JAERI-Conf 2001-002

ICANS-XV

15th Meeting of the International Collaboration on Advanced Neutron Sources November 6-9, 2000 Tsukuba, Japan

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Development of Long-Lived Cluster and Hybrid Carbon Stripper Foils for High Energy, High Intensity Ion Beams

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Abstract

We have developed thin and thick long-lived carbon stripper foils for high energy, high intensity ion beams. The foil thicknesses are about 10 μ g/cm² (cluster foil) and 200 μ g/cm² (hybrid foil) for thin and thick, respectively. The thin foil is made by a controlled DC-arc discharge (CDAD) method, by using the size effect of the carbon particles. The size effect was the difference between the carbon particle sizes emitted from the cathode and the anode electrodes in the DC arc discharge, in which the particle size from the cathode is large (0.3 μ m ϕ) and the other is small (0.003 μ m ϕ). The thin foils composed of large particle size are not mechanically strong, but long-lived under low energy ion bombardment with a 3.2 MeV, 2-3 μ A Ne⁺ beam. The mean lifetime is 900 mC/cm² in average which corresponds to 25 times longer than that of commercially available standard foils. In this method, the key point in producing long-lived foils is to control the amount of carbon particles ablated from the cathode by adjusting temperature at the cathode emission spot.

The thick hybrid carbon foils (multi-layer thickness about 200 $\mu g/cm^2$) have been developed for use in 800 MeV, H+ ion beam at the Proton Storage Ring (PSR) of Los Alamos National Laboratory. The thick foils are prepared by means of the controlled ACDC arc discharge (CADAD) method, and are mechanically strong. The lifetime measurements of thick foils made by various methods were carried out using 800 MeV, 85-100 μ A proton beams in the PSR. The foils made by the CADAD method showed very long lifetime, compared to other foils tested.

1. Introduction

With great improvements of ion sources in accelerators, carbon stripper foils have become very important for heavy ion or H- ion beams with high intensity for efficient acceleration. However, if the lifetime of the carbon foils is very short, then the operational efficiency of the accelerator becomes low and also stripper foil replacement at high energy irradiation such as 800 MeV H- ion beam is one of most dose-intensive activities that maintenance personal are concerned with. Carbon stripper foils with longer lifetime than that of commercially available (CM) foils are, thus, indispensable to high intensity heavy ion accelerators. There are many preparation methods to produce the carbon stripper foils [1-7] as shown in Table 1. First of all, we aim to produce long-lived carbon foils by means of a simple method without any special handling such as the slackening technique [8,9] and the substrate treatment techniques[10]. We have investigated the relation between the lifetime and the preparation

method listed in Table 1. For this purpose, we have manufactured and assembled equipments, in house with the except of the laser (cw and pulse) and the electron beam gun listed in Table 1 [11]. The best preparation method has been found by considening lifetime, reproducibility, mechanical strength, smooth surface, pin holes, limitation of the foil thickness and area. The foils with the best combination of these properties were serected. Lifetime measurements of these foils made by various method were performed using a 3.2 MeV, Ne+ ion beam. Through experimental investigation, we have successfully developed the following three methods, as listed in Table 2, for preparing of long-lived carbon foils: 1) CDAD and CADAD methods (cluster and hybrid foils), 2) Ion Beam sputtering MIBS and HIBS methods (sputter foil) and 3) Ion beam sputtering with reactive nitrogen (IBSRN) method (nitrided carbon foil). The micro structures of the foils were investigated by means of an electron microscope, and the thickness of irradiated area was compared with that of nonirradiated areas [13,17].

By a systematic study of the present work, we found reliable and reproducible conditions to

prepare not only long-lived, but also mechanically strong carbon stripper foils.

In this paper, we present the preparation methods and the lifetime of carbon stripper foils made by means of the CDAD and the CADAD methods. The lifetime measurements were performed with a low energy a 3.2 MeV Ne⁺ ion beams for the thin cluster and thick hybrid carbon stripper foils and with a high energy an 800 MeV, H⁻ ion beam for the thick hybrid carbon stripper foils.

