 lifetime improvement of nitrided carbon stripper foils by
ion-beam sputtering with a binary gas mixture
I. Sugai,a,* Y. Takeda,a M. Oyaizu,a H. Kawakami,a Y. Hattori,b K. Kawasaki,b
K. Yoshidac, A. Itohc

a High Energy Accelerator Research Organization, Oho 1-1, Tsukuba-shi, Ibaraki-ken 305-0801, Japan
b Tokyo Institute of Technology, Ookayama, Meguro, Tokyo 152-8550, Japan
c Quantum Science and Engineering Center, Kyoto University, Gokasho, Uji, Kyoto 611-0011, Japan

Abstract

The lifetimes of our carbon stripper foils (10–25 μg/cm²) prepared by reactive nitrogen ion-beam sputtering were
distributed in a wide range. Expressed in units of integrated beam current received, lifetimes of 750 ± 400 mC/cm² were
usual for a 3.2 MeV Ne⁺ beam. This wide distribution of lifetimes have been improved by applying a binary gas
mixture (N₂ + Ne) for sputtering of carbon. With high reproducibility, lifetimes of 475 ± 80 mC/cm², approximately 15
times those of the best standard commercially available foils, have been achieved. We have also investigated the
influence light-element admixtures had on the lifetimes of carbon stripper foils.

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1. Introduction

In a previous paper [1], we reported that nitrided
carbon foils made by the Ion Beam Sputtering
with Reactive Nitrogen (IBSRN) method had
extremely long lifetimes when compared to hydro-
genated, oxidized carbon foils or the best commer-
cially available standard foils. As shown in Fig. 4
of Ref. [1], however, the beam lifetime of
individual foils varied by a factor of three with a
maximum exposure of 2300 mC/cm² for the
3.2 MeV Ne⁺ beam. In order to prepare nitrided

*Corresponding author. Fax: +81-298-64-5576.
E-mail address: isao.sugai@kek.jp (I. Sugai).
gases such as Kr and Xe also introduced contaminants because they sputtered more effectively the materials used to construct the ion source.

Because we had previously obtained long lifetimes from foils made with a Kr + Ne gas mixture [3,4], in the present work we conducted additional trials with mixtures of Ne and other gases. Also investigated were the lifetime effects of light-element additions to the carbon foils, for example: hydrogen, oxygen, enriched isotopic $^{15}$N$_2$, air and metallic Be, and B and Al.

2. Experimental

Carbon foils were prepared with the focused ion-beam sputtering (IBS) system, as described in Ref. [2]. The typical operating parameters are listed in Table 1. The size of the carbon rod sputtered was 13 mm diameter by 2 mm thick. The impurity content of the rod was less than 20 ppm. During sputtering, the rod was atop a graphite beam stopper. The sputtered carbon was deposited onto three glass slides (26 × 32 mm$^2$) coated with CsI releasing agent by vacuum evaporation–condensation. These substrates were placed 5 cm from the graphite source and at an angle of 45° to the primary beam direction. A rotating substrate holder was used to improve the thickness uniformity of the deposited films. A thin Al foil served as a thickness monitor positioned near the substrates and weighed with an electronic micro-balance.

The reactive gases used were purified. In the case of nitrogen and neon gases, the impurity levels were less than 0.2 ppm of oxygen and 1 ppm of carbon dioxide and, overall purity of 99.999%. The flow rates of the binary gas mixtures were about 1 ml/min for Ne and 5 ml/min for H$_2$, N$_2$, $^{15}$N$_2$, O$_2$ and air. Silica gel powder was used to dehydrate the air when air was mixed with $^{15}$N$_2$ gas of 98.55% enrichment.

The pressed-carbon sputtering disks were prepared by hand mixing graphite powder (of < 20 ppm total impurities) with Be, B and Al powders of 99.99% purity. The mixtures produced were C with a 10% B content, C with a 10% Be content and C with 20% Al. The mixed powder was formed by compression into 13 mm diameter by 2 mm thick disks, as used previously.

