ELSEVIER

Journal of Nuclear Materials 313–316 (2003) 364–369

journal of nuclear materials

www.elsevier.com/locate/jnucmat

Chemical sputtering of carbon in Tore-Supra outboard pump limiter

A. Cambe [a], E. Gauthier [a,*], J. Hogan [b], J.M. Layet [c]

[a] *Association Euratom – CEA sur la Fusion Contrôlée, Centre d'Etude de Cadarache, F-13108 Saint Paul Lez Durance cedex, France*
[b] *Fusion energy Division, ORNL, P.O. Box 2009, Oak Ridge, TN 37831-8072, USA*
[c] *UMR 6633 CNRS-Université de Provence Physique des Interactions Ioniques et Moléculaires, centre Universitaire de Saint Jérôme service 241, avenue de l'Escadrille Normandie Niemen 13397, Marseille cedex 20, France*

Abstract

The use of a large amount of carbon in tokamaks induces two major problems mainly due to the chemical sputtering: erosion and tritium retention. In the Tore-Supra outboard pump limiter, the in situ chemical sputtering yield of the carbon neutralizer plate has been studied. Investigation of methane and heavier hydrocarbon (C_2D_x and C_3D_y) emission has been performed in ohmic and lower hybrid heated discharges, by means of mass spectrometry and optical spectroscopy. Simulation performed with a Monte Carlo code (BBQ) allows validation of the sputtering yield calculation method and show good agreement with experimental data.

PACS: 52.55.Fa
Keywords: Chemical sputtering; Erosion; Redeposition; Tokamak; Tore-Supra; Carbon

1. Introduction

Carbon materials are used widely as plasma facing components (PFC) in most existing magnetic fusion experiments and will be used, at least for specialized elements, in future reactors. These materials are considered because they have low atomic number (*Z*) and good thermo-mechanical properties. On the other hand, a critical point is that these materials have relatively large erosion during hydrogen bombardment from the fusion plasma. There are three different mechanisms of erosion: physical sputtering, chemical sputtering and radiation enhanced sublimation, inducing a large erosion rate on the target plate, consequently reducing the life time of the PFC, and leading to plasma dilution with impurities. This erosion is correlated with the production of co-deposited layers (redeposition of carbon with hydrogen isotopes) that tends to build a large tritium inventory in the wall, limiting the operation of the machine for safety reasons [1]. The use of divertors (axisymetric or ergodic) allows now the improvement in the control of the plasma edge parameters, limiting the power flux and the electron temperature [2,3]. As a result, the physical sputtering by D^+ ions can be reduced or even suppressed if the electron temperature is kept below the threshold energy. If we add to the limitation of the power flux the active cooling of the PFC, the RES can also be suppressed. Contrary to the two other mechanisms, the chemical sputtering does not show any threshold value either in particle energy or in surface temperature. Under these conditions, chemical sputtering will be probably the main mechanism of erosion for a carbon-based PFC. Various experimental studies have been undertaken in laboratory or on several fusion devices in order to develop theoretical models. Laboratory experiments show a decrease of the sputtering yield with increasing flux. A flux dependence of $\varphi^{-0.1}$ has been found by Roth et al. [4]. These experiments do not allow to reproduce the tokamak conditions (high flux). In tokamak, in situ measurements are difficult to

---

[*] Corresponding author. Tel.: +44-225 4204; fax: +44-225 4990.
E-mail address: e.gauthier@cea.fr (E. Gauthier).

0022-3115/03/$ - see front matter © 2003 Elsevier Science B.V. All rights reserved.
PII: S0022-3115(02)01583-0

achieve and are very dependent on the machines and of the diagnostics used (mass spectrometry or optical spectroscopy). Thus the incident particle flux (φ) dependence of $\varphi^{-\alpha}$ is found by different authors with $0.06 < \alpha < 1.25$ [5–10]. This leads to a high uncertainty on the PFC lifetime, specially for fusion reactors such as ITER. The purpose of this study is to validate the existing models, extrapolate the sputtering yield for higher flux (10^{24} m^{-2} s^{-1}), and to study the contribution of the hydrocarbons on chemical erosion in tokamak. At present, the chemical sputtering is attributed by many authors to the CD_4 emission, but at low energy the production of heavy hydrocarbons is expected to become dominant [11].

In the Tore-Supra outboard pump limiter (OPL), the in situ chemical sputtering yield of the carbon neutralizer plate has been studied. Investigation of methane and heavier hydrocarbon (C_2D_x and C_3D_y) emission has been performed in ohmic and lower hybrid (LH) heated discharges, by means of mass spectrometry and optical spectroscopy. In this study we will first focus on the variation of the sputtering yield of CD_4 and on the variation of the heavier hydrocarbons production versus the incident D^+ flux. Then, we will study the dependence of the sputtering yield of CD_4 versus surface temperature.

