ~1 A), the DLC stripping film lifetimes are 4 times and 13 times those of carbon arc carbon stripping films, respectively; for 107Ag-, 70Ge-, 48Ti-, 28Si-, and 127I- ion beams, the DLC stripping film lifetimes are 2.6 to 10 times those of carbon arc carbon stripping films, and the advantage of DLC stripping film lifetime over carbon arc carbon stripping film lifetime becomes more pronounced with heavier ions and stronger beam currents; the DLC stripping film lifetime exhibits a certain relationship with substrate bias voltage, with current test results showing a general trend of first increasing and then decreasing with increasing substrate bias voltage, and the longest DLC stripping film lifetime is achieved at a substrate bias voltage of approximately -400 V.

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Preamble

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Preparation and Lifetime Study of Diamond-like Carbon Stripper Foils

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Abstract

Self-supporting diamond-like carbon (DLC) stripper foils with a thickness of ~5 g/cm^2 were prepared using a composite technique combining filtered cathodic vacuum arc (FCVA), alternating current carbon arc (ACCA), and relaxation methods. The uniformity and microstructure of the DLC foils were characterized using an XP2U ultra-microbalance, scanning electron microscopy (SEM), Raman spectroscopy, and X-ray photoelectron spectroscopy (XPS). The results demonstrate that the prepared DLC foils exhibit smooth surfaces, are essentially free of droplets, and consist of typical amorphous DLC with sp^3 hybridized bonds exceeding 70%. The maximum non-uniformity across a Φ100 mm area was 8.82%. The lifetimes of the DLC stripper foils were evaluated using various heavy ion beams provided by the Beijing HI-13 tandem accelerator. The findings reveal that the lifetime of relaxed DLC stripper foils is approximately three times that of unrelaxed foils. For 63Cu$^-$ and 197Au$^-$ ion beams (accelerating voltage ~9 MV, low-energy end current ~1 A), the DLC stripper foil lifetimes were 4 times and 13 times longer than those of carbon arc carbon stripper foils, respectively. For 107Ag$^-$, 70Ge$^-$, 48Ti$^-$, 28Si$^-$, and 127I$^-$ ion beams, the DLC stripper foil lifetimes ranged from 2.6 to 10 times those of carbon arc foils, with the advantage becoming more pronounced for heavier ions and higher beam currents. The DLC stripper foil lifetime also depends on the substrate bias voltage, showing a general trend of initially increasing and then decreasing with higher bias voltages, with peak lifetime achieved at approximately -400 V.

Keywords: diamond-like carbon; stripper foil; relaxation; lifetime

Introduction

The Beijing HI-13 Tandem Accelerator National Laboratory is one of China's key nuclear physics research facilities, and its efficient operation is crucial for advancing fundamental nuclear physics research in the country. In recent years, increasing demands from nuclear physics experiments have led to the extraction of more high-intensity, low-energy heavy ion beams from this accelerator, imposing stricter requirements on stripper foil quality and lifetime. Conventional carbon stripper foils prepared by the carbon arc method [1] previously used at the facility exhibit extremely short lifetimes under these conditions. For example, when stripping 197Au$^-$ ions (accelerating voltage 9–11 MV, low-energy end current ~1 A), the lifetime is only 4–8 minutes, whereas a typical experiment requires 100 hours of beam time. The limited number of stripper foils installed in the accelerator, combined with their short lifetimes, directly leads to beam delivery interruptions, shortened maintenance cycles, reduced accelerator operational efficiency, increased operating costs, and decreased experimental productivity.

The development of long-lifetime carbon stripper foils has long been a focal point and challenge in the international target preparation community. Many accelerator laboratories worldwide face similar issues with short stripper foil lifetimes when extracting high-intensity, low-energy heavy ion beams. To address this, researchers have developed numerous carbon stripper foil preparation techniques, including contact spark discharge [2], electron bombardment [3], carbon arc [4–8], ion beam sputtering [9–10], ethylene pyrolysis [11–12], laser ablation [13–15], and DC glow discharge sputtering [16–18], seeking more effective methods and processes for producing long-lifetime foils. Several accelerator laboratories have successfully replaced conventional carbon stripper foils with diamond-like carbon (DLC) stripper foils, achieving significant lifetime improvements [13–18].

This work employs filtered cathodic vacuum arc (FCVA) technology combined with carbon arc and relaxation techniques to prepare self-supporting DLC stripper foils with a thickness of ~5 g/cm^2 for the Beijing HI-13 tandem accelerator. The surface morphology and microstructure of the foils were characterized, and their lifetimes were evaluated using various low-energy heavy ion beams from the Beijing HI-13 tandem accelerator, yielding remarkable results.

