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THE BEHAVIOUR OF CARBON STRIPPER FOILS UNDER HEAVY ION BOMBARDMENT

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Résumé. — On a étudié la durée de vie de feuilles de *stripping* en carbone par bombardement d'ions A^{++} de 4,8 MeV sous vide en absence d'hydrocarbures. La vie des feuilles est limitée non pas par l'épaississement mais par la contraction et la rupture, et se laisse prolonger par un facteur 3 en menant l'irradiation à une température de 600 °C. Des feuilles plus minces (nominale de mi-épaisseur) produites sur un support de cellulose ont une vie plus courte mais, vu que le faisceau transmis est plus intense, on constate pour elles une amélioration de 30 % en ce qui concerne la vie utile. Pour augmenter la durée de la vie des feuilles de *stripping*, il faudra ou bien en réduire le taux de contraction ou bien en améliorer la résistance à la contraction.

Abstract. — The lifetime of carbon stripper foils when bombarded with 4.8 MeV A^{++} ions in a hydrocarbon-free vacuum environment has been investigated. The lifetime of the foils is limited not by thickening but by shrinkage and rupture, and can be improved by a factor of 3 by irradiating at a temperature of 600 °C. Thinner foils (nominally half thickness) produced on a cellulose backing had a shorter life, but since the transmitted beam is greater, they show an improvement of 30 %. Further improvements in foil lifetime can only be achieved by reduction of the shrinkage rate, or by improvements to the resistance of the foils to shrinkage.

1. Introduction. — Carbon stripper foils are preferred for the generation of high energy heavy ion beams in tandem electrostatic accelerators since they produce higher mean charge stages than gas stripping systems. However, the lifetime of carbon stripper foils can become intolerably short when bombarded with heavier ions. Consequently attention has been widely paid [1-7] to the causes of the reduced lifetime and to methods of extending the useful lifetime when bombarded with heavy ions.

In the first phase of a systematic study of the factors controlling the lifetime of carbon stripper foils [1], foils (nominally 5 $\mu\text{g}/\text{cm}^2$) were bombarded with 4.8 MeV doubly charged argon ions from the Harwell 6 MeV Van de Graaff accelerator using a conventional vacuum system. The foils, produced by evaporation from a carbon arc, were found to thicken considerably when bombarded at room temperature at current densities less than 0.1 $\mu\text{A}/\text{mm}^2$ and the lifetime was usually limited by the resulting excessive scattering of the transmitted beam. However, if the foils were heated in an oven to 600 °C the thickening rate was reduced considerably and the lifetime was increased by at least a factor of 2 and then was usually limited by rupture of the foil. A similar behaviour was found when the foil was bombarded at nominally room temperature but with a sufficiently high current density to cause local heating of the foil. It was evident that two processes were contributing to the

failure of the foil : thickening, which was attributed to the build-up of carbon on the surface of the foil due to the cracking by the ion beam of hydrocarbons present in the vacuum environment, and rupturing, which was caused by shrinkage of the foil associated with the radiation damage to the foil by the ion beam.

Since the hydrocarbon-thickening process can readily be avoided by using vacuum techniques to exclude hydrocarbon contamination, a special experimental system has been constructed to test the lifetime of stripper foils in hydrocarbon-free vacuum conditions, in order to determine the rupture resistance of foils under varying bombardment conditions.

2. Experimental Details. — The *clean line* system for irradiating carbon stripper foils in hydrocarbon-free conditions is shown schematically in figure 1. 4.8 MeV A^{++} ions from the Harwell 6 MeV Van de

FIG. 1. — Hydrocarbon-free carbon foil irradiation facility.

Graaff accelerator were directed down the stainless steel flight line and focussed by a pair of magnetic quadrupole lenses to a spot of 1.7 mm diameter on the stripper foil positioned in the target chamber and the transmitted beam was measured in the Faraday cup. The beam spot was examined with profile monitors before and after the foil position to give both spot size and relative intensity. The beam could also be observed at the foil position by replacing the foil by a quartz disc.