In parallel, simulation of plasma wall interaction in the OPL throat, have been performed with a Monte Carlo code (BBQ), in order to compare the experimental results with modelling.

2. Experimental set up

The Tore-Supra OPL and the diagnostics used in the experiment are presented in Fig. 1. There are five diagnostics used to provide information data useful for the study of chemical sputtering.

The Langmuir probes installed in the throat of the limiter measure the electronic density and temperature and also the incident flux of particles. In order to estimate the sputtering yield, we need to know the incident deuterium ions flux. This flux is calculated as described in Ref. [12], taking into account the effective charge of impurities, which are mainly carbon ions C^{4+}.

The partial pressures in the plenum of the different hydrocarbons are obtained from a mass spectrometer differentially pumped and absolutely calibrated. The partial pressure of methane is deduced from the mass 20 signal. The partial pressures respectively of C_2D_x and C_3D_y is deduced from mass 30 and 42 (the emission of C_2D_2 and C_3D_8 is neglected [13]) and extrapolated from ionisation coefficient and mass spectrum [14]. As the signal of masse 4 is saturated, we have approximated the partial pressure of D_2 by the total pressure [15]. The total pressure is measured on both parts of the limiter plenum (ionic side and electronic side) and above the pumps, using three capacitor gauges.

The surface temperature is monitored by an infrared detector, equipped with a filter peaked at 4 μm and with

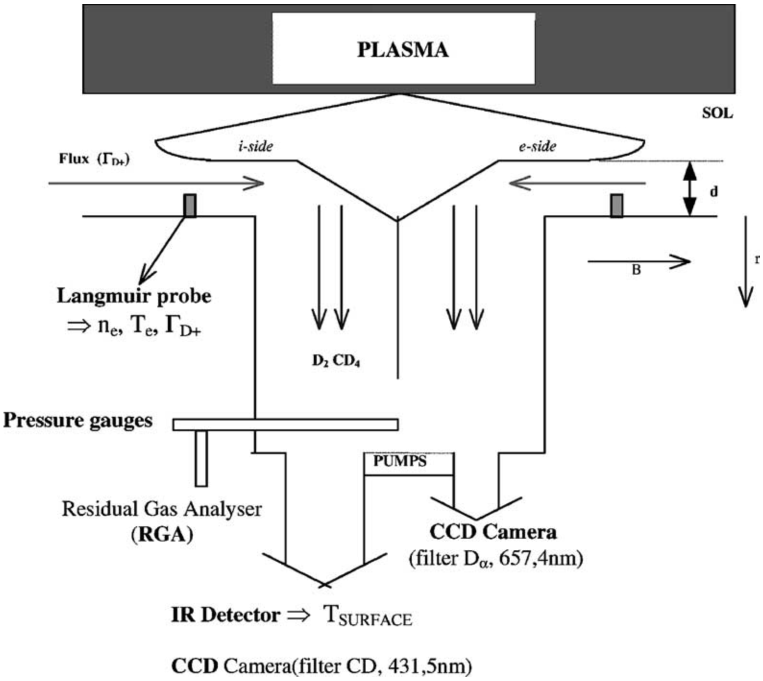

Fig. 1. Experimental set up.

0.1 μm bandwidth, looking at the neutralizer plate on the ion side.

Two CCD cameras are installed on both sides of the OPL. The ion side camera is equipped with an interference filter peaked at 431.5 nm with 0.6 nm bandwidth to measure the CD band emission, and the other one located on the electron side, allows to observe the Dα signal through the interference filter peaked at 657.4 nm with 0.9 nm bandwidth.

3. Chemical sputtering yield

The chemical erosion is quantified by a chemical sputtering yield, defined as the ratio of the impurities flux leaving a surface to the deuterium ions flux impinging the surface. For each hydrocarbon molecules emitted (C_nD_m) the chemical sputtering yield $Y_{C_nD_m}$ is defined by:

$$Y_{C_nD_m} = \frac{\varphi_{C_nD_m}}{\varphi_{D^+}},$$

where $\varphi_{C_nD_m}$ is the C_nD_m flux and φ_{D^+} the deuterium flux.