All prepared carbon stripper foils had a thickness of about 10–25 μg/cm$^2$, which were measured with an electronic micro-balance. The size of the foils was 15 × 15 mm$^2$.

The lifetime measurements were carried out using 3.2 MeV Ne$^+$ beams of 3–4 μA distributed in a beam spot of 3.5 mm diameter at the Van de Graaff accelerator of Tokyo Institute of Technology. The lifetime (mC/cm$^2$) was defined as integrated transmitted Ne$^+$ current density upon the foils until a rupture was indicated by a sudden decrease of the current in a Faraday cup located downstream from the foil. The setup is described in detail in Fig. 1 of Ref. [4]. The total pressure in the scattering chamber for the lifetime measurement

![Fig. 1. Comparison of the relative lifetime of carbon stripper foils prepared by focused ion beam sputtering applying different mixtures of sputter gas. The lifetimes were measured by using a 3.2 MeV Ne$^+$ beam of 3–4 μA with a 3.5 mm diameter beam spot. The foil thickness was 10–25 μg/cm$^2$. The average integrated incident current until rupture is plotted in units of mC. The numbers of samples investigated are given in parentheses.]

<table>
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<th>Table 1</th>
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<td>Typical run parameters for this ion-beam sputtering system</td>
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<td>Ion species binary mixture gas: N$_2$ + Ne, H$_2$ + Ne, Ne + O$_2$, $^{15}$N$_2$ + Ne, air + Ne</td>
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<td>Ion current: 0.5–2.0 mA</td>
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<tr>
<td>Source gas: &gt; 99.99% purity</td>
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<td>Indicated pressure: $1 \times 10^{-4}$ Pa</td>
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<tr>
<td>Ta filament current (1.5-mm $\varnothing$ wire): 40–80 A at 3 V</td>
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<tr>
<td>Extraction voltage: 10–13 kV</td>
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<td>Beam spot: 2–4 mm $\varnothing$</td>
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was maintained at about $2 \times 10^{-4}$ Pa total pressure. The partial pressures recorded for other gases were $H_2 : 8.6 \times 10^{-7}$ Pa; $H_2O : 6.1 \times 10^{-5}$ Pa; $N_2 + CO : 9.7 \times 10^{-5}$ Pa; $O_2 : 2.2 \times 10^{-5}$ Pa; $Ar : 1.3 \times 10^{-6}$ Pa; $CO_2 : 2.7 \times 10^{-7}$ Pa.

Contamination of light elements such as hydrogen, nitrogen and oxygen in the foils were quantified by the Rutherford Back Scattering (RBS) method. For this, a $He^{2+}$ beam from the Van de Graaff accelerator at Kyoto University was used. Inspections of the foils were made with a camera during ion beam bombardment.

3. Results and discussion

3.1. Lifetime measurements

Fig. 1 shows the results of the lifetime measurements of carbon stripper foils produced by sputtering with binary-mixture gases. In addition, the lifetimes of different foil types were measured for comparison:

(a) foils made by the IBSRN method as described in Ref. [1],
(b) foils prepared by sputtering with pure Ne gas,
(c) foils prepared by sputtering with a gas mixture of $H_2 + Ne$,
(d) foils prepared by sputtering with a gas mixture of $O_2 + Ne$,
(e) commercially available carbon foils (CM).

These results are averaged values from several data sets. The number of foils measured is given in parentheses. The foils made by the binary mixture gas of $(N_2 + Ne)$ show a promising long lifetime of 475 mC/cm$^2$ on average with $780 \pm 70$ mC/cm$^2$ in root-mean-square variation. This value corresponds to 15-times longer, on average, than for the CM foils. Although the average lifetime 475 mC/cm$^2$ is shorter than that of the one for $N_2$ (IBSRN) foils, it is longer than that of foils produced in pure Ne. The difference of the lifetime seems to be due to the neon addition. As obvious in Fig. 1, the lifetime spread of the foils prepared by the binary gas mixtures was improved by about 25%, compared to the foils made by the IBSRN.