1 Experimental Apparatus and Methods

Self-supporting DLC stripper foils with a thickness of ~5 g/cm^2 were prepared for the Beijing HI-13 tandem accelerator using a combination of FCVA, ACCA, and relaxation techniques. Polished mirror-finish stainless steel sheets (1 mm thick) served as deposition substrates, with betaine used as a release agent. The preparation process involved first depositing a ~1 g/cm^2 protective carbon layer via ACCA, followed by deposition of a ~4 g/cm^2 DLC layer using a double 90° bent FCVA system. A thin collodion coating was applied before water release, and the freestanding foils were then transferred to dedicated stripper foil holders for relaxation and drying. The holder aperture diameter was 13 mm, corresponding to an original membrane size of Φ13 mm that was stretched by 2 mm during relaxation.

[Figure 1: see original paper] shows a schematic diagram of the carbon arc apparatus, which was modified from a DM-450A coating system. The device consists of a transmission mechanism, workpiece holder, thickness control system, power supply, and the main DM-450A coating chamber. Rotating the handwheel on the worktable drives the transmission shaft via chains, gears, and bevel gears, causing the sliding frame to move along guide rails and adjust the distance between the carbon electrodes to initiate or extinguish the arc. Selecting an appropriate arc current is critical when preparing protective carbon films by ACCA. Excessive current causes rapid temperature rise in the carbon rods, leading to significant carbon particle splashing and numerous pinholes in the film, while insufficient current prevents stable arc ignition. The relationship between carbon electrode diameter and arc current was investigated and is presented in Table 1 . Based on deposition performance, 6 mm diameter carbon rods and an arc current of 90 A were selected for this work.

Film thickness was monitored in real-time using a quartz crystal thickness monitor. [Figure 2: see original paper] illustrates the FCVA apparatus for DLC film preparation, with its core components being the arc source and double 90° magnetic filter bend. Detailed descriptions of this system are available in references [19–20]. Table 2 lists the optimized process parameters for DLC deposition by FCVA. Film thickness was controlled by deposition time, and [Figure 3: see original paper] shows the relationship between deposition time and DLC film thickness, which exhibits approximately linear growth.

2 Uniformity Testing and Analysis

The uniformity of DLC films was evaluated over an effective coating area of 100 mm × 100 mm using a weighing method with deposited foil samples. The procedure was as follows: (1) Thirty-six stepped holes (13 mm through-hole diameter, 15 mm step diameter, 2 mm spacing) were uniformly machined via CNC in a 100 mm × 100 mm region of a 150 mm × 150 mm × 3 mm aluminum substrate, as shown in [Figure 4: see original paper]; (2) Aluminum foils (~10 m thick) were placed in each stepped hole, and the mass of each foil was measured before and after DLC deposition using an XP2U ultra-microbalance (0.1 g precision) to calculate the mass thickness of each DLC sample; (3) The uniformity across the 36 positions was determined to characterize the DLC film uniformity over the 100 mm × 100 mm area. The measurement data are compiled in Table 3 .

The data indicate that: (1) Under these process conditions, the average DLC film thickness is 6.7 g/cm^2, with a distribution that is thicker in the center and thinner at the periphery, particularly at the four corners; (2) The maximum non-uniformity is 14.9% over the full 100 mm × 100 mm area, which improves to 8.82% when considering only the Φ100 mm region (excluding the four corner samples).

3.1 Surface Morphology

Since the plasma generated by vacuum arc contains macroparticles or microdroplets that significantly degrade film properties, magnetic filters are employed to remove most of the neutral particles. The filtering efficiency of the 90° magnetic filter was evaluated by depositing DLC films on 1 mm thick single-crystal Si wafers under three conditions: with double 90° magnetic filter bends, with a single 90° magnetic filter bend, and without any magnetic filter. The effectiveness in filtering macroparticles was observed by SEM, as shown in [Figure

5: see original paper]. The images reveal that films deposited without magnetic filtering contain numerous carbon particles, those with single 90° filtering contain fewer particles, and those with double 90° filtering exhibit virtually no macroparticle contamination. Consequently, double 90° magnetic filter bends were adopted for DLC stripper foil preparation.

3.2 Raman and XPS Spectroscopic Analysis

Raman spectra were acquired using a Nexus 670 micro-confocal Raman spectrometer, while XPS spectra were obtained with an AXIS ULTRADLD XPS system. [Figure 6: see original paper] presents the visible-light Raman spectrum (wavelength 623.8 nm) (a) and C1s XPS spectrum (b) of a DLC film deposited on silicon. The Raman spectrum Figure 6: see original paper shows an asymmetric broad peak between 1100–1900 cm^{-1}, which can be deconvoluted into two Gaussian components centered at 1360.9 cm^{-1} (D peak) and 1548.6 cm^{-1} (G peak). The G peak is significantly shifted to lower frequency compared to single-crystal graphite, and the integrated intensity ratio I_D/I_G is 0.78, characteristic of typical amorphous DLC films [20,21]. XPS peak fitting analysis using Xps Peakfit software reveals an sp^3 bond content of 71.56%.

