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Electron emission and molecular fragmentation during hydrogen and deuterium ion impact on carbon surfaces

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Abstract

Molecular fragmentation and electron emission during hydrogen ion impact on graphite surfaces has been investigated in the eV to keV impact energy region typical for fusion edge plasma conditions. As a target surface graphite tiles for the Tokamak experiment Tore Supra in CEA-Cadarache/France and highly oriented pyrolytic graphite (HOPG) have been used. For both surfaces studied, the experimentally observed threshold for electron emission is at about 50 eV/amu impact energy. Electron emission from the high conductivity side of the carbon tile is 15–20% less as compared to its low conductivity side, whereas results for HOPG are generally between these two cases. Deuterium and hydrogen ions are almost equally effective in liberating electrons from graphite when comparing results for the same impact velocity. Surface-induced dissociation of deuterium ions D_3^+ upon impact on Tore Supra graphite tiles, in the collision energy range of 20–100 eV, produced only atomic fragment ions D^+. The other possible fragment ion D_2^+ could not be observed.
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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 for producing, sustaining and controlling such plasma [1,2]. In order to understand and elucidate the role of the radiative and collisional processes in the plasma edge region, in particular their (i) influence on the plasma properties and dynamics and their (ii) use for controlling the plasma conditions, it is essential to have a detailed and quantitative knowledge on (i) the elementary processes in the gas phase [2], such as cross-sections, reaction rate coefficients etc., and on (ii) interaction of the gas phase particles with the plasma walls [1]. These data are necessary as input for edge plasma modeling and various diagnostic techniques.

The use of low-*Z* materials such as carbon, boron and beryllium has been beneficial in present large tokamaks [3]. In ITER design, graphite-based low-*Z* material is recommended for divertor plates and first wall protection for the initial operation phase, in order to minimize the risk of plasma contamination. The available databases on plasma-surface interaction (PSI) mainly include data on physical and chemical sputtering/erosion, material deposition and hydrogen/deuterium recycling. Less reli-

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able data are known for electron emission from wall materials under impact of energetic ions, atoms or electrons [4] and data on molecular fragmentation during molecular ion scattering are practically non-existing [1]. The last group of data has so far been limited to laboratory studies mainly involving H^+ and O^+ projectiles [5], and only recently extended to systems such as CH_3^+ and CH_4^+, but with conflicting results [6,7]. However, electron emission plays an important role in PSI by influencing the boundary plasma (e.g. via the sheath potential) and the intensity of plasma wall interaction accordingly. Basically, an extensive electron yield is expected to reduce the sheath potential. This in turn reduces the impact energy of ions and consequently the ion flux to the surface and the related sputtering yield. Molecular fragmentation and reactive collisions, on the other hand, may significantly change the ion and neutral compositions in the plasma edge due to production of neutrals and ions not a priori present in the plasma.

In view of the lack of available data, the main objective of this paper is to present measured data from a joint study of our laboratories for fusion-relevant ion species (e.g. H_n^+, D_n^+, $n = 1$–3) in the eV to keV energy region typical for fusion edge plasma conditions.

2. Ion-induced electron emission from carbon surfaces

As projectiles for our studies we have used atomic and molecular hydrogen and deuterium ions. These ions have been produced for the electron emission studies in a 5 GHz ECR ion source [8], extracted and accelerated to a few keV, mass-to-charge separated in a sector magnet and directed onto the carbon target situated in an UHV chamber (typically 10^{-8} Pa). To obtain impact energies typical for the plasma edge (several eV to several keV), the ions were decelerated in front of the target by means of a deceleration and focusing lens. Total electron yields γ (electrons per ion) were determined by means of current measurements of impinging ions and emitted electrons [9], and resulting total errors are estimated to about $\pm 7\%$. Data were obtained for normal incidence only.

As targets we studied not yet used specimens of carbon tiles which are applied on limiter surfaces of the tokamak experiment Tore Supra in CEA-Cadarache/France (manufacturer Soc. S.E.P. Bordeaux, France). Different surface orientations of these carbon compound tiles exhibit different thermal conductivity, i.e., a high conductivity of 230 W/m K due to carbon fibers in the direction of the thickness of the tiles and a low conductivity of 150 W/m K in the direction of the length of the tile. Since electron transport in a conductor is related to its thermal conductivity, some connection between thermal conductivity and the total electron yield can be expected. Consequently, we have investigated electron emission from the high (HC) and low (LC) thermal conductivity sides separately. For comparison we have also investigated a highly oriented pyrolytic graphite (HOPG) sample, constituting a standard material in surface science. Before taking data targets have been sputter-cleaned by impact of 3 keV Ar^+ ions and all investigations were carried out under ultra-high-vacuum (UHV) conditions (typically 10^{-8} Pa).