Fig. 2. Thickness decrease of a nitrided carbon foil made by ion beam sputtering with the binary gas mixture $N_2+Ne$ during irradiation with 3.2 MeV $Ne^+$ ions with 3–4 $\mu$A beam current and a 3.5-mm-diameter beam spot. The thickness decreases gradually with beam dose. The thickness reduction corresponds to approximately one-half of the initial thickness.

The admixtures of oxygen and hydrogen in the sputtering gas, and the additions of Be, B and Al elements, resulted in films with lower brittleness and fragility. We thus infer that, for carbon stripper foils, the bonding between carbon and nitrogen atoms is probably stronger and more stable than the bonds to hydrogen and oxygen atoms.

The thickness change of the nitrided foils made with the present binary-gas mixture IBM technique was measured during the beam irradiation. During the long-term irradiations, the transmitted beam current through the foils was stable at about 9 ± 0.5 $\mu$A. Fig. 2 shows the average thickness as a function of the beam current density measured in mC/cm$^2$. The thickness was estimated from the number of elastically scattered $Ne^+$ ions detected with a Si detector located downstream of the carbon foils. With irradiation time, the thickness of a carbon foil gradually decreased until the foil ruptured in the irradiated area. This occurred at a thickness of about 9.5 $\mu$g/cm$^2$, corresponding to an ablation of 8.5 $\mu$g/cm$^2$ or 47% of the initial thickness.

3.2. Contaminant measurements by RBS

Impurities in the foils were quantified by the RBS method. For this, a $He^{2+}$ beam from the Van
de Graaff accelerator at Kyoto University was used.

Fig. 3(a) through (h) illustrate the RBS spectra from the foils made with the various types of gases mixed in Ne sputtering gas and the spectra from foils made with the metallic-element admixtures. Fig. 3(a) shows the spectrum from the foil made by the binary mixed gas N₂+Ne. The nitrogen element line is clearly observed and small amounts of oxygen, sodium and tantalum are also detected. The thicknesses of carbon and nitrogen are 12.4 and 1.3 μg/cm², respectively. The contaminant thicknesses were estimated to be 0.18 and 0.24 μg/cm² for oxygen and tantalum, respectively. We suppose the sodium is from the release agent and tantalum is from the thermal filament. Fig. 3(b) shows the spectrum of foils made with the enriched ¹⁵N₂ gas added to the Ne. The isotopic nitrogen element ¹⁵N₂ is clearly seen in the spectrum and the composite thicknesses of the elements were estimated, respectively, to be 15.0, 0.8 and 4.0 μg/cm² for ¹²C, ¹⁴N and ¹⁵N. This result suggests that this method might be applicable to the preparation of an enriched solid nitrogen target for use in nuclear physics experiments, provided the ¹²C content would not be problematic. Fig. 3(d) shows the RBS spectrum from the foil made by an addition of atmospheric air during sputtering. To dehydrate the gas, the air was passed through phosphoric anhydride (P₂O₅) and silica gel. In accordance with their abundance in air, the foils contain much nitrogen and a small amount of oxygen.

The spectra of Fig. 3(e), (f) and (g) are for metallic Be, B and Al, respectively admixed to the carbon powder. The associated foils include large amounts of oxygen, which might come from the surface-oxide layers of the Be, B and Al powders. In Fig. 3(f), however, there is no peak from B. We suppose this is due to the low sputtering yield for boron. Fig. 3(h) shows the typical RBS spectra from the CM foil made by the Arizona Carbon Foil Company. The foil contains only small amounts of oxygen but nearly no other impurities.

4. Conclusion

Foil production with good reproducibility of long lifetimes has been achieved by using a binary mixed gas of N₂+Ne. The variation of foil lifetimes with this method improved significantly. On average, these lifetimes correspond to 15 times those of the CM foils. We have also investigated the influence of light-element admixtures on the foil lifetimes and have found that these elements did not play a positive role. From the present work, the most important conclusion for production of long-lived carbon stripper foils is that one...
should reduce the amount of the hydrogen and oxygen contaminants as much as possible.

References


