

## GRAPHITE THIN FILMS FOR ACCELERATOR APPLICATIONS

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### Abstract

Graphite thin films were prepared through a simple two-step carbonization-graphitization process from polyimide films and were applied as rotating charge stripping disks for high intensity ion beam irradiation. These films demonstrated high mechanical robustness within the range of 300 – 1000 rpm under vacuum conditions and worked remarkably well as charge stripping foils. Changing the thickness of graphite thin films also enabled precise control of the ion beam charge states. Further demonstrating its exceptional durability, a total of  $3.31 \times 10^{18}$  calcium ion particles was stripped of their charge with the intensity of 10  $\mu\text{A}$ . The graphite thin film disks exhibit great potential for next generation stripping foils by significantly extending stripping foil lifetimes under the highest intensity ion beam irradiation.

### INTRODUCTION

Charge stripping foils are used at accelerators to change the charge state of ion beams to appropriate values for acceleration. Due to its low density and excellent thermal and mechanical properties, carbon has been widely used for this purpose. Common carbon foils are amorphous carbon foils [1], Diamond-like carbon foils [2], CNT-carbon foil [3], and graphene foils [4]. Recent upgrades of accelerators, however, have led to higher ion beam intensities, which in turn have increased the demand for carbon charge stripping foils with extended lifetimes [3, 5-7]. To meet the requirement of long life charge stripping foils, the foils should have high thermal conductivity to avoid evaporation or damage from local beam heating. Other important characteristics include thermal stability, large thermal loading area, and mechanical robustness.

Kaneka has been a supplier of graphite sheets, in which the sheets are prepared from polyimide films annealed at temperatures of around 2900 °C [8]. The sheets exhibit approximately 4 times higher thermal conductivity relative to that of copper. Other key features include their high thermal stability of up to 2000 °C under vacuum, and high tensile strength of about 40 MPa. All of these features provide strong support for these sheets as candidates for highly durable charge stripping foils.

RIKEN Nishina Center has been testing these graphite sheets as charge stripping foils since the autumn of 2014 [9]. Graphite rotating disks were previously prepared for uranium ion charge stripping using two sheets of 35  $\mu\text{m}$  thickness.

As shown in Table 1, the rotating graphite disks withstood heavy ion radiation damage, which compared favorably to stationary amorphous carbon foils or glassy carbon disks. The graphite disks also showed higher ion beam current and charge stripping efficiency relative to the often-used Beryllium disk. Furthermore, the lifetime of this graphite disk was two times longer than that of Beryllium.

Table 1: History of Last Charge Stripping Foils in RIKEN Nishina Center

Charge Stripping Foils	Maximum Beam Intensity ( $\mu\text{A}$ )	Life Time (Charge)
Arizona Carbon Foil	2-3	$7.12 \times 10^{15}$ (71 <sup>+</sup> )
Beryllium Disk	12	$1 \times 10^{18}$ (64 <sup>+</sup> )
Glassy Carbon	12	Not measured
Graphite Sheet	15	$2.19 \times 10^{18}$ (64 <sup>+</sup> )

Recent efforts at Kaneka have led to new techniques that enabled the preparation of graphite thin films (GTF) with less than 10  $\mu\text{m}$  thickness. Precise thickness control was found to be attainable through careful control of the starting polyimide film thickness. The thickness of an appropriate charge stripping foil strongly depends on the type of ion and ion beam energy. For example, approximately 1.5  $\mu\text{m}$  ( $0.30 \text{ mg} \cdot \text{cm}^{-2}$ ) thickness is required for changing  $\text{U}^{35+}$  ion to  $\text{U}^{71+}$ . The aforementioned capability of precise and facile thickness control in GTF is therefore highly advantageous. Two aspects that required testing on GTFs prior to their application as charge stripping foils were their mechanical robustness, under high rotation speed, and lifetime. In this paper, we describe the rotating and charge stripping test of the GTFs, as well as measurements on their lifetime under high intensity ion beams irradiation.