Fig. 1 shows measured total electron yields γ induced by impact of H^+ on the different carbon surfaces (C_L and C_H denote results for the low and high conductivity sides of the carbon tiles and HOPG refers to the highly oriented pyrolytic graphite sample). Fig. 2 compares measured total electron yields γ from HOPG induced by

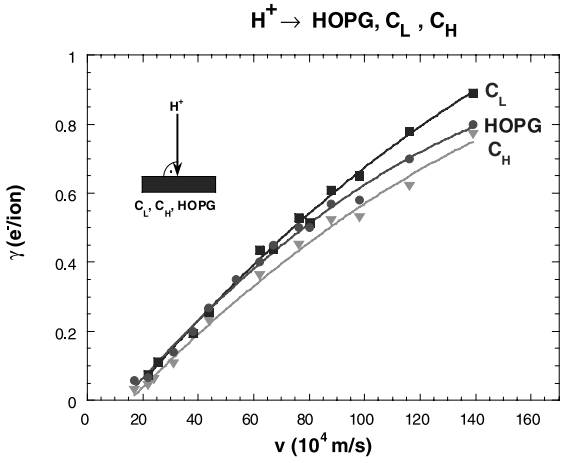

Fig. 1. Total electron yields vs. impact velocity v for impact of H^+ ions on different carbon tile surfaces (see text).

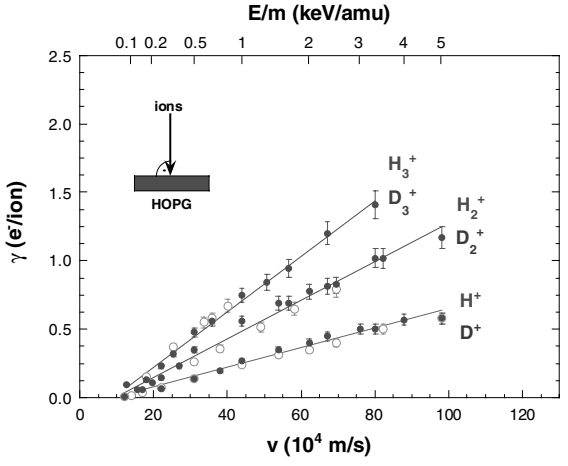

Fig. 2. Total electron yields vs. impact velocity v for impact of atomic and molecular hydrogen (full symbols) and deuterium (open symbols) ions on HOPG.

impact of atomic and molecular hydrogen ions H_n^+ ($n = 1$–3, full symbols) to results for atomic and molecular deuterium ions D_n^+ ($n = 1$–3, open symbols).

As a general trend we note that electron emission starts near an ion impact velocity threshold of about 10^5 m/s, corresponding to an impact energy per atomic mass unit of 50 eV/amu. For singly charged ions in the relevant impact energy range, ion induced electron emission is prominently caused by the kinetic energy of the projectile ions (kinetic electron emission – KE). For conducting targets the most prominent KE mechanism (at least for sufficiently fast projectiles) is momentum transfer in collisions of the projectile with quasi-free metal electrons ('eKE process') [10]. In such collisions, however, quasi-free electrons from the Fermi gas can only be ejected into vacuum above the so-called 'classical threshold' impact velocity $v_{\mathrm{th,e}}$ which depends on the respective work function W_ϕ and also on the Fermi velocity v_F (Fermi energy E_F) of the electrons in the solid [10–14] according to Eq. (1).

$$v_{\mathrm{th,e}} = \frac{1}{2} v_F \left(\sqrt{1 + \frac{W_\Phi}{E_F}} - 1 \right). \tag{1}$$

For carbon this classical threshold impact velocity is given by $v_{\mathrm{th,e}}(C) \approx 1.5 \times 10^5$ m/s or ≈120 eV/amu and thus somewhat higher than the experimentally obtained threshold value. This points to the importance of other KE mechanisms like electron promotion into continuum in projectile collisions with individual target atoms [14,15] and/or a newly proposed 'surface assisted KE' process [16], which is related to the abrupt change of the target electron density as seen by the impinging projectile ion. Recent calculation show indeed that surface-assisted KE can explain both the energy dependence and magnitude of the observed electron yields quite well [17].

