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# Preparation and comparative testing of advanced diamond-like carbon foils for tandem accelerators and time-of-flight spectrometers

V.Kh. Liechtenstein[a,*], T.M. Ivkova[a], E.D. Olshanski[a], A.M. Baranov[b], R. Repnow[c], R. Hellborg[d], R.A. Weller[e], H.L. Wirth[c]

[a]*Russia Research Centre, "Kurchatov Institute", 123182 Moscow, Russia*
[b]*Research Institute of Vacuum Technique, 113105 Moscow, Russia*
[c]*Max–Planck-Institute für Kernphysik, 69117 Heidelberg, Germany*
[d]*Lund University, S-22362 Lund, Sweden*
[e]*Vanderbilt University, Nashville, TN 37235, USA*

**Abstract**

The sputter preparation technique for thin diamond-like carbon (DLC) foils, advantageously used for ion-beam stripping and timing in accelerator experiments, has been optimized to improve the quality and the performance of the foils. Irradiation lifetimes of 5 μg/cm$^2$ DLC foils prepared by this technique have been compared with those for foils of approximately the same thickness, prepared by laser plasma ablation and for ethylene cracked foils when bombarded by 11 MeV $Cu^-$ - and $Au^-$-ion beams of $\sim 1$ μA beam current at the Heidelberg MP-tandem. Standard carbon arc-evaporated foils were used as references. In these experiments, DLC stripper foils appeared to have a mean lifetime approximately two times longer than ethylene-cracked foils regardless of ion species, and compared favorably with foils prepared by laser ablation method. All these foils lasted at least, 10 times longer than standard carbon foils, when irradiated in the MP terminal. Approximately, the same improvement factor was confirmed with 3 μg/cm$^2$ DLC stripper foils irradiated with 2.3 MeV Ni-beams at the Pelletron accelerator in Lund. Unlike standard carbon foils, most of the advanced lifetime foils exhibited thinning during long irradiation, under clean vacuum. This suggests that sputtering of the foil by the heavy-ion beam might be a dominant process, responsible for the observed failure of these long-lived strippers. Along with specifically corrugated self-supporting DLC beam strippers, we succeeded in the fabrication of very smooth and ultra thin ($\sim 0.5$ μg/cm$^2$) DLC foils, mounted on grids and used as start foils for the ToF spectrometers applied in ion beam analysis. 

*Keywords:* Spectrometers; DLC foils; Tanden accelerators

## 1. Introduction

Thin and stable carbon foils are essential, e.g. for the performance of tandem accelerators and time-of-flight (ToF) spectrometers. Such foils are most critical relating to tandem accelerators of heavy ($Z > 16$) ions, for which beam strippers of 2–3 μg/cm$^2$ thickness are needed, to obtain an optimum ion transmission at about 10 MV terminal voltage [1]. However, standard evaporated carbon strippers do suffer from irradiation damage,

* Corresponding author. Fax: 7-(095)-943-0073.
*E-mail address:* liechten@qq.nfi.kiae.su (V.Kh. Liechtenstein)

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resulting in a lifetime of only several minutes when used with high-intensity heavy ion beams in modern tandems [2].

Despite the existence of several methods for the preparation of long-lived carbon stripper foils, short lifetimes of very thin foils for very heavy ions is still a serious operational limitation. Analysis of literature shows that well-known ethylene-cracked foils are ineffective as very thin beam strippers [3], while thin carbon foils prepared by laser plasma ablation deposition [4,5] are credited with the highest irradiation hardness. Unfortunately, this method is too expensive so far. Sugai et al. presented the development of long-lived foils using combined DC and AC carbon arc evaporation [6] or ion beam sputtering [7]. They conceded, however, that it is very difficult to fabricate such carbon foils thinner than 5 $\mu g/cm^2$ by these methods [8].

