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Journal of Nuclear Materials 290–293 (2001) 291–294

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Detection of sputtered and evaporated carbon aggregates: relative and absolute electron ionization fragmentation yields

C. Mair [a,*], H. Deutsch [b], K. Becker [c], T.D. Märk [a,d], E. Vietzke [e]

[a] *Institut für Ionenphysik, Universität Innsbruck, Technikerstr. 25, A-6020 Innsbruck, Austria*
[b] *Institut für Physik, Ernst-Moritz-Arndt Universität, Domstr., D-17487 Greifswald, Germany*
[c] *Department of Physics and Engineering Physics, Stevens Institute of Technology, Hoboken, NJ 07030, USA*
[d] *Katedra fyziky plazmy, Univerzita Komenskehu, Mlynska dolina F2, SK-84215 Bratislava, Slovak Republic*
[e] *Institut für Plasmaphysik, Forschungszentrum Jülich, D-52425 Jülich, Germany*

Abstract

The present study is a first attempt to determine electron impact ionization efficiencies for C_2 and C_3. A novel method has been applied to obtain the partial cross-section values for the reactions $C_2 + e \rightarrow C^+, C_2 + e \rightarrow C_2^+$ and $C_3 + e \rightarrow C^+, C_3 + e \rightarrow C_2^+$ and $C_3 + e \rightarrow C_3^+$. The neutral target consisting of C, C_2 and C_3 is produced by thermal evaporation from a heated graphite sample and the neutral precursors in the subsequent ionization process can be distinguished by their different flight-time distributions acquired in the evaporation process. The partial ionization cross-section ratios obtained in this experiment have been calibrated with calculated absolute total ionization cross section curves of C_2 and C_3 using the Deutsch-Märk (DM) formalism.

Keywords: Sputtering; Particles; Ionization

1. Introduction

Recent studies in the field of thermonuclear fusion based on the magnetic confinement of high temperature plasmas have demonstrated that the conditions at the plasma periphery ('plasma edge') play an important role in achieving, sustaining and controlling the thermonuclear fusion [1–3]. Carbon materials are the most extensively used materials for plasma-facing components in present fusion devices. Carbon and its clusters are sputtered from the walls by energetic particles and enter the plasma as impurities. Eventually they are ionized in the plasma volume and finally they hit the walls again, following the electric and magnetic fields present. For models which describe the phenomena in the plasma edge region, it is essential to obtain a detailed knowledge about the impurity sources as well as the ionization cross-sections of these species [1–3]. The present study constitutes a first attempt to determine electron impact ionization efficiencies for C_2 and C_3 using a novel method to obtain for instance the partial cross-section ratios $\sigma(C^+/C_2)/\sigma(C_2^+/C_2)$ and $\sigma(C^+/C_3)/\sigma(C_2^+/C_3)$. Moreover, the partial ionization cross-section ratios obtained in this experiment have been calibrated with calculated absolute total ionization cross-section curves of C_2 and C_3 using the Deutsch-Märk (DM) formalism [4].

2. Experimental

The experimental setup is schematically shown in Fig. 1 (a detailed description is given in [5]). It consists of a primary chamber with a graphite sample mounted on a rotatable manipulator. The sample can be heated via resistive heating (typical temperatures used were

* Corresponding author. Tel.: +43-512 507 6243; fax: +43-512 507 2932.
E-mail addresses: Christian.M.Mair@uibk.ac.at (C. Mair), Tilmann.Maerk@uibk.ac.at (T.D. Märk).

0022-3115/01/$ - see front matter
PII: S0022-3115(00)00577-8

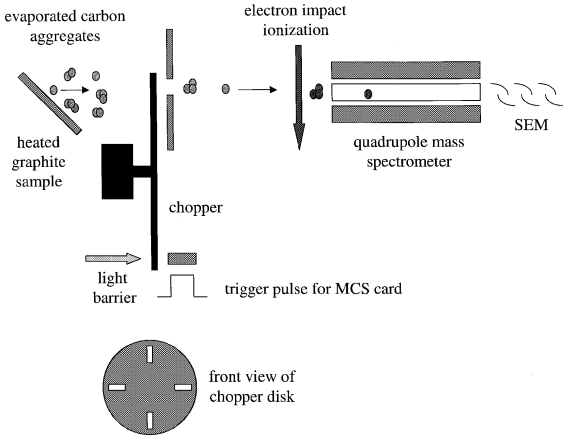

Fig. 1. Schematic view of the apparatus after [5].