Table 1. Present available preparation methods of carbon stripper foil in the world:

Electron Beam Gun AC arc Discharge DC arc Discharge Ohmic Heating CW Laser Ablation	Thermal Evaporation-Condensation (Hot Temperature)
Magnetron Sputtering Focused Beam Sputtering	Sputtering Evaporation (Cold Temperature)
Glow Discharge	Decomposition of Gases
Pulsed Laser Ablation	Combined Evaporation (Hot and Cold)

Table 2. Preparation methods of long-lived carbon stripper foil successfully developed:

Controlled DC Arc Discharge (CDAD) method	Cluster Carbon Foil (5-50 µg/cm ²)
Controlled ACDC Arc Discharge (CADAD) method	Hybrid Carbon Foil (50-130 μg/cm ²)
Mixed Ion Beam Sputtering (MIBS) Heavy Ion Beam Sputtering (HIBS)	Sputter Carbon Foil (5-50 μg/cm ²)
Ion Beam Sputtering with Reactive Nitrogen (IBSRN)	Nitrided Carbon Foil (10-40 μg/cm ²)

2. Experiment

2.1 Thin foil preparation

The complete set-up for preparing the carbon foils was installed into the same vacuum deposition equipment used previously for the CDAD method. [12]

The arc-discharge system for the CADAD method consists of two arc-discharge evaporation sources. One is a DC arc-discharge unit and the other is an AC arc-discharge unit. In this experiment concerning the cluster carbon foil preparation, we only used the DC arc discharge

unit consisting of a 15 mm diameter carbon rod as anode, and a 10 mm diameter cathode carbon rod. The cathode ablates about 0.3 μ m large particles, on the other hand, the anode ablates about 0.003 μ m fine particles. We call this process it the controlled DC arc-discharge (CDAD) method. The DC arc-discharge power supply (20V x 300A) was operated at 20V, 300 A for evaporation, until the desired deposition thickness was achieved. A crystal thickness monitor monitored the deposition thickness. The vacuum in the evaporation chamber was 1.3 x 10^{-4} Pa without the arc-discharge evaporation and 1.3×10^{-3} Pa with the arc-discharge one.

The carbon layers were deposited onto substrates coated with a releasing agent of Cream-Coat (manufactured by J. Varley and Sons, St. Louis, Missouri).

Figure.1 shows schematic drawings of the present method. Figure.2 shows the TEM (transmission electron micrograph) images of the foils produced by the present CDAD method, and Fig.2 (a) shows the structure of a foil composed of particles evaporated almost from the anode electrode only and Fig.2 (b) shows the structure of a foil composed almost completely of particles from the cathode electrode. The experimental apparatus for the lifetime measurement of the thin cluster carbon foils is almost the same as mentioned in the previous system [17]. The lifetime of the foils was performed with a 3.2 MeV Ne⁺ ion beam of 2-3 µA and beam spot size of 3.5 mm in diameter.

The lifetime is defined as integrated ion current per cm² until carbon foil rupturing occurs. Figure.3 shows the histograms of the foils (10 - 26 µg/cm²) produced by various preparation

rigure.3 shows the histograms of the foils (10 - 26 μ g/cm²) produced by various preparation methods. In Fig.3, the lifetime of Lp-foil made by laser plasma ablation deposition is attached as a reference because this foil of very thin 3 - 4 μ g/cm² thick has considerably long lifetime compared to other same thin thickness as foils in the world. [18,19]

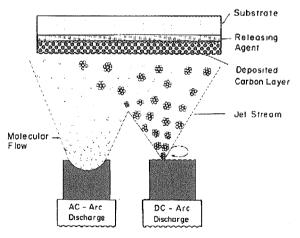


Fig.1 Schematic drawing of the carbon deposition images of the DC and AC arc-discharge sources. The CDAD method uses only DC-arc discharge source, and the CADAD method uses both AC and DC arc-discherge sources.

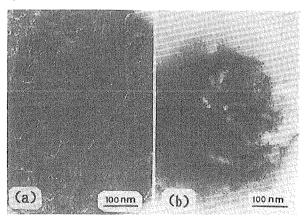


Fig.2 Transmission electron micrographs (TEM) of carbon foils. (a) Structure of a foil composed of carbon particles emitted from the anode only (Wc=0). (b) Structure of a foil composed of carbon particles from the cathode only (Wa=0).

