The irradiation study of carbon stripper foils by a 12 MeV Ar beam

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Abstract. Irradiation performance of multilayer HBC foils which layers are more than 20 were conducted for the first time, and different types of carbon stripper foils were dosed under a series of irradiation conditions systematically. Eight (8) types of carbon stripper foils were struck by Ar4+ beam under different intensities and fluences. Multilayer foils performed generally better than the monolayer ones. The existence of boron layer helped to increase the resistance of the foils to irradiation damage, elongating their lifetimes.

1 Introduction

Carbon stripper foils belong to one of the dominant types of stripper foils and have been studied for decades [1]-[5]. Diamond like carbon (DLC) and hybrid boron doped carbon (HBC) foils are two popular kinds among all carbon stripper foils. Multilayer HBC foils have been considered for their long lifetime, and attracted much interest [6-10]. The preparation method of the HBC foils has been the subject of many studies, yet the relationship between the characteristics of the layers and the lifetime of the foils has not yet been systematically studied. The irradiation performance of foils with more than 20 layers has not yet been reported. For such foils with thickness around 2μm or less, where each layer is less than 100nm thick, some nanoscale effects [11-12] are expected to take place. In this paper, 8 types of carbon stripper foils, including foils of 48 layers and 100 layers, were irradiated with a Ar4+ beam of 12 MeV under different intensities and fluences. Multilayer foils performed generally better than the monolayer ones. The existence of boron layer helped to increase the resistance of the foils to irradiation damage, elongating their lifetimes.

2 Materials

The stripper foils used in this study were all supplied by UHV Technologies, Inc. There were totally eight types of foils: 2 types of monolayer DLC foils, 10 wt% boron mix 90 wt% carbon monolayer foil, and 7 layers, 9 layers, 17 layers, 48 layers, 100 layers HBC foils. The layout of the foils is listed in Table 1, and each type of foil was assigned a number, for the convenience of discussion. The index of the foils is showing in Table 1.

Table 1. Types of the foils. The nominal thicknesses were between 2-2.5μm (around 500 μg/cm²), except the 10% boron mix 90% carbon monolayer foil, which thickness is 1.6μm.

| Num. | Foil type/layout | Composition |
|------|-----------------|-------------|
| 1    | One layer DLC, type 3 | Amorphous carbon |
| 2    | One layer DLC, type 1 | Amorphous carbon |
| 3    | 7 layers HBC, 4B:3C | 6% B + 94% C |
| 4    | 9 layers HBC, 5B:4C | 20% B + 80% C |
| 5    | 17 layers HBC, 9B:8C | 20% B + 80% C |
| 6    | 48 layers HBC, 24B:24C | 83% B +17% C |
| 7    | 100 layers HBC, 50B:50C | 83% B + 17% C |
| 8    | One layer mixture | 10% B+90% C |

3 Experiment

All the foils were mounted on brass frames, glued by either Aquadag® (graphite coating) or double-sided graphite tape. The effective irradiation area of the foils is 6.35×5.84 mm². The frames were further mounted on copper plates to be irradiated. Each plate bears 4 different foils, that can be irradiated under the same condition. Thus the 8 types of foils were mounted on 2 plates for each run’s condition. The plates were then loaded on a target holder with water-cooling system. See Fig. 1. The spot size was adjusted to be large enough to cover all 4 foils and it was re-defined each time the beam intensity was changed, or after several rounds had been done.

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Fig. 1. Sample assembly. (a) Samples loaded on holder. Water goes through the bottom flange, behind the copper plate; (b) Irradiation line. The 2 plastic tubes belong to water cooling system.

Before irradiation, plastic sheets were irradiated to confirm the beam size. The beam size was re-measured every time the beam intensity was changed, or after several runs had been done. Alpha particle attenuation was used to determine the thickness of the foils. The alpha source used was Am-241, 3μCi.

4 Irradiation of the foils

Ar⁴⁺ beam had an energy of 12 MeV. Multiple beam intensities and fluences were used. The initial intensity we aimed at was 10μAe for Ar⁴⁺. Yet to prevent extreme damage of the foils before reaching higher fluence, we had to use lower intensities. One example of the extremely damaged samples after irradiation is shown in Fig. 2.

Pictures were taken each time after irradiation, the damage of all the foils tested is evaluated based on the pictures. The damages were classified to 6 grades:
- 0-No damage;
- 1-No visible cracks or peelings, but uneven surface appeared;
- 2-Ralatively even surface, with visible cracks or peelings;
- 3-Local deformation, with small scale cracks or peelings;
- 4-Badly damaged, with severe cracks or peelings (may accompany with large deformation, depends on foils);
- 6-Foil failed and removed.

Three variables were used for evaluation: intensity, fluence and the type of foil. Upon a correlation analysis, we showed that the beam intensity is a less significant factor with respect to the foils’ damage. Therefore, the average damage under certain magnitudes of fluence were taken for each type of foil regardless of beam intensity. The result is shown in Fig. 3.