### RESULTS AND DISCUSSIONS

#### *Preparation and Characterization of Graphite Thin Films [10]*

The thin polyimide films with thickness in the range of 2 – 25  $\mu\text{m}$ , as shown in Fig. 1 (a), were subjected to carbonization at temperatures of up to 1400 °C.

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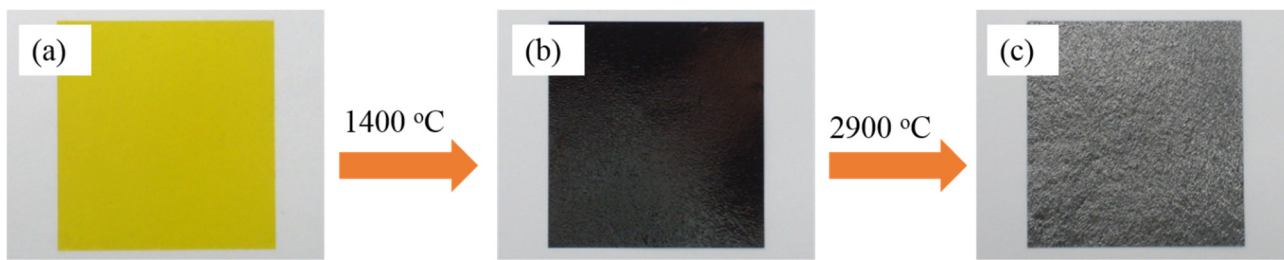


Figure 1: Preparation of graphite thin films (a) Kaneka polyimide film (b) Carbonized film (c) Graphite thin films (GTFs).

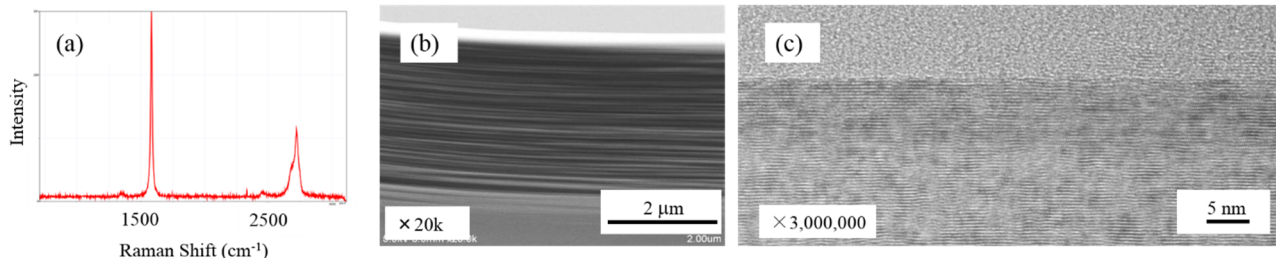


Figure 2: (a) Raman Spectra (b) Cross sectional SEM image and (c) TEM image of GTFs.

The resulting carbonized films shown in Fig. 1 (b) were then subjected to graphitization at 2900 °C to obtain GTFs with approximately 1 – 9.8  $\mu\text{m}$  (0.23 – 2.2  $\text{mg}\cdot\text{cm}^{-2}$ ) thickness (shown in Fig. 1 (c)). The thickness and area of GTFs was controllable by carefully selecting polyimide films with the appropriate thickness and area, respectively.

The Raman spectra of GTF with thickness of 2.1  $\mu\text{m}$  as shown in Fig. 2 (a) exhibited a symmetric peak at around 1585  $\text{cm}^{-1}$  and an asymmetric sharp peak at around 2717  $\text{cm}^{-1}$ . These features indicate that the GTF was composed of highly oriented graphite. The cross-sectional SEM image in Fig. 2 (b) and TEM image in Fig. 2 (c) both show highly oriented multilayer graphene layers in the depth profile.