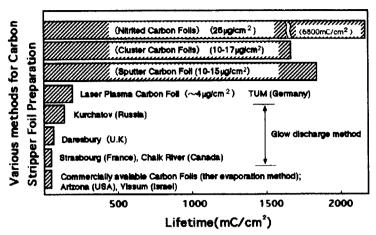


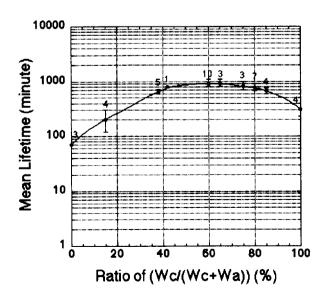
Fig.3 Lifetime results for carbon stripper foils made by various preparation methods. The measurement was performed with a 3.2 MeV Ne $^+$ at 2 - 3 μ A beam current with a 3.5 mm ϕ beam spot.

2.2 Lifetime dependence on the weight loss of carbon electrodes

In the preparation procedure, we measured the current intensity of the carbon arc-discharge, and the lifetime dependence on the arc current intensity.

We, then, believed that the electrodes in a DC arc discharge play an important role in producing carbon foils with long lifetimes. Therefore, we measured the weight loss of the electrode due to ablation from the cathode electrode. The lifetimes of foils depend on the ratio R=Wc/(Wc+Wa) x 100%, where Wc and Wa stand for the weight loss of the cathode and the anode, respectively. The DC power of the arc (300A x 20V) was pulsed with an interval of 3-5 seconds in order to suppress rising of the cathode spot temperature. The amount of Wc is sensitively to the cathode spot temperature and hence we controlled the ratio R by adjusting the discharge interval. The lifetime results as a function of the ratio R are shown in Fig.4.

On the anode-rich emission side, the lifetime is remarkably decreased, and on the cathode-rich emission side, the lifetime is decreased only slightly. The optimum condition for long-lived carbon foils is in the Wc range from 50 % to 80 %. The foils for the present preparation were deposited by R=60-70 % onto the glass substrate. The foil thickness was $15\pm5~\mu g/cm^2$. We named the foils made by R larger than 60 % and 5-50 $\mu g/cm^2$ in thickness, cluster carbon foils. Mixing of carbon particles emitted from both the anode and cathode rod is very essential in making strong carbon foils. Foils composed of particles from only the cathode rod are found to be very weak under mechanical stress.



The lifetime of the foils made with the by R=0 % was nearly the same as that of commercially available standard (CM) foils. On the other hand, the lifetime of foils composed of R=100 % did not reach a maximum, but were longer than 10 times as compared with the CM-foils.

Fig.4 Relation between the ratio R=Wc/(Wc+Wa) and the lifetimes. Error bars represent mean square errors and the numbers attached to the points are the number of measured sample. Tested foil thickness are 10 - 15 μg/cm² and the lifetime measurement were performed with a 3.2 MeV, Ne⁺ at 2 - 3 μA of 3.5 mmφ beam spot.

3.1 Thick foil preparation

Thick carbon foils of about 130 μ g/cm² were prepared by means of electron beam gun at Westinghouse Hanford Company, Richland, Wa. USA, and by AC arc discharge of spectroscopically pure graphite at the INS, University of Tokyo. In latter cases, the carbon foils were deposited onto substrate covered with a releasing agent of cream-coat. The deposition system used in this work is nearly the same as described in Ref [12, 13]. The electron beam evaporation was performed using a high tension of 4 kV and 500 mA. For the carbon deposition by the AC arc discharge graphite rods of 8 mm diameter at a current of AC-250A were used, and the unusual thick carbon foils of 130 μ g/cm² were prepared without any problem.

Next, we tried to apply the DC arc discharge method for the required range of thickness. This method, however, showed a limit on the thickness obtained. Deposited carbon layer always curled and peeled off from the substrate at approximately $60 \mu g/cm^2$ as shown in Fig.5(a), 5(b) and 5(c). Then, in order to overcome the peeling off, we applied the CADAD method [12, 13]. We, however, had no experience of foils thicker than $60 \mu g/cm^2$ so far.