Diamond-like carbon (DLC) foils, developed at the Kurchatov Institute [9] have shown lately to be promising as long-lived heavy-ion beam strippers for tandem accelerators [10] and improved start foils for the state-of-the-art ToF spectrometers applied to ion beam analysis [11]. However, thin DLC strippers may need the ethylene cracked foils to be mechanically supported by an organic film. In this paper, next investigations are described which are aimed at the optimization of the preparation procedure of DLC foils, to fulfill the expanding requirements of both, the applications and the correct evaluation of the strength of the DLC stripper foils against heavy-ion irradiation at full-scale comparative experiments.

## 2. Experimental

### 2.1. Preparation of DLC foils

DLC foils in the thickness range of nominally 0.5–30 $\mu g/cm^2$ can be produced by a DC glow discharge sputter deposition process of pure carbon, which is described in detail [9,10]. The schematic set-up of the sputtering system originally constructed at the Kurchatov Institute to fabricate hard carbon coatings, is presented in Fig. 1(a) of [10]. Sputtered by a low-density krypton plasma, DLC films are formed by the deposition of carbon atoms outside the glow discharge region onto the glass slides, which are coated with a release agent and cooled to liquid nitrogen temperature. Characteristic properties of these DLC deposits, as measured on bare substrates are their extreme hardness, high electrical resistivity and high chemical inertness, making them attractive as protective coatings for metals [12].

Along with this special-purpose DC glow discharge system, a commercially available RF magnetron sputtering system was used to produce DLC foils (see Fig. 1(b) in [10]), which proved to be promising for the mass production of foils with properties close to those of the DLC ones. However, it became apparent that all foils from magnetron sputtering, tested so far, show less mechanical and irradiation hardness compared to foils from the DC setup. As a consequence, all measurements described below are related to DLC foils produced by DC glow discharge.

It is well known that the parting agent substantially influences the major properties of the resulting foil [13–15]. This is particularly the case for the deposition of DLC films onto a substrate, which might suffer from energetic carbon atoms during film growth. Besides, the characteristic features of DLC films may depend substantially on the substrate composition [16]. We have examined a variety of organic and inorganic release agents and found betaine and potassium oleate to be the most suitable ones for preparing, thin, corrugated stripper foils and ultra thin, smooth target DLC foils, respectively. Betaine was protected with about 0.5 $\mu g/cm^2$-thick evaporated carbon, against destruction during the deposition of DLC. This procedure yields strong and flexible DLC foils which withstand the usual low-pressure slackening procedure with an efficiency of nearly 100% for 3 $\mu g/cm^2$, with collodion backing.

To fabricate very smooth and ultra-thin foils, we examined several types of glass slides using an Alpha-Step 200 (Tencor Instruments) profilometer and found that ESCO slides worked best, yielding total RMS roughness of less than 3 nm for a slide coated with an alcohol solution of potassium oleate and polishing.

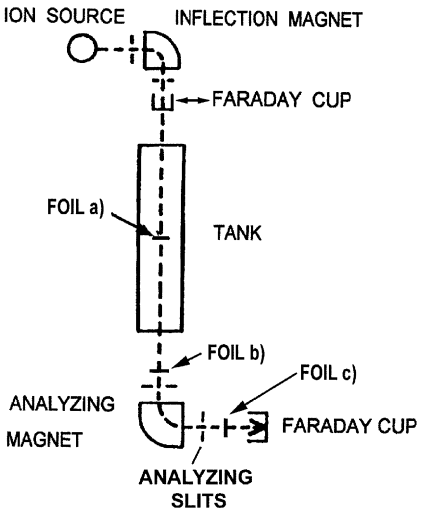


Fig. 1. Irradiation schemes of the stripper foil lifetime tests at the Heidelberg MP-tandem: (a) MP-terminal (negative ions), (b) MP-object (energy mixture), (c) MP-image (analyzed beam).