Fig. 2. Foils before and after irradiation. (a) (from left to right, up to bottom) 9 layers HBC, 7 layers HBC, DLC type1, DLC type3 foils before irradiation; (b) 9 layers HBC, 7 layers HBC, DLC type1, DLC type3 foils after irradiation; (c) (from left to right, up to bottom) 17 layers HBC, 48 layers HBC, 100 layers HBC, B+C mixture foils before irradiation; (d) 17 layers HBC, 48 layers HBC, 100 layers HBC, B+C mixture foils after irradiation.

Fig. 3. The damage degree of 8 types of foils under different magnitudes of ion fluence.

It can be seen that below 10¹³ ions/cm², all the foils are “safe”, but above this fluence, DLC foils and B/C mixture foil become heavily damaged (average grade >4) and failures start to appear above 10¹⁴ ions/cm².
Specifically, two sets of samples are compared: one DLC foil, one HBC foils, and the B/C mixture; and foils of various number of layers (See Fig. 4 (a) and (b) respectively). The error bars correspond to the deviation of foils struck by beams with different intensities. From Fig. 4. (a), it can be observed that DLC foil and B/C mixture foil became damaged rapidly above 10^{13} ions/cm², and are both near to failure at 10^{14} ions/cm². On the contrary, 100 layers HBC foil remained relatively undamaged up to 10^{17} ions/cm². From Fig. 4 (b), one can find that below 10^{14} ions/cm², the performance of all the foils is similar, and starts to deviate after 10^{14} ions/cm². Despite no obvious relationship between damage degree and the number of layers, it is not until 10^{16} ions/cm² when some foils start to fail. Surprisingly, 17 layers HBC behaved almost the same as 100 layers one.

5 Swelling study of the irradiated foils

To quantify the irradiation performance of the foils, one method was to characterize the swelling of the foils. This parameter can be deduced from the thickness of the samples using equation (1).

\[
\text{Swelling} = \frac{(\text{thickness of the irradiated samples} - \text{thickness of the unirradiated sample})}{\text{thickness of the unirradiated sample}} \times 100\% \quad (1)
\]

The thickness of the foils was determined by the Energy loss of Alpha-particles method using the 5480 keV alpha energy of an Am-241 source detected by a Si detector with a full width at half maximum (FWHM) of 47 keV.

Stopping and range of ions in matter (SRIM) was used to evaluate the thickness of all sorts of foils. For the α particles of Am-241 in carbon foils, \( dE/dx = 783.27 \text{ keV/mg/cm}^2 \) considering a density of the foils as 2.25 g/cm³.

100 layers HBC and DLC (type 1) foils irradiated under different conditions were measured. The analysis of the measurements is found in table 2 and table 3 respectively where thickness (in μm) and swelling (in %) are given. The result indicates that the thickness of foils irradiated under different conditions are not much changed.

| Irradiation fluence (ions/cm²) | Irradiation intensity (μAe) | Thickness evaluated by SRIM (μg/cm²) | Thickness (μm) | Swelling (%) |
|-----------------------------|---------------------------|--------------------------------------|----------------|--------------|
| 7 layers HBC                | 0                         | 261.03                               | 1.16           |              |
| 9 layers HBC                | 0.23                      | 267.34                               | 1.19           | 2.29         |
| 17 layers HBC               | 1.4                       | 263.83                               | 1.17           | 0.95         |
| 10 layers HBC               | 1.4                       | 252.12                               | 1.12           | -3.53        |
| 100 layers HBC              | 1.4                       | 267.87                               | 1.19           | 2.49         |
| 17 layers HBC               | 1.4                       | 259.19                               | 1.15           | -0.83        |
| 10 layers HBC               | 1.4                       | 269.01                               | 1.19           | 2.93         |
| 100 layers HBC              | 4                         | 262.87                               | 1.17           | 0.58         |
| 10 layers HBC               | 10                        | 267.52                               | 1.19           | 2.36         |
| 100 layers HBC              | 10                        | 259.25                               | 1.15           | -0.80        |

5 Discussion

As discussed in section 4, the multilayer HBC foils generally had a longer lifetime than DLC foils. The B/C mixture foils performed worst among HBC foils, even worse than DLC foils sometimes. The 100 layers HBC foil had the best resistance toward irradiation, yet the lifetime of other multilayer HBC foils did not show a regular trend with increasing number of layers as expected. In many cases, the 7 and 17 layers foils had longer lifetime than 48 layers ones. This is particularly eminent for 17 layers foils, which behave almost identical to the 100 layers ones.
The 100 layers foils performed best among all the tested foils, also with respect to their structural steadiness. When radiation damage appears, most of the other multilayer foils exhibit layer debonding, while the layers of 100 layers foils, despite the top layer peels sometimes, stayed essentially undamaged even under high intensities or high fluences. A comparison between the 7 and 100 layers is shown in Fig. 5. The pictures were taken by a microscope.