We deposited by using a type ||| (large particle layer is sandwiched by fine particle layers) multi-layer configuration (see Fig.2 of Ref.[13]). The DC power of 20 V x 300 A and AC power of 20 V x 300 A were switched on and off alternately for periods necessary to obtain the required thickness. We were able to prepare foils by keeping the ratio parameter R=Wd/(Wd+Wa) at 50 - 80 %, where Wd and Wa are the carbon source weight losses due to ablation from the DC and AC arc discharge, respectively. In this preparation, three layers were deposited onto the glass substrate: The first layer with a thickness of 30 µg/cm² emitted from the AC arc discharge method, the second layer of 60 µg/cm² was deposited using the DC arc discharge procedure, and finally the third layer of 30 µg/cm² was deposited with the AC method, resulting in a total thickness of about 120 µg/cm². The vacuum in the arc discharge was ~1 x 10⁻³ Pa from 1 x 10⁻⁴ Pa without the arc discharge. Figures. 5(d), 5(e) and 5(f) are photographs of such carbon foils of 120 µg/cm² made by the CADAD method, (d) R=70 %, (e) R=60 % and (f) R=50 %. We found that such foils close to 100 μg/cm² in thickness tended to curl while removing the substrate by floating on water. This problem was solved by an annealing technique, in which the deposited layers on the glass substrate were placed 3 cm above a Mo filament heater, and heated to temperature of about 350°C for 5 h in vacuum of ~10⁻⁴ Pa. The foil thus prepared was supported by carbon fibers, as shown in Fig.6(a) to reduce scattering with the circulating beam. The stamp support as shown in Fig. 6(a) reduce beam losses. Recently, the new ribbon foil support was developed to lengthen the lifetime of the stripper (Fig.6(b)). This technique supports the foil by attaching it to the aluminum frame. The doubling of a foil improves the stripping efficiency because the probability of accidental coincidence of pinhole positions between two sheets is extremely low. The foils of 15 x 50mm² size was then sandwiched between tight-stretched carbon fibers of 4-5 µm diameter in a cross pattern as shown in Fig.6 and described in Ref.[14, 16]. The size of doubled foil is then 15 x 50 mm² glued along one to the aluminum frame with a conductive paint. The two aluminum frames are bolted together with the doubled foil suspended between the tight-stretched carbon fiber grids.

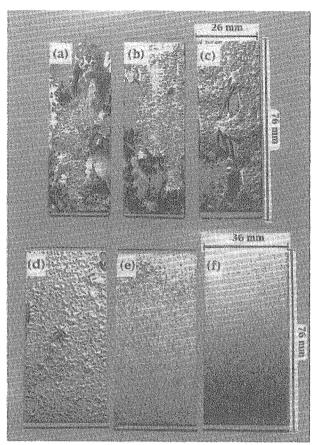


Fig.5 Photographs of carbon deposited onto the substrates by the DC arc discharge method; (a), (b) and (c), and the CADAD method; (d): R=70 %, (e): 60 %, (f): 50 %.

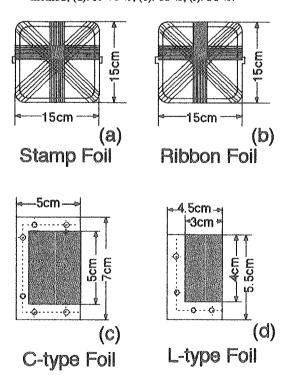


Fig.6 Carbon stripper foils drawn by dotted area on various type frames supported by 4-5 μm diameter carbon fibers. (a) "Stamp" type carbon stripper foil. (b) "Ribbon" type carbon stripper foil, (c) C-type frame carbon stripper foil, (d) L-type frame carbon stripper foil

3.2 Lifetime measurement with high energy 800 MeV, H ion beam

Lifetime measurements were performed with an 800 MeV proton beam in the PSR at Los Alamos National Laboratory.

Figure.7 shows the layout of the PSR during the period of these tests with injection and

extraction beam transport lines.

The negative hydrogen ions before entering the PSR are neutralized by a stripper magnet to H^0 and then stripped to H^+ by ~200 $\mu g/cm^2$ carbon foils supported with carbon fiber. The pressure in PSR was about 1 x 10⁻⁴ Pa. The PSR now uses direct H^- injection and strips directly to H^+ proton with a 400 $\mu g/cm^2$, four layer, CADAD carbon foil. The foil was initially conditioned with a low intensity beam 1-10 μA for 5 to 10 minutes and then irradiated with the production beam at a maximum injection current of 80-100 μA .

The effective foil lifetime was determined by the maximum tolerable beam current at the injection beam dump.