### Rotation Test of Graphite Thin Film

A donut shaped GTF (0.23  $\text{mg}\cdot\text{cm}^{-2}$ ) with an outer diameter of 55mm in Fig. 3(a) was used to perform a rotating test using the RIBF accelerator at RIKEN Nishina Center. The thickness uniformity of charge stripping foils influences the fluctuation of beam intensity. Notably, thinner disks are preferentially rotated at higher speeds for preventing undesired fluctuation during charge stripping. The rotation speed of the disk was therefore set at 300 rpm at the beginning and was increased up to 1000 rpm to mimic

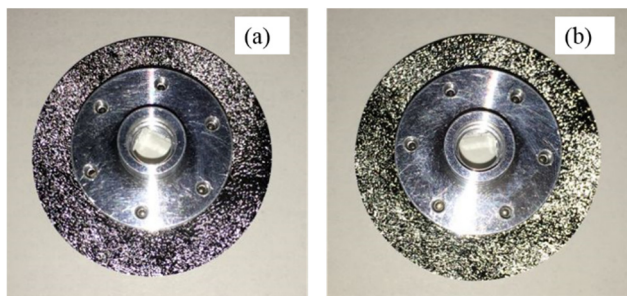


Figure 3: Optical images of the GTF disk (a) before and (b) after rotation test.

real conditions. The result of the test is shown in Fig. 3 (b), where the disk after testing exhibits no damage nor deformation.

### Charge Stripping Test with High Intensity Uranium Ion Beam [11]

Charge stripping tests were performed for GTF disks (0.45 – 2.2  $\text{mg}\cdot\text{cm}^{-2}$ ) and Kaneka graphite sheet disks (14  $\text{mg}\cdot\text{cm}^{-2}$ ). The rotation speed of disks was set at 300 rpm and was increased up to 1000 rpm. The incident beam used was  $\text{U}^{64+}$  with 10  $\mu\text{A}$  at a beam energy of 50 MeV/u and a beam diameter of about 4 – 5 mm; the beam intensity corresponds to a thermal load of 4.4 – 164 W. The charge distribution after the GTF disks was measured and shown in Fig. 4.

The mean charge states of the GTF disks with thickness of 0.45 (Red), 0.91 (Green), 2.2 (Blue) and 14 (Black)  $\text{mg}\cdot\text{cm}^{-2}$  were  $74^+$ ,  $78^+$ ,  $82^+$ , and  $87^+$  respectively. The thicker charge stripping foils provided a higher fraction and narrower charge states of uranium ion. Beam intensity fluctuation was not observed in the downstream in all GTF disks.

### Life Time Measurement at Calcium Ion Beam [11]

Disk lifetime is a key characterization parameter for stripping foils. A donut shaped GTF disk with an outer diameter of 110mm and a thickness of 2.2  $\text{mg}\cdot\text{cm}^{-2}$  was subjected to calcium ion beam. The disk was rotated at 300 rpm and the  $\text{Ca}^{16+}$  ion was stripped into  $\text{Ca}^{20+}$  at an incident energy of 45 MeV/nucleon. A total of  $3.31 \times 10^{18}$  calcium particles with an intensity of 10  $\mu\text{A}$  were irradiated on the GTF disk and the thermal load of the disk was 6.4 W. Fig.5 (b) shows the disk after irradiation. Slight footprints of the ion beam were observed near the outer edge, in addition to small deformations within the disk. Although a minor beam swing was observed, these deformations, nevertheless, did

not affect the beam intensity at the downstream. Collectively, these results indicated that GTF disks possess a long lifetime and the ability to strip the charges away from high intensity ion beams. Furthermore, the disk showed enough mechanical robustness under high speed rotation while being exposed to high intensity ion beam irradiation.

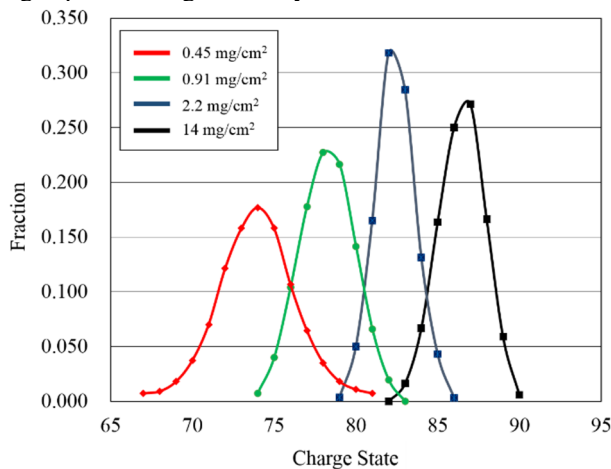


Figure 4: Charge distribution of  $^{238}\text{U}^{64+}$  ion beam using GTF disks.