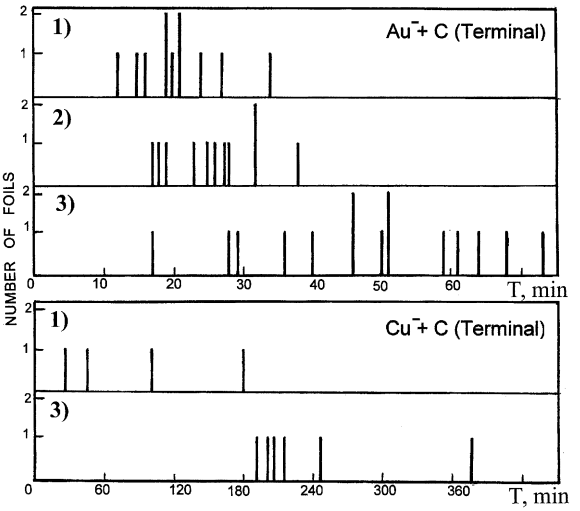


Fig. 2. Histograms of lifetimes of various foils when irradiated in the MP-terminal: 1 – ethylene-cracked foils; 2 – laser-plasma ablation foils; 3 – DLC foils.

### 2.2. Comparative lifetime tests of DLC stripper foils

The comparative lifetime tests of DLC stripper foils have been carried out at the MP–tandem (Max-Planck-Institute für Kernpysik, Heidelberg) in a clean, vacuum environment [17]. Two different locations have been used for the irradiations, as shown in Fig. 1. The foil condition during lifetime tests was monitored by a TV camera (beam-line runs) and by strip chart records of the ion currents. Also, all changes in the terminal voltage were chart-recorded. The terminal voltage is regulated by a signal from the analyzing slits. Thus, an increase in the thickness of the terminal stripper foil leads to an increase in the terminal voltage. The terminal vacuum pressure was in the $10^{-5}$ Pa range.

The $\sim 5\ \mu g/cm^2$ DLC foils were mounted on frames with about 13 mm diameter apertures and irradiated, together with ethylene-cracked [18] and laser plasma ablation foils of approximately the same thickness, prepared as described in Refs.[4,19]. Most of the foils were slackened by the low-pressure procedure. The laser plasma ablation foils came on bigger frames than was suitable for this terminal. That is why they had to be re-mounted and thus, lost their slackening.

Some of these foils, made by different methods, were irradiated in the MP-terminal with 11 MeV $Cu^-$ or $Au^-$ ions of $\sim 1\ \mu A$ beam current and $\sim 5$ mm diameter beam spot. The lifetimes of DLC, ethylene-cracked and laser ablation foils, defined as, the time at which the analyzed beam drops to half its initial value, are presented in Fig. 2. In spite of the spread of obtained data, which is due to the slightly different properties of the irradiated foils, it is clear that DLC foils essentially outlast ethylene-cracked foils for both ion species, and compare favorably with laser-plasma ablation foils. To enable the most accurate comparison of the lifetime of the foils made by different methods, it was thought worthwhile to select and compare the analyzed beam plots for different foils with the same initial ion transmission, as shown in Fig. 3. The plots displayed in Fig. 3 confirm the encouragingly long lifetimes of DLC foils, with an enhancement factor $> 10$ in lifetime, compared to those of standard carbon foils. Some of the results for a large number of different foils of $\sim 5\ \mu g/cm^2$ thickness, tested as terminal beam strippers during a variety of irradiation runs at the Heidelberg MP-tandem, are compiled in Table 1. Along with these results, irradiation lifetimes are presented for

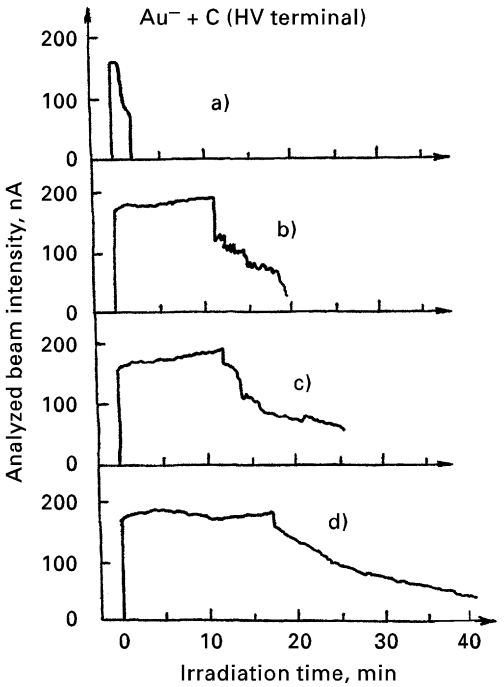


Fig. 3. Transmission plots of $^{197}Au^{14+}$ ions through the MP-tandem under high vacuum for different foils: (a) carbon arc evaporated foils, (b) ethylene-cracked foils, (c) laser-plasma ablation foils, (d) DLC foils.