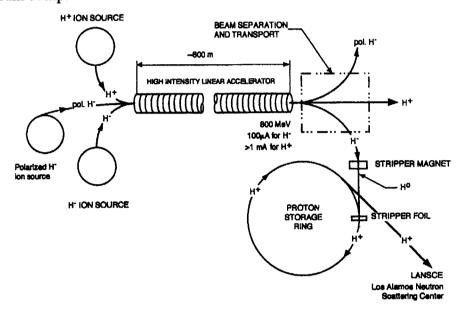


Fig.7 The layout of the former Los Alamos Meson Physics Facility (LAMPF)

If the lifetime measurements, standard foils from Arizona and from Israel (CM-Y) were compared with foils from Hanford (CM-A) and our first special CADAD (shown as CADAD in the Figure) foils during a beam time of about three months in 1992.

The results are shown in Fig.8(a) in 1992, where the vertical and the horizontal axis are each first turn loss rate and lifetime (date), respectively. The lifetime of those foils is generally shorter by fast thickening. The foil made by the CADAD method unfortunately dropped out of its frame due to failure of the supporting carbon fibers, and the measurement was stopped after one week of beam irradiation. As can be seen from the graph in Fig.8(a) or (b) the foil is not broken and shows relatively little thickening as indicated by the small changes in "first turn" losses. During about 17 days beam time in 1993, one of our (nitrided carbon foil) IBSRN foils [15], one foil from Hanford in its "ribbon" configuration and another CADAD foil were tested. Their performance is shown Fig.8(b) and "Hanford" (Westinghouse) is referred to the ribbon foil shown in Fig.8(b). The nitrided carbon foil was fold quadruple in order to get a sufficiently thick foil. The CADAD foil showed a long lifetime of 28 days, corresponding in a minimum of 4 times the lifetime of all other foils tested. Data from the diamond foil are not shown in Fig.8, because the foil ruptured in a short beam time of only 4 hours.

From the results performed in 1992 and 1993, the foils made by the CADAD method have shown to have reproducibly longer lifetimes, as compared to other foils tested.

In 1995, a ribbon foil with double layered (240 μ g/cm²) made by the same CADAD evaporation method as used in 1993, was used for the beam production. The beam condition was 800 MeV and 80-100 μ A. The beam production was continuously performed except

during ion source recycles once per month and outages. As shown in Fig.8(c), the foil showed an extremely long lifetime of three months and 17 days as compared to the (from August to November, 1995) previous 28-days obtained in 1993. The lifetime of the foil corresponds to about 4 times longer lifetime than the foil tested in 1993 and about 17 times, as compared to commercially available standard foils.

The foil did not broken, and survived until the end of the beam time and could be used for next run. After irradiation, the ribbon foil was placed in a box, as shown in Fig.9. Visual inspection of the irradiated foil showed slight bending the top of the foil (Fig.9). Supporting fibers broken by besm irradiation may have contributed to the bending and curling of the foil.

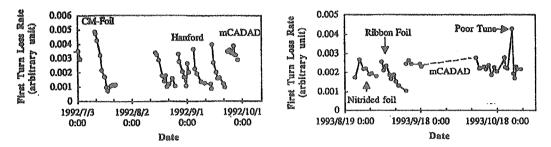


Fig.8(a) and (b) The results obtained in the lifetime measurements with 800 MeV, $85\,\mu\text{A}$ H beam. in (a) 1992 (b) 1993.

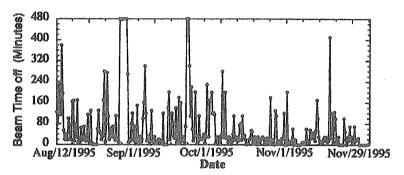


Fig.8(c) Beam delivery performance during the record-breaking lifetime for 800 MeV, H⁺ion beam bombardment in the proton storage ring (PSR) at Los Alamos National Laboratory.

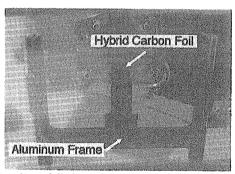


Fig.9 A photograph of the thick carbon stripper foil showed the record-breaking lifetime by irradiation for 103 days of an 800 MeV, 80-100 μ A, H⁺ion beam