At the present time, the hydrocarbons flux leaving the neutralizer cannot be measured. Thus, the chemical sputtering yield has to be estimated by the partial pressure of the gas involved. In the case of the CD_4 emission and as described in Ref. [13], the CD_4 and the D^+ flux are expressed by the following relations:

$$\varphi_{CD_4} = P_{CD_4} V^{CD_4}_{\text{pumping}}$$

with P_{CD_4} the CD_4 partial pressure and $V^{CD_4}_{\text{pumping}}$ the CD_4 pumping speed.

$$\varphi_{D^+} = 2P_{D_2} V^{D_2}_{\text{pumping}}$$

with P_{D_2} the D_2 partial pressure and $V^{D_2}_{\text{pumping}}$ the D_2 pumping speed.

We can then, determine the D_2 pumping speed from the CD_4 pumping speed.

$$V^{CD_4}_{\text{pumping}} = V^{D_2}_{\text{pumping}} \sqrt{\frac{m_{D_2}}{m_{CD_4}}} \Rightarrow V^{CD_4}_{\text{pumping}} \cong 0.45 V^{D_2}_{\text{pumping}}.$$

Finally, the CD_4 emission yield is given by:

$$Y_{CD_4} \cong 0.23 \frac{P_{CD_4}}{P_{D_2}}.$$

4. Chemical sputtering yield variation versus deuterium flux

To investigate the variation of the chemical sputtering yield versus deuterium flux, a series of ohmic discharges (plasma current set at 1.4 MA and toroidal field at 3.3 T) in reproducible conditions of plasma wall interaction have been performed. The plasma was leaning on the OPL located at 3.12 m. The steady state line average density was varied shot by shot in order to construct a density scan. The deuterium flux increased from 10^{22} to 1.3×10^{23} $m^{-2}\,s^{-1}$. As the flux increased, the electronic density moved from 6×10^{17} to 4×10^{18} m^{-3}, and the electronic temperature decreased from 30 to 10 eV. The Fig. 2 represents the variation of the electronic temperature and density in the throat of the OPL versus the deuterium flux for each ohmic shots. Measurements have been taken during the current plateau of the discharges (between 6 and 7 s) where the flux can be considered as stationary. We can consider that during the time interval on which the measurements are made, the temperature does not change. So all the data have been taken at the same temperature 350 °C. This point is very important because for the ohmic shots, the chemical sputtering is dependent only on the flux variation and not on the surface temperature. The dependence on surface temperature will be investigated later.

It has been found that the production of chemical impurities increase with the deuterium ions flux. This increase depends on the type of hydrocarbons emitted. The CD_4 production versus deuterium flux follow the same relation that one established by Ruggiéri et al. [15] and is comparable to what is found on JT-60 [16]. This relation is:

$$P_{CD_4} \propto \varphi^{0.73},$$

where P_{CD_4} is the partial pressure of methane and φ is the deuterium ions flux.

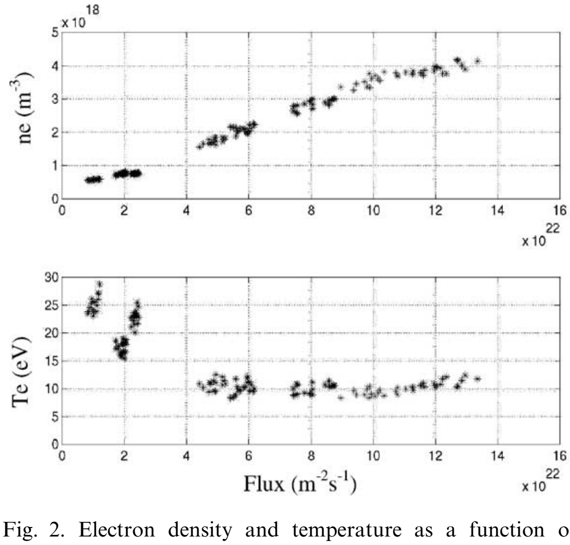

Fig. 2. Electron density and temperature as a function of deuterium ions flux in the throat of the OPL for each ohmic shots.

For heavier hydrocarbons the production is less important compared to the CD_4 production. It is two times lower for the C_2D_x and five to eight times lower for the C_3D_y. The C_2D_x production has almost the same flux dependence that CD_4 and the relation is also close to the one found on JT-60. The flux dependence is defined by:

$$P_{C_2D_x} \propto \varphi^{0.68},$$

where $P_{C_2D_x}$ is given by the signal of mass 30 and φ is the deuterium ions flux.

The increase of C_3D_y is lower than CD_4 and C_2D_x hydrocarbons and shows the following flux dependence:

$$P_{C_3D_y} \propto \varphi^{0.5},$$

where $P_{C_3D_y}$ is given by the signal of mass 42 and φ is the deuterium ions flux.

From this result, we can conclude that methane is the principal impurity emitted by chemical sputtering. But, in order