These observations made it reasonable to believe that there is a connection between the firm layer structure of 100 layers foils and their long lifetime. All the multilayer foils were produced by the same technique, Pulse-Laser Deposition. Therefore, it is less possible to be the processing which made the difference. We tried to explore the reason for the outstanding performance of 100 layers foils from the microstructure’s side. Table 2 indicates that the total thickness of this type of foil is around 1 μm, such that the thickness of each layer is about 10 nm. The carbon layers of all the multilayer HBC foils used were supposed to be consisted of DLC structure. It was suggested that upon irradiation, the sp3 bond of DLC tend to transform to sp2 bonds, possibly forming nanographite structures [11, 12]. The displacement of carbon atoms due to energetic particles, heavy ions here particularly, can subsequently occur. As a result, interstitial defects will cause crystallite growth perpendicular to the layer planes, whereas coalescence of vacancies will cause a shrinkage parallel to the layer planes [13]. This crystallite dimensional changes leads to the generation of new porosity [13], which has the potential to become the source of cracks. This can explain the initiation of the cracks in the foils in Fig. 2. The interstitial defects, one form of Frenkel defects, and crystallite distortion discussed above are also known as “Wigner Effect”, which is an important issue of graphite when subjected to high energy irradiation[14], resulting in energy storing through lattice swelling. This energy storage to nanodiamond is even higher [15]. At high temperature (>600°C), the defect can be annealed, while below 600°C the distortion of lattice grows as the temperature increases [14]. Without temperature controlling system, the temperature within our samples during irradiation rose as the irradiation intensity increased. The highest temperature (at 10 μAe) was estimated to be 670°C, when the highest degree of lattice distortion occurs-- further explains the derivation of cracks. In terms of multilayer structure, it is reasonable to believe that boron atoms can act as interstitials which diffuse into diamond lattice and recombine with vacancies, forming a stable interface tolerant to irradiation. This may be similar to the promotion of radiation tolerance in metal/metal and metal/oxide interfaces due to the point defects (interstitials, vacancies) recombination [15]. This hypothesis may explain the prominent performance of 100 layers HBC foils. With the increase of thickness of carbon layer in fewer layers foils, the Wigner Effect is expected to be dominant, and the resistance to irradiation declines. Even so, the multilayer foils still have longer lifetime than monolayer ones, including B/C mixture foil, proving that boron layers increase the resistance of the DLC layers toward irradiation.

According to section 5, it is easy to find that the thickness of 100 layers HBC foils irradiated under different conditions did not change too much. Considering the uncertainty, the thickness of DLC foils didn’t change too much either, despite a decreasing tendency seemed to appear for DLC foils at higher fluences, see figures 6 and 7.

![Fig. 5. A view of the border of 7 layers and 100 layers HBC foils after an irradiation of a fluence of 7*10^{14} ions/cm² at 1.4 μAe.](image)

![Fig. 6. Swelling of 100 layers HBC and DLC foils irradiated under 0.23 (N/A for DLC foils), 1.4, 10 μAe intensities and the same fluence of 1.40*10^{15} ions/cm².](image)

![Fig. 7. Swelling of 100 layers HBC foils irradiated under 1.4 μAe intensity and the increasing fluence.](image)
the expansion of the surface area is accompanied by the attenuation in thickness, when the bulk density remains the same.

The expansion of the surface area of the foils is majorly caused by thermal effect [16, 17]. On the other hand, the thickness could reduce due to the “ion hammering effect” [18], which has an outcome of swelling. Yet the reduction in measured thickness does not always indicate swelling. It is very possibly due to the sputtering of surface atoms or the cracks on the foil as well. Therefore, thermal expansion and “ion hammering effect” are not enough to explain the thickness change of the foils. The thickness measurements give an effective thickness, which is a combination of the “true thickness” with the angle of the ripples (deformation), especially for irradiated DLC foils. This argument is also indicated by the high uncertainty of the “swelling” percentage.

Despite the complication discussed above, it is still possible to tell by comparing the alpha particle test results of 100 layers HBC foil and DLC foil that multilayer foils maintain better their sizes than DLC foils and are more resistant to irradiation. This result is coherent with the conclusion made earlier.

6 Conclusion

In the present study, irradiation performance of multilayer HBC foils with more than 20 layers were observed for the first time, and different types of carbon stripper foils were systematically irradiated under a series of irradiation conditions. 8 different types of carbon stripper foils were irradiated using the Ar⁺12 MeV beam from the 5U accelerator of the University of Notre Dame. It was found that below 10¹³ ions/cm², regardless of beam intensity, all the foils are “safe”. Then non-multilayer foils start to show heavy damage and eventually fail. Among all the foils, 100 layer HBC foils performed best upon irradiation, not only having longest lifetime (10¹⁷ ions/cm² under 10 μAe), but also having the most steady multilayer structure. This is believed to be the reason for the better radiation resistance. Multilayer foils performed generally better than the monolayer ones. Boron layer may help to increase the resistance of the foils to irradiation damage, elongating their lifetimes. The swelling of the irradiated DLC type I foil and 100 layers HBC foil were measured via alpha particle attenuation. Although the “swelling” here is caused by complicated factors, it can still be concluded that the 100 layers HBC foils is relatively stable in size upon irradiation, and the swelling of DLC foils decrease with the increasing of fluence at 1.4 μAe intensity.

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