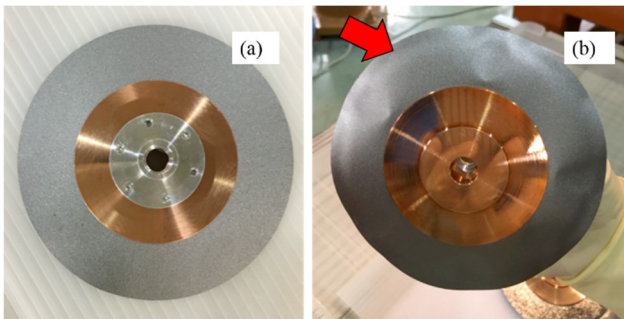


Figure 5: GTF disks (a) before and (b) after calcium ion beam irradiation.

## CONCLUSION

Graphite thin films were prepared through a simple two-step carbonization- graphitization process from polyimide films. Prepared GTFs were processed into rotating charge stripping disks and showed enough robustness against both high speed rotation and high intensity ion beam irradiation. Furthermore, GTF disks exhibited long lifetime. These results indicated that this GTF disks have great potential for charge stripping foils under highest intensity ion beam irradiation.

## REFERENCES

- [1] B. Lommel, W. Hartmann, B. Kindler, J. Klemm, J. Steiner “Preparation of self-supporting carbon thin films”, Nucl. Instr. Meth. Phys. Res. A 480, pp. 199-203, 2002.
- [2] G. Dollinger, P. Maier-Komor, C.M. Frey, E. Hammann, H.J. Körner, “New plant for fabricating carbon stripper foils by laser plasma ablation deposition”, Nucl. Instr. Meth. Phys. Res. A 328, pp. 168-172, 1993.
- [3] H. Hasebe *et al.*, “Development of a new foil compounded from carbon nanotubes and sputtered-deposition carbon”, J. Radioanal Nucl. Chem. vol. 299, pp. 1013-1018, 2014.
- [4] I. Pavlovsky and R.L. Fink, “Graphene stripper foils”, J. Vac. Sci. Technol., B 30, pp. 03D106-1, 2012.
- [5] W. Barth *et al.*, “Carbon stripper foils for high current heavy ion operation”, J. Radioanal Nucl. Chem. vol. 299, pp. 1047-1053, 2014.
- [6] K. Kupka *et al.*, “Intense heavy ion beam induced temperature effects in carbon-based stripper foils”, J. Radioanal Nucl. Chem. vol. 305, pp. 875-882, 2015.
- [7] S. Korenev *et al.*, “The use of graphene as stripper foils in Siemens Eclipse cyclotrons”, in Proc. Cyclotron2016, Zurich, Switzerland, Sep. 2016, paper TUP08, pp. 181-183.
- [8] Kaneka Corporation; <http://www.elecdiv.kaneka.co.jp/english/graphite/>
- [9] H. Hasebe *et al.*, “History of solid disk improvement for rotating charge stripper”, in 13th International Conference on Heavy Ion Accelerator Technology (HIAT2015), Sep. 2015, Tokyo, Japan, paper MOA1C01, pp. 17-19.
- [10] A. Tatami, M. Tachibana, M. Murakami, T. Yagi, M. Murakami “Preparation of multilayer graphene sheets and their applications for particle accelerators”, in 28th World Conference of the International Nuclear Target Development Society (INTDS 2016), Cape Town, South Africa, Nov. 2016, O-3 in press.
- [11] H. Hasebe *et al.*, “Developing of rotating graphite carbon disk stripper”, in 28th World Conference of the International Nuclear Target Development Society (INTDS 2016), Cape Town, South Africa, Nov. 2016, O-15 in press.