A pressure less than 10^{-7} torr was obtained in the flight line by connecting the flight line to the accelerator with a long, narrow restrictive aperture and by pumping the line with a 4" liquid nitrogen trapped diffusion pump charged with polyphenyl ether oil. Hydrocarbons from the accelerator were trapped by an in-line refrigerated baffle. The remaining sections of the flight line were pumped with three ion pumps, whose main function was to act as holding pumps, since their pumping speed for argon is poor. At the end of the flight line another restriction isolated the target chamber, which was also pumped by a 4" diffusion pump. After a short bakeout at 150 °C a pressure of 10^{-8} — 10^{-9} torr was obtained in the foil chamber. The partial pressure of hydrocarbons was less than 5×10^{-10} torr as measured by a MS 10 mass spectrometer attached to the chamber. The residual gases were mainly hydrogen, water vapour, methane and carbon monoxide. In order to facilitate foil changes, the foils were inserted into the chamber *via* either of a pair of independently pumped air locks. The foils could be heated to 1 000 °C using an electron-beam heated oven surrounding the foil. During irradiation at 1 000 °C the pressure rose initially due to an increase in the H₂ and CO partial pressures but remai-

ned below 5×10^{-8} torr thereafter. Some ethylene was also released from the foil by the ion beam.

During bombardment, the behaviour of the foils was followed by observing the shape of the transmitted beam on the downstream profile monitor. If the foil split, a large peak of unscattered beam could be seen, superimposed on the normal scattered beam profile. Often the split could also be seen by examining the foil through windows in the target chamber but this was not always possible. The foil lifetime was set arbitrarily at the time when the sharp peak of the unscattered beam on the profile monitor was the same height as the scattered beam.

3. **Results.** — The hydrocarbon-free irradiation facility has been used to compare the lifetime of carbon stripper foils under various conditions. Ini-

FIG. 2. — Histograms of measured foil lifetimes at ambient, 600 °C and 1 000 °C.

TABLE I
Results of Lifetime Measurements

Type of foil	No. of foils irradiated	Temperature	Approx Irradiated area (mm ²)	Mean Lifetimes* Dose to first split (μA min.)	Dose to rejection (μA min.)
A Standard					
~ 5μg/cm ²	16	ambient	~ 20	21.3 ± 3	25.4 ± 4
Standard					
~ 5μg/cm ²	7	600 °C	~ 20	39.6 ± 5	77.5 ± 10
Standard					
~ 5μg/cm ²	5	1 000 °C	~ 20	40.7 ± 3	53.4 ± 6
B Scanned beam	12	nominally ambient	~ 40	28.9 ± 3	33.9 ± 4
Scan off	6	nominally ambient	5-10	19.1 ± 1	27.9 ± 10
C Thin backed	7	ambient	5-10	12.9 ± 2	16.2 ± 3
Standard	7	ambient	5-10	19.3 ± 3	21.8 ± 3

(*) Charge current : divide by 2 to obtain particle current for A⁺⁺ beam.

tially the improvement in lifetime achieved by heating the foils [1] was re-examined by bombarding foils ($\sim 5 \mu\text{g}/\text{cm}^2$) with 4.8 MeV A^{++} ions with a measured beam current of 0.5 μA spread over an area of $\sim 20 \text{ mm}^2$. Contrary to the first experiments in a hydrocarbon contaminated system the foils did not thicken when bombarded at ambient temperatures and the foil eventually ruptured. The lifetime of the foils varied considerably, as shown in figure 2, so that many foils had to be tested in order to achieve a reliable value for the mean lifetime. The mean lifetimes of foils tested during these studies are compiled in table 1, together with the error of the mean estimated from the distribution of lifetimes. Foils irradiated at 600 °C lasted 2-3 times as long. The lifetime of foils tested at 1 000 °C was slightly less than at 600 °C, but since this result was derived from only five foils it is possible that the difference is merely due to the small number of observations. It was noted that part of the improvement in lifetime was realised due to the increased stability of heated foils after they began to split. Usually, cold foils curled over rapidly after a split appeared, but hot foils often remained usefully in the beam.

Some improvement in foil lifetime might be expected by scanning a fine beam spot slowly around the foil [2]. Accordingly, by applying suitable magnetic deflection, a 0.6 μA beam spot 3 mm diameter was rotated around a circular path 6 mm diameter at a frequency of 0.3 Hz. The improvement in lifetime achieved was somewhat less satisfactory than that expected on the basis that the lifetime would be enhanced by the ratio of the bombarded areas.

The lifetime of standard foils has also been compared with that of thinner foils (nominally half thickness) prepared on a cellulose backing which is burnt away during irradiation leaving a foil thinner than can be made directly. Although the lifetime of the thin foils was only ~ 0.7 that of the standard foils, these thinner foils may be advantageous since the beam is scattered less and the resulting lifetime in terms of the useful *transmitted* beam is a factor of 1.4 greater than standard foils.