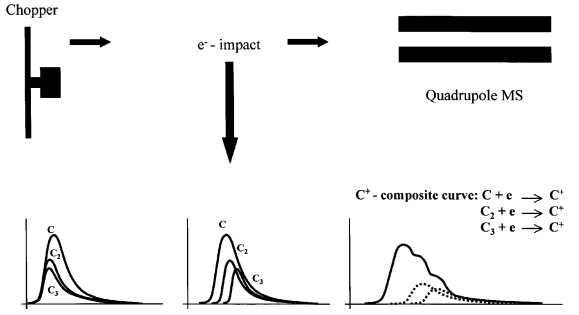

Fig. 2. Schematic view of the experimental technique.

2600 K), thus emitting by thermal evaporation besides carbon atoms also carbon aggregates such as C_2 and C_3. During the measurement the temperature is monitored by a quotient pyrometer system. The neutral beam of evaporated particles coming from the target is then chopped by a disk rotating with a frequency of 360 Hz. A light barrier creates the trigger pulse for the multichannel scaler card to finally record a flight-time correlated mass spectrum. Instead of heating, an ion gun can be used for sputtering the target material by bombarding the sample with a high-energy ion beam [5]. After a flight path of approximately 15 cm, the evaporated particles are ionized by an electron impact ion source, which is part of a differentially pumped line-of-sight quadrupole mass spectrometer. The ion signals are finally collected by a multichannel counting device (2 μs channel width) which directly records the total TOF spectra, the corresponding flight times including the flight time from the chopper to the ionization region (approximately 15 cm) and the flight time of the ionized particles flying through the quadrupole mass spectrometer. The flight lengths and times have been determined in earlier experiments [6]. The zero line of the TOF spectra was obtained by averaging the noise at large flight times.

As can be seen schematically in Fig. 2, depending on their original mass the neutral particles coming from the target (e.g., (C, C_2 and C_3) have different Maxwellian flight-time distributions. Moreover, the main difference in flight times will result from the distance between the chopper and the ion source (see above).

Therefore, if C_2 and C_3 break up into C^+ upon the electron impact ionization process, there should be an additional contribution to the C^+ distribution (originating from the reaction $C + e(C^+ + 2e)$ at the positions of the maxima of the flight-time distributions for the parent ions C_2^+ and C_3^+ (originating primarily from the reactions $C_2 + e \rightarrow C^+ + 2e$ and $C_3 + e \rightarrow C^+ + 2e$, respectively). This would then, as C^+ from C_2 and C^+ from C_3 are flight-time shifted, result in a broadening or in additional humps of the composite C^+ curve depending on the magnitude of the effect (see Fig. 2). The contribution of C_4 and larger clusters can be neglected safely in this analysis, because vapor pressures of these aggregates become of importance only for temperatures above 4000 K [7].

The amount of fragmentation can be evaluated in the following way: the experimental curves for C_2^+ and C_3^+ have to be fitted with the corresponding theoretical Maxwellian distributions, which are then in a kind of fitting procedure partly added to the C^+ distribution to obtain the best agreement with the measured C^+ distribution. Obviously, before this fitting procedure the ion signals of C_2^+ and C_3^+ have to be corrected for the different evaporation rates of the corresponding neutral precursors C_2 and C_3, respectively (ratio of measured evaporation rates of C, C_2 and C_3 species: 1:0.5:1.03 at $T = 2510$ K; evaporation rate $\propto p(T) * (MT)^{-0.5}$ with p being the partial pressure of the carbon species, M the molecular weight and T the temperature) [7].

The use and accuracy of a Maxwellian-fit was tested with He and Ne effusing from a heated gas cell at different temperatures at the position of the target, with the corresponding theoretical curves being very close to the experimental data (see Fig. 3).