As another result we note that about 15–20% less electrons are emitted from the high conductivity side of the carbon tiles as compared to the low conductivity side, whereas results for HOPG are generally situated between those for C_L and C_H (cf. Fig. 1). Reasons for the observed difference between results for HC- and LC-oriented carbon tiles should be related to the orientation of the carbon fibers. There might be differences in projectile penetration and stopping power, or an influence due to preferred directions for electron transport out of the solid.

Within our experimental errors deuterium and hydrogen ions are equally effective in liberating electrons from graphite when compared for the same impact velocity (cf. Fig. 2). For a fusion plasma, however, this means that at the same impact energy (edge temperature, sheath potential) deuterium ions will eject electrons less efficiently than their hydrogen counterparts.

Our data for protons are in very good agreement with measurements performed on bulk carbon samples by Large and Whitlock [18] and Cawthron [19], and somewhat lower than data obtained on self-supporting carbon foils [20]. To our knowledge no data for the other ion systems are available so far.

Although we did not perform measurements with fast neutral hydrogen atoms on graphite, we can evaluate a hypothetical H^0 yield (D^0 yield) under the assumption that the relation for equally fast ions

$$\gamma(H_n^+) = \gamma(H^+) + (n-1)\gamma(H^0), \tag{2}$$

holds for graphite in the same way as for our previously applied Au target [21]. Values for $\gamma(H^0)$ derived in this way are about 10% smaller than yields for equally fast protons.

3. Fragmentation of molecular ions

Ion surface (reactive) collisions is a research area which is rapidly growing in an effort to identify and explore new methods for characterizing gaseous ions and the nature of the surface and for elucidating plasma/wall interaction phenomena. Besides physical and chemical sputtering the following processes have been identified and investigated in the past years for collisions in the range of tens of eV laboratory energy:

(1) reflection,
(2) surface induced dissociation (SID),
(3) charge exchange reactions and
(4) surface induced reactions (SIR).

In order to allow a quantitative investigation of SID and SIR processes it is important to control and determine accurately the collision energy and to achieve energy spreads as small as possible. In a recent effort to achieve this situation we have constructed the tandem mass spectrometer set-up BESTOF (consisting of a B-sector field combined with an E-sector field, a Surface and a Time Of Flight mass spectrometer) [22,23] which allows the investigation of ion/surface reactions with high primary mass and energy resolution.

The ions produced in a Colutron ion source are extracted/focussed from the ion source region and accelerated to about 3 keV for mass and energy analysis by the double-focusing two-sector-field BE mass spectrometer. After passing the exit slit of the mass spectrometer, ions are refocused by an Einzel lens and the deceleration optics positioned in front of the graphite surface. The incident impact angle of the primary ions at the surface is usually kept at 45° and the scattering angle is fixed at 91°. The collision energy of ions impacting on the surface is defined by the potential difference between the ion source and the surface. The potential difference (hence, the collision energy) can be varied from 0 to about 2 keV with a typical resolution better than 200 meV.

A fraction of the secondary ions formed at the surface exits the shielded chamber through a 1 mm diameter orifice. These ions are then subjected to the pulsed extraction and acceleration field which initiates time-of-flight analysis of these ions. The mass selected ions are detected by a double stage multi-channelplate which is connected to a fast scaler (with a time resolution of 5 ns per channel) and a laboratory computer. Mass resolution has been improved steadily the past years and is to date approximately $m/\Delta m = 100$.

Measurements were first carried out for comparison (with earlier studies) and standardization with stainless steel and HOPG surfaces and then with fusion relevant carbon based materials, i.e., using the same carbon tile samples from Tore Supra as described in Section 2. The surface samples have been studied (i) either 'as is' under the vacuum conditions present in our machine (10^{-8} Pa) in which case the surface is assumed to be covered with adsorbed gases from the background or (ii) after cleaning the surface with an ion sputter gun. In each case we have not only measured the total reflected ion current, but also performed mass analysis of the product ions being produced by the impact of the projectile ions. Furthermore, the reactive interactions have been studied as a function of collision energy (thus yielding energy resolved mass spectra, ERMS) up to an energy of about 200 eV (see details given in [22,23]).