4. Discussion. — These results have shown that thickening may be eliminated as a cause of foil life limitation by suitably excluding hydrocarbon vapours from the vicinity of the foil, either by ultra-high vacuum techniques or by reducing the molecular residence times on the foil by heating. It is evident that cryogenic trapping of hydrocarbon vapour would be equally successful.

The main limitation to the use of carbon stripper foils with heavy ions is therefore the shrinkage and rupture of the foils. Improvements to the lifetime must therefore be directed towards reducing the shrinkage rate and increasing the rupture resistance.

Shrinkage of the foil is associated with the radiation

damage created by the bombardment. Carbon stripper foils prepared by evaporation on to a cold substrate are amorphous, with a disordered structure consisting of bonds partially graphitic and diamondine in nature [8] which give broad electron diffraction bands. During bombardment radiation enhanced diffusion of the carbon atoms, using energy gained by elastic collisions with the ion beam, enables sintering and reordering to occur as shown by narrowing of the broad bands in the electron diffraction pattern. The resulting shrinkage has also been observed in bulk graphite and pyrocarbons and found to be a minimum at 600 °C [9, 10]. Surface energy effects also contribute to shrinkage, since a thin foil may reduce its surface energy by decreasing its surface area by surface diffusion mechanisms.

The carbon atom displacement rate can be estimated from the nuclear energy loss [11] using the improved Kinchin and Pease [12] displacement model. If the displacement energy for carbon is taken as 30 eV, each atom is displaced ~ 3 times during the lifetime of the foil when bombarded with 4.8 MeV A^{++} ions. If the lifetime is assumed to be dependent only upon the total number of atom displacements, the lifetime of the foils may be predicted for bombardment with other ions and energies. At higher energies the nuclear energy loss is decreased, resulting in an increase of 6 in the lifetime for 40 MeV A^+ ions. The energy loss is increased for heavier ions, so that the lifetime is predicted to be only 0.6 $\mu\text{A min.}/\text{mm}^2$ for 100 MeV I^+ ions, under similar conditions.

At low temperatures, carbon atom mobility is restricted to the irradiated area, so that only the irradiated area is affected. However, at sufficiently high temperatures (e.g. 1 500 °C) thermal diffusion will occur, with spontaneous graphitisation of the foil and rapid failure. The shrinkage rate is therefore dependent upon the ion mass, energy, flux and temperature. The resistance of the foil to shrinkage will depend upon the foil's elasticity, absence of defects, initial slackness and any processes, radiation or thermal, which relieve the stresses induced by the shrinkage strains. The increased lifetime of foils irradiated at 600 °C may be attributed to the reduced shrinkage rate at this temperature. Also it is probable that the stresses are relieved by radiation enhanced creep. From the limited information on foils irradiated at 1 000 °C it is not clear whether the lifetime is as long and it is possible that the shrinkage may be starting to increase again.

In some cases, the shrinkage resistance of the foils can be improved by optimization of the irradiated area. It can be shown that the radial stresses in the foil due to the shrinkage caused by a given uniform ion dose are constant within the irradiated area, and outside decrease towards the holder. Decreasing the irradiated area increases the maximum stress considerably. Calculations also show that the tangential

stresses change sign at the edge of the irradiated zone. Since the strength of the foil is greater than the interfacial strength between the foil and its holder and additionally the probability of finding a defect in the foil is greatest at the edge, it is reasonable to assume that the optimum diameter of the irradiated area is approximately half the total diameter of the foil. When this condition is reached the foil is likely to rupture either outside the irradiated zone or at the edge of the holder, depending upon the position of the weakest point. The split then runs normal to the maximum stress, i.e. tangentially to the irradiated zone, as observed in practice. If the irradiated spot size is too small, the foil splits rapidly around the edge of the irradiated area. Thermal fatigue effects are also likely to promote early failure of the foil. Since the foil is thin, with a thermal response time of ~ 10 ms any periodic fluctuations in the ion beam may cause oscillation of the foil temperature with the possibility of fatigue and reduced lifetime.

5. Conclusions. — Measurements of the lifetime of carbon stripper foils irradiated with 4.8 MeV A^{++} ions in a clean vacuum environment have shown that the lifetime of foils is limited by radiation induced shrinkage. The lifetime may be increased by a factor of 3 by bombardment at a temperature of 600 °C. An analysis of the cause of failure indicates that substantial improvements to the lifetime may only be achieved by reducing the shrinkage rate or by increasing the capacity of the foil to resist shrinkage.

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