3. Results and discussion

Fig. 4 shows as an example the flight-time distributions of the ions C^+, C_2^+ and C_3^+ produced by ionization with 200 eV electrons of the neutral particles evaporated from a graphite sample heated to 2620 K. Also shown in this figure as an example is the Maxwellian fit to the measured C^+ distribution demonstrating clearly that at longer flight times there is a deviation of the experimental points from the theoretical curve indicating additional sources of C^+ ion production. As these additional ion signals appear at the positions at around the maxima of the flight-time distributions of the two heavier carbon aggregates (as indicated in Fig. 4 by the

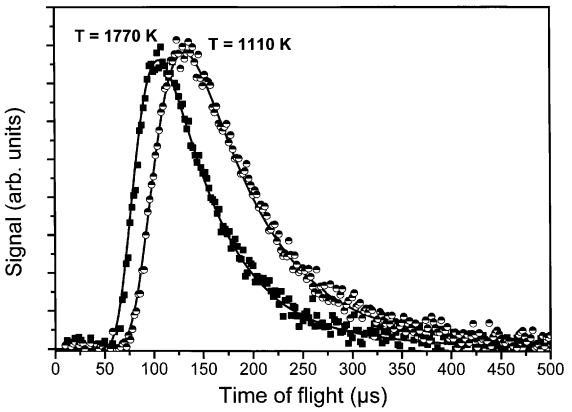

Fig. 3. Measured Ne^+ ion flight-time distributions for different temperatures (designated by full boxes for a temperature of 1770 K and with half circles for 1110 K) as compared to calculated Maxwellian distributions designated by full lines.

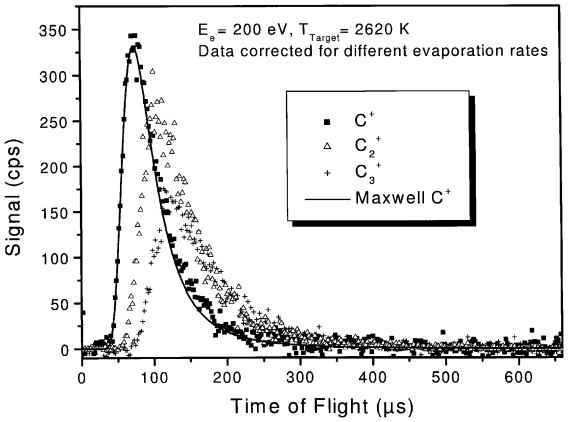

Fig. 4. Measured flight-time distributions of C^+, C_2^+ and C_3^+ as well as a Maxwellian fit to the C^+ distribution (without taking into account for this fit the dissociative ionization of C_2 and C_3 to C^+).

measured parent ion distributions) this deviation can be attributed to the additional C^+ ion production from C_2 and C_3 via dissociative ionization of these heavier carbon aggregates.

It is interesting to note that dissociative ionization of the two heavier carbon aggregates apparently does not lead to a large contribution to the experimentally determined C^+ distribution (Fig. 5 shows as an example the measured C^+ ion distribution and the fitted curve after adding corresponding contributions from the heavier carbon aggregates, see above). There can be several reasons for this small effect. On the one hand it indicates that electron impact induced fragmentation from C_2 and C_3 leading to the formation of C^+ is a relatively minor ionization channel. On the other hand, this may partly be due to the fact that the arrival time

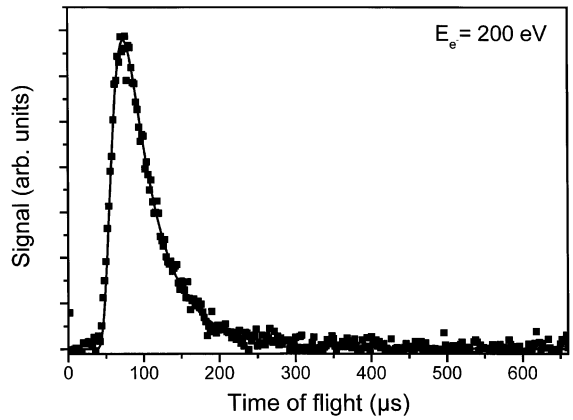

Fig. 5. Measured flight-time distribution of C^+ and Maxwellian fit to the data (designated by full line) taking into account for this fit possible contribution from the dissociative ionization of C_2 and C_3 giving C^+ (corresponding values given in Table 1).

differences for the C, C_2 and C_3 particles are rather small and thus make it more difficult to distinguish between the different contributions in the experimental C^+ distribution. Nevertheless, we tried to estimate an upper limit for the fragmentation of C_2 and C_3 for different electron energies from the corresponding fits (e.g. see Fig. 5). The values for the corresponding cross-section ratios are given in Table 1. It is interesting to note that cross-section ratios for other 'dimer' molecules show values of similar order of magnitude [8].