3 μg/cm$^2$ DLC beam strippers and standard carbon foils, exposed to 2.3 MeV $Ni^-$-beams at the Lund University Pelletron accelerator. Lifetimes, as presented in Table 1, are defined in the usual way as, the time after which the intensity of the analyzed beam drops to half its initial value. As is well known, the lifetime of a carbon stripper foil is not very meaningful, the so-called quality factor [20–22] takes into account the integrated analyzed ion beam over the lifetime which is more useful for the user of the beam.

If we look again at Fig. 3, we see for all three types of improved carbon stripper foils, a sharp breakdown after 12–18 min. This indicates a rupturing of the foil and informs that the slackening was not sufficient. With more slackening, such carbon stripper foils could live even longer.

Although the terminal lifetime measurements are of most interest to one who wants to choose the stripper with optimum transmission and lifetime, beam line tests are a useful test of stripper performance. On the other hand, in most beam line tests current densities are higher and lifetimes are shorter than for foils exposed in a terminal. Results of lifetime tests for different foils irradiated in a beam line under significantly poorer vacuum as compared to the terminal, are shown in Fig. 4. It can be seen that, in contrast to the terminal measurements demonstrated in Fig. 3, the analyzed ion beam decreases monotonically during irradiation for all foils tested, and no remarkable improvement for any advanced lifetime foil is observed above the standard foil. The reason for this might be the carbon built up due to the cracked hydrocarbon deposits from the poor vacuum environment as

Table 1
Results of lifetime measurements in the terminals of the Heidelberg tandem (runs 1–3) and the Lund tandem (run 4)

| Run | Ion species | Energy | Foil type | Number of foils | Mean lifetime | | Quality factor (arb. units) |
|---|---|---|---|---|---|---|---|
| | | | | | (min) | (mc) | |
| 1 | $^{197}Au^-$ | 11 MeV | Ethylene | 11 | 21 ± 4 | 1.3 | 33 ± 6 |
| | | | Laser plasma | 11 | 26 ± 5 | 1.6 | 44 ± 6 |
| | | | Diamond-like | 15 | 48 ± 12 | 2.9 | 56 ± 8 |
| | | | Arc-evaporated | 1 | 4 | 0.2 | 5.1 |
| 2 | $^{197}Au^-$ | 11 MeV | Ethylene | 5 | 37 ± 14 | 2.2 | 58 ± 13 |
| | | | Diamond-like | 10 | 82 ± 45 | 4.9 | 98 ± 21 |
| 3 | $^{63}Cu^-$ | 11 MeV | Ethylene | 4 | 81 ± 45 | 4.8 | 96 ± 26 |
| | | | Diamond-like | 6 | 238 ± 50 | 14.2 | 282 ± 42 |
| 4 | $^{58}Ni^-$ | 2.3 MeV | Arc-evaporated | 3 | 10 ± 2 | 0.6 | — |
| | | | Diamond-like | 4 | 120 ± 22 | 7.2 | — |