4 Lifetime Testing and Analysis of DLC Stripper Foils

Lifetimes were measured using ion beams from the Beijing HI-13 tandem accelerator. Following the internationally accepted definition, stripper foil lifetime is defined as the time from initial beam exposure until the ion beam current at the image point decreases to 50% of its initial value, under constant low-energy end injection current [17]. [Figure 7: see original paper] shows a schematic of the Beijing HI-13 tandem accelerator used for lifetime measurements. The stripper foils tested in this study were installed in the first stripper position between the fourth and fifth accelerating tubes, with beam current measured at the image point indicated in the diagram.

The first test series evaluated the lifetime of DLC stripper foils before and after relaxation using a 197Au⁻ ion beam (accelerating voltage ~9 MV, low-energy end current ~1 A). The results, shown in [Figure 8: see original paper], demonstrate that the lifetime of relaxed DLC stripper foils is approximately three times that of unrelaxed foils.

The second test series compared the lifetimes of DLC stripper foils and carbon arc carbon stripper foils using 63Cu⁻ and 197Au⁻ ion beams at 9 MV accelerating voltage and 1 A low-energy end current. The results, presented in [Figure 9: see original paper], show that DLC stripper foil lifetimes are 4 times and 13 times longer than those of carbon arc foils for 63Cu⁻ and 197Au⁻ beams, respectively.

The third test series measured and compared lifetimes using six representative ion beams: 107Ag⁻, 70Ge⁻, 48Ti⁻, 28Si⁻, 197Au⁻, and 127I⁻ (accelerating voltage and low-energy end current detailed in Table 4). Under constant accelerating voltage and beam current, the lifetime results are summarized in Table 4. The data indicate that DLC stripper foil lifetimes are 2.6–10 times those of carbon arc foils, with the difference becoming more pronounced for heavier ions and higher beam currents.

The fourth test series investigated the effect of substrate bias voltage on DLC stripper foil lifetime using a 197Au$^-$ ion beam (accelerating voltage ~9 MV, low-energy end current ~0.25 A). The results, shown in [Figure 10: see original paper], reveal that DLC stripper foil lifetime is related to substrate bias voltage (carbon ion energy), exhibiting a general trend of initially increasing and then decreasing with higher bias voltage. Maximum lifetime is achieved at approximately -400 V substrate bias, which explains why -400 V was selected as the optimal bias voltage in our process parameters. It should be noted that due to limited accelerator beam time, only one foil was tested for each bias condition, introducing statistical uncertainty. Therefore, these results represent a reference trend rather than definitive quantitative values.

A common phenomenon observed during lifetime testing is an initial increase in image point current followed by gradual decline until foil failure. The initial current rise is attributed to two factors: (1) the collodion coating applied during foil preparation is gradually removed by beam heating, improving beam quality, and (2) the heating process repairs microscopic defects in the carbon film. Subsequent current decline results from radiation damage to the carbon film as energy deposition reaches critical levels.

Summary

Using a composite technique of "filtered cathodic vacuum arc + alternating current carbon arc + relaxation," self-supporting DLC stripper foils with ~5 g/cm^2 thickness were successfully developed for the Beijing HI-13 tandem accelerator. Characterization via XP2U ultra-microbalance, SEM, Raman spectroscopy, and XPS demonstrated that DLC films prepared with double 90° filtering exhibit smooth surfaces with virtually no macroparticle contamination, typical amorphous DLC structure with I_D/I_G = 0.78, sp^3 bond content exceeding 70%, and maximum non-uniformity of 8.82% across a Φ100 mm area. Lifetime testing at the Beijing HI-13 tandem accelerator showed that for 197Au$^-$ ion beams (accelerating voltage 9–11 MV, low-energy end current ~1 A), relaxed DLC stripper foils achieved lifetimes approximately three times those of unrelaxed foils. Compared to carbon arc carbon stripper foils, DLC foils exhibited 4-fold and 13-fold longer lifetimes for 63Cu$^-$ and 197Au$^-$ beams (accelerating voltage 9–11 MV, low-energy end current ~1 A), respectively, and 2.6–10-fold longer lifetimes for 107Ag$^-$, 70Ge$^-$, 48Ti$^-$, 28Si$^-$, and 127I$^-$ beams, with the advantage increasing for heavier ions and higher beam currents. DLC stripper foil lifetime shows a dependence on substrate bias voltage, peaking at approximately -400 V. These developed DLC stripper foils have completely replaced conventional carbon arc foils at the Beijing HI-13 tandem accelerator, significantly extending stripper foil lifetime, reducing accelerator maintenance cycles, improving operational efficiency, and lowering operating costs.

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Note: Figure translations are in progress. See original paper for figures.