Calculations for the total ionization cross-sections for C_2 and C_3 have been carried out using the DM approach [9,10], a semiclassical formula, which is a combination of the classical binary encounter approximation, the Born–Bethe approximation and the additivity rule (for details and a review see [4]). As an example, the total electron impact ionization cross-sections for C_2 and C_3 are plotted in Fig. 6. Using these theoretical total cross-section values it is possible to convert the derived cross section ratios into absolute partial cross-sections. Obviously, due to the large statistical scatter in the data and the small fragmentation probability the resulting partial cross-sections are only rough estimates with rather large error bars. Nevertheless, as these are the first results concerning electron impact induced dissociative ionization of C_2 and C_3

Table 1
Upper limits for the cross-section ratios after the fitting procedure

Electron energy (eV)	$\sigma(C^+/C_2)/\sigma(C_2^+/C_2)$	$\sigma(C^+/C_3)/\sigma(C_3^+/C_3)$
70	0.01	0.06
200	0.01	0.09
250	0.08	0.02

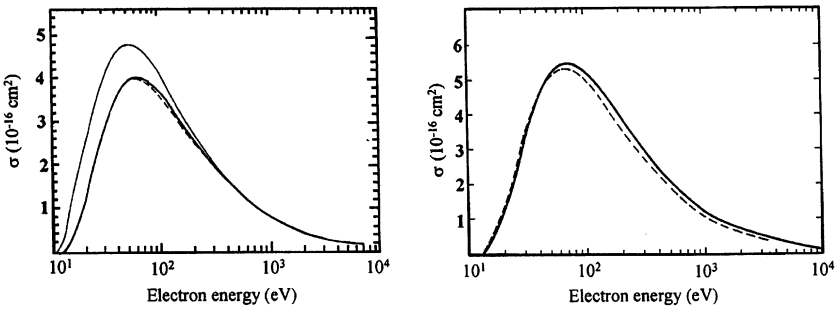

Fig. 6. Calculated absolute total electron impact ionization cross-sections for C_2 (left) and C_3 (right) as a function of electron energy using different input parameters (Mullikan analysis sets) for the DM formula used (for details see [4]).

these values may serve as a first guiding principle for corresponding modeling calculations.

4. Conclusion

From flight-time distributions of evaporated carbon and carbon aggregates we have determined estimates for the partial ionization cross sections for the reactions $C_2 + e \rightarrow C^+ + 2e$ and $C_3 + e \rightarrow C^+ + 2e$. The partial ionization cross-section ratios obtained in the experiment can be calibrated with calculated absolute cross-section curves for total electron impact ionization of C_2 and C_3. The effect of dissociative ionization in the case of C_2 and C_3 into C^+ has been found to be very small. On the one hand fragmentation of the carbon aggregates into C^+ upon the ionization process may be indeed only a minor reaction channel, on the other hand the arrival time differences in the evaporation experiment are rather small and thus make it difficult to distinguish between the different contributions. An upper limit has been given, but the associated error bars are relatively large due to the small effect and the large scatter of the data. Future experiments will be performed, where the carbon aggregates will be sputtered by energetic particles. In these measurements the arrival time differences of the particles will be larger and thus even a small effect should be more clearly distinguishable. There exist older measurements of this kind by Vietzke et al. [6], where a rough upper limit for the fragmentation has been estimated, but the new experiments will be performed with better statistics and higher reproducibility of the data.

Acknowledgements

This work has been carried out within the Association EURATOM-ÖAW and was partly supported by FWF, Wien, Austria. C.M. would like to thank the European Commission for supporting his visit to Jülich by a 'mobility grant'.

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