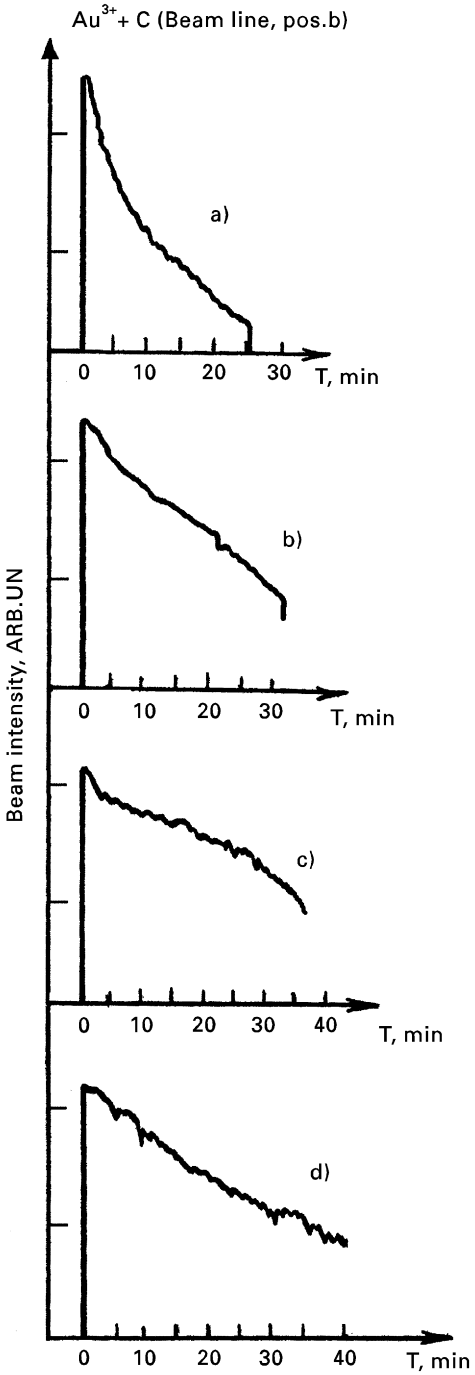


Fig. 4. Variations of the analyzed beam intensity with time for various foils when irradiated at the beam line (b) in poorer vacuum environment: (a) carbon arc evaporated foil, (b) ethylene -cracked foil, (c) laser-plasma foil, (d) DLC foil.

explained in Ref. [5]. Besides, it was revealed by the TV camera that the standard carbon foils ruptured, most likely, outside the beam spot soon after all slackened area was stretched, while holes in the irradiated area might cause foil failure much later.

Analysis of ion-transmission plots for different foils, irradiated in the MP-terminal (Fig. 1a) as well as in the beam line with the analyzed ions (Fig. 1c), together with measurements of changes in the terminal voltage and analyzing magnetic field, reveals that, unlike standard carbon foils, all advanced lifetime foils become thinner during the long irradiation under clean vacuum conditions. This suggests that sputtering of the foil by the heavy-ion beam, might be a dominant process for the observed failure of these long-lived strippers, which is in agreement with some published theoretical predictions [23] and experimental results [3].

### *2.3. The application of DLC foils to the ToF spectrometer for medium energy ion beam analysis*

Thinnest possible and very uniform secondary-electron emitting foils are needed for state-of-the-art ToF spectrometers, being used, in particular for ion beam analysis of semiconductors. It is most critical for ToF beam analysis in the medium-energy range (shown in Fig. 5) where ultimate resolution is often limited by the straggling in the foil and its non-uniformity due to roughness [24].

The improved tensile strength of the DLC foils, in combination with enhanced (due to negative electron affinity of diamond) secondary-electron yield, make them very attractive as substitutes for evaporated carbon foils for such applications. Some very smooth DLC foils with nominal thickness of about 0.5 $\mu g/cm^2$ were prepared as described above and picked up on frames with an 85%-transmission mesh covering a circular opening of 12.5 mm diameter. Typical atomic force microscope images of such a DLC foil and the thinnest possible standard foil used in earlier setups, are shown in Fig. 6. One such DLC foil was installed in the start detector of the ToF spectrometer instead of a nominally 1.2 $\mu g/cm^2$- thick standard carbon foil. As a result, experimentally observed energy resolution was improved from 2600 to 1350 eV for 104 keV He-ions.