3.3 Lifetime measurement with a low energy beam of 3.2 MeV, Net Ion

The experimental apparatus for this measurement was nearly the same as that described in Fig. 1 of Ref. 13. The beam from the Van de Graaff accelerator at the Tokyo Institute of Technology was used. The current intensity was $2 - 3 \mu A$ with a spot size of $3.5 \text{ mm}\phi$. Twenty sheets of carbon foils from $50\text{-}200 \mu g/\text{cm}^2$ in thickness were mounted on 0.2 mm thick stainless steal holders made by a photo-etching technique. The beam profile was checked with a quartz disk of 1 mm thick and 20 mm ϕ . The measurements were performed in a high vacuum chamber. The pressure of the chamber was 1.2×10^{-4} Pa (the partial pressure

were; H_2 : 8.6 x 10^{-7} Pa, H_2O : 6.1 x 10^{-5} Pa, N_2+CO : 3.7 x 10^{-5} Pa, O_2 : 2.2 x 10^{-5} Pa: Ar: 1.3 x 10^{-6} Pa and CO2: 2.7 x 10^{-7} Pa). In this measurements, foils of 50-200 $\mu g/cm^2$ thickness, made by the CADAD and foils made by the glow discharge methods from Russia (GD-R) and Chalk River Nuclear Lab. (GD-C), and foils from Hanford, diamond foils by the Kobe Steel Co. in USA, Al_2O_3 foil from the Rutherford Appleton Lab. and CM foils were tested. The single foils of ~50 $\mu g/cm^2$, ~100 $\mu g/cm^2$ and double foils of ~200 $\mu g/cm^2$ were sandwiched with the same flames.

The lifetime results are shown in Fig.10(a) \sim 200 µg/cm², (b) \sim 100 µg/cm² and (c) \sim 50 µg/cm². The "AC-250A" indicated the foil was made by an AC arc discharge method with AC current of 250 A. The CM(A) is from Arizona carbon foil Co., and CM(Y) from Yissum Research Development Co., of Israel.

3.4 The behaviors of foils under 3.2 MeV Net heavy ion beam bombardment

The foil surface was observed with a camera during irradiation with a 3.2 MeV, Ne⁺ ions. These visual observations are important not only to check the lifetime, but also to obtain information on the appearances of shrinkage and destruction phenomenon, which depends on

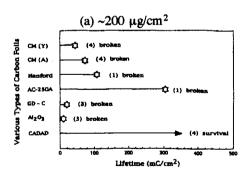
the foil preparation method.

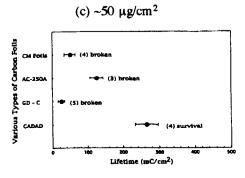
The typical behaviors of thick foils are photographed as shown in Fig.11. A foil of 124 µg/cm² thick made by the CADAD method shown in Fig.11(a) survived for 2092 mC/cm². This corresponded to the longest lifetime among the foils tested without rupture. The lifetimes of foil (b) (200 µg/cm² thick double layers) made by Hanford and (c) CM foil (110 µg/cm²) were 126 mC/cm² and 32 mC/cm², respectively. The foil (d) (202 µg/cm² thick double layers) made by the glow discharge method (GD-R) survived up to 718 mC/cm² of integrated current. The aluminum oxide foil (e) (169 µg/cm², folded in double) was made by the anodic oxidation method, ruptured at 2.0 mC/cm². The lifetime of the foil in Fig.11(d) is long, but visual observation indicated that one layer broke at the early stage of 23 mC/cm², as shown in Fig.5(d).

Figure 11(a) shows the surface appearance of the foil. The temperature at the beam spot was about 1500 °C. The temperature was measured through a BaF₂ window using a spot thermometer (TR-630A) of Minolta Camera Co. Ltd. Japan. From this photograph, the

shrinkage due to the bombardment can not be seen clearly.

Figure 11 indicates the great difference especially shrinkagein surface appearance between (a) and the others. It was found that the present CADAD method can produce very stable and long lived foils under heavy ion beam bombardment at high intensity.





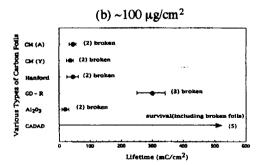


Fig. 10(a) The results of the lifetime measurements with 3.2 MeV, $3{\sim}4~\mu\mathrm{A}~\mathrm{Ne}^+$ beam. The foil thicknesses are ${\sim}200~\mu\mathrm{g/cm^2}.$ The numerical values in the figure represent the numbers of samples and the error bars represent standard deviations calculated from the data obtained. The foils made by the CADAD method show long lifetimes, regardless of the thickness, compared to the other foils tested.

Fig.10(b) The results of the lifetime measurements with 3.2 MeV, $3\sim4~\mu A$ Ne⁺ beam. The foil thicknesses are $\sim100~\mu g/cm^2$.