As mentioned in the introduction, so far laboratory studies in the frame of fusion oriented investigations concerning reactive interactions in the low energy regime below about 200 eV have been limited, i.e., involving only H^+ and O^+ projectiles ions [5]. Here we have extended these studies to molecular hydrogen ions for the first time including in a systematic fashion the species D_3^+ (see earlier studies on H_2^+ and H_3^+ given in [24]). Possible dissociation pathways (and its concomitant energetics [25]) for this ion are listed below:

$$D_3^+ \Rightarrow D^+ + D_2 \quad 4.70 \text{ eV} \tag{3}$$

$$D_3^+ \Rightarrow D + D_2^+ \quad 6.55 \text{ eV} \tag{4}$$

$$D_3^+ \Rightarrow D^+ + D + D \quad 9.21 \text{ eV} \tag{5}$$

In Fig. 3 the present measurements concerning the D_3^+ ion interacting with a sputter-cleaned Tore Supra carbon tile surface are summarized in form of an ERMS plot. The primary ion current of D_3^+ was 1–1.5 nA, whereas the secondary ion current was about three orders of magnitude less than the primary ion current. As a matter of fact due to the surprising results obtained we have repeated all of these measurements several times and under differing experimental conditions. The triatomic projectile ion D_3^+ quite unexpectedly produced only atomic fragment ion D^+, the other possible fragment ion D_2^+ could not be detected. This is in line with the energetics shown above which demonstrates that production of the deuterium ion plus one neutral D_2 is the energetically favored reaction; however, in the present experimental set-up it is not possible to measure these neutral fragments. This is in accordance with results concerning the angular dependence of backscattering of keV molecular hydrogen ions studied by Eckstein et al. [26] and photofragmentation of these molecular ions by Carrington and Kennedy [27]. The fragmentation pattern is very similar for HOPG and stainless steel target surfaces. From the energy value at the crossing point and the energetics of the dissociation pathway the energy transfer efficiency into the internal degrees of freedom during the collision process can be estimated to about 7%.

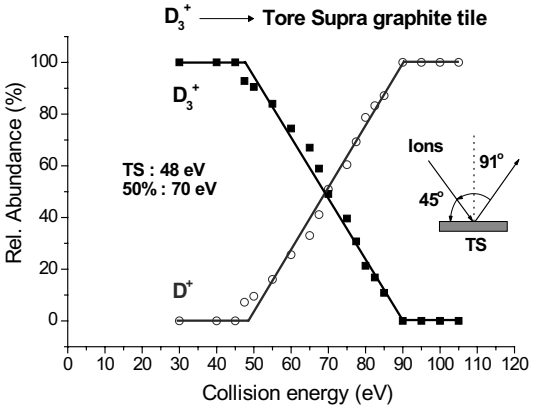

Fig. 3. ERMS plot for the interaction of D_3^+ with a graphite surface of a Tore Supra tile from Cadarache. TS: threshold energy, 50%: energy at crossing point.

It is clear that the outcome of these ion/surface collisions in terms of the reaction products will crucially influence the composition of the plasma in the plasma edge and may also be of importance in the H-retention process.

4. Summary and conclusions

As presented above, electron emission induced by impact of singly charged ions on carbon targets starts above an impact energy of about 50 eV/amu. Electron emission from the high conductivity side of the carbon tile is 15–20% less as compared to its low conductivity side, whereas results for HOPG are generally between these two cases. Deuterium and hydrogen ions are almost equally effective in liberating electrons from graphite when comparing results for the same impact velocity. At typical plasma sheath potentials of 200 V the total electron yield even for the light hydrogen ions is

still rather small (typically 0.2 electrons/ion) if compared to the electron induced emission yield. Singly charged ion-induced electron emission is therefore usually neglected in plasma simulation codes. However, ion-induced kinetic electron emission exhibits an impact angle Θ dependence of approximately $\gamma(\Theta) \approx \gamma_0 \cos^{-1}(\Theta)$ [10], which means that for more inclined incidence the yields may become much larger. The presence of an (oblique) magnetic field further complicates the situation due to the prompt return of emitted electrons within their first gyration inside the sheath [28]. A more definite answer for the role of (singly charged) ion-induced electron emission can only be gained from complex simulations. The data presented in this paper might serve as a valuable input for such efforts.

SID of deuterium ions D_3^+ on a graphite tile (as is and sputter-cleaned) from Tore Supra showed that in the collision energy range below 100 eV, which is relevant in fusion edge plasmas, only single-atom fragment ions D^+ were produced. Further studies on HD^+ and HD_2^+ are presently in progress and will contribute to a better understanding of the underlying processes in the scrape-off layer of fusion devices.

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