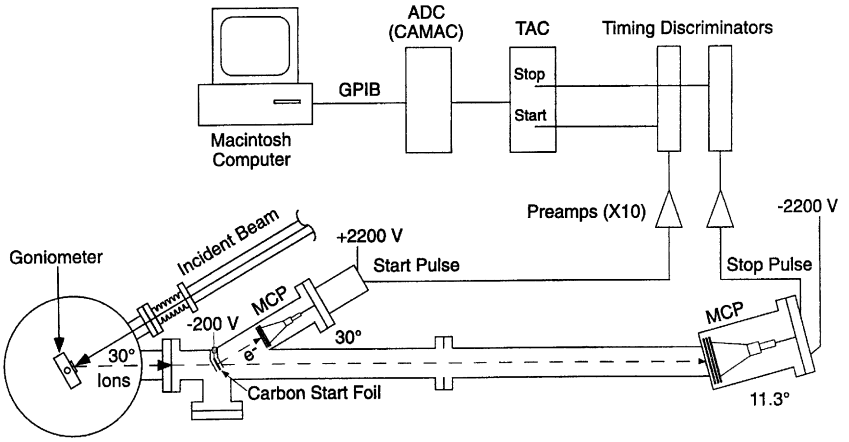


Fig. 5. Schematic of the ToF spectrometer system for medium-energy ion beam analysis.

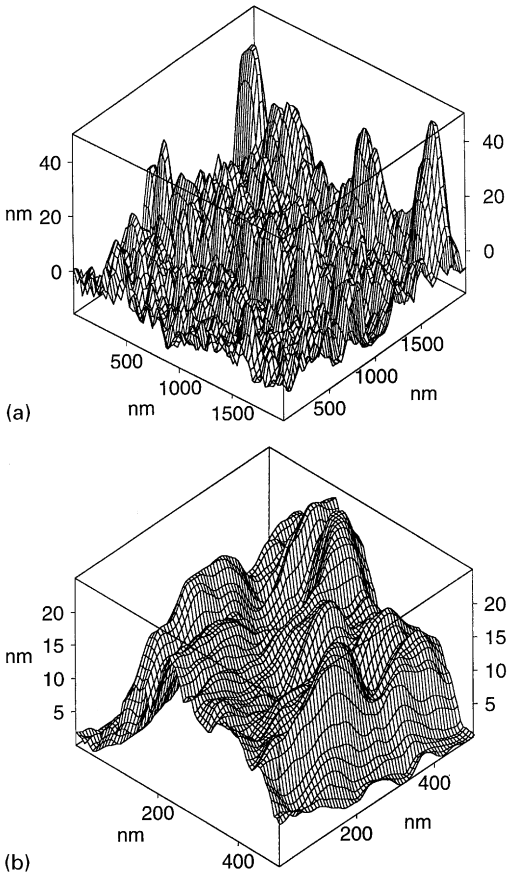


Fig. 6. Atomic force microscope images of a very smooth DLC foil (a) with about 0.5 μg/cm$^2$ nominal thickness and of a standard 1.2 μg/cm$^2$ carbon foil (b).

## 3. Conclusion

Thin DLC strippers for heavy-ion beams have proved superior with reasonable statistics, to ethylene-cracked foils with an improvement factor of two, for Cu and Au beams and compare favorably with laser-plasma ablation foils. All these foils lasted at least 10 times longer than the standard carbon foils, when irradiated at the HV terminal in good high vacuum, while the beam line runs showed improvement factors to be dependent on the irradiation conditions. Sputtering by ion bombardment is suggested to be a dominant process, responsible for lifetime of advanced lifetime strippers under very heavy-ion irradiation in clean vacuum environment. Such an outcome clearly directs further improvement efforts.

Ultra-thin and very smooth DLC foils for ToF spectrometers are developed and used advantageously for medium-energy ion beam analysis. Also, such foils can be used profitably in Coulomb-Explosion Imaging and other experiments which require very thin and stable carbon foils. High hardness of DLC foils against heavy-ion bombardment makes them favorable for use as backings and protective layers for thin sandwiched isotopic targets. Expanding possibilities of DLC target foils clearly need to establish methods for mass production.

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