Fig. 10(c) The results of the lifetime measurements with 3.2 MeV, $3\sim4~\mu A~Ne^+$ beam. The foil thickness are $\sim50~\mu g/cm^2$.

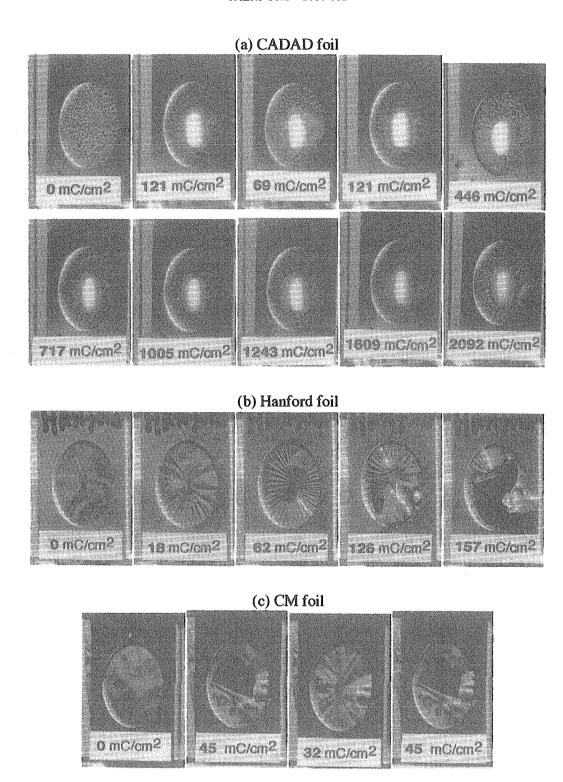
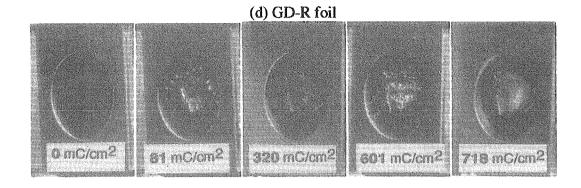


Fig. 11 The behavior of a foil surface inspected by a camera; (a) foil made by the CADAD method (thickness is $124 \,\mu\text{g/cm}^2$), (b) Hanford foil (200 $\,\mu\text{g/cm}^2$ thickness double layer) and (c) CM foil (100 $\,\mu\text{g/cm}^2$)



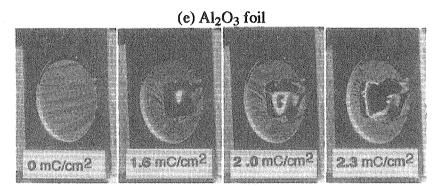


Fig.11 The behavior of a foil surface inspected by a camera; (d) GD-R foil (202 μ g/cm² thick double layers) and (e) made by the anodic oxidation method (169 μ g/cm²)

4. Conclusion

We have investigated the lifetimes of various carbon stripper foils from 10 $\mu g/cm^2$ to 200 $\mu g/cm^2$.

The ribbon carbon stripper foil made by the CADAD method accomplished a great achievement of a record-breaking lifetime of 103 days at the continuous operation of the 800 MeV, $80-100 \mu A$, as performed in 1995.

This lifetime corresponds approximately to 4 times longer than 28 days performed in 1993 and 17 times, compared to the commercially available standard foils.

The cluster thin foils made by the CDAD method showed the longest lifetime, compared with those of other foils tested, including commercially available standard foils.

The CADAD foils also showed the longest lifetimes compared to all other foils, for a high energy proton beam of 800 MeV and low energy Ne⁺ beam of 3.2 MeV.

The CADAD method is confirmed to be highly reproducible and could produce carbon stripper foils of a wide thickness range from 10 to about 120 μ g/cm². The thick CADAD foils were very stable and showed very small shrinkage rates when irradiated with a high intensity beam.

The decreased shrinkage rates associated with these foils allow the use of narrower foils, thereby decreasing stored beam losses. Lower beam losses imply that the ring components are less radioactivited and therefore worker radiation exposures will be reduced.

From the experiments performed in 1992, 1993 and 1995, we found a serious problem that the supported carbon fibers showed very fragile against the long time ion irradiation with high intensity 800 MeV, >85 μ A. We also need stronger carbon fiber materials for higher beam intensity in future. We also would like to do more pioneering work for new materials for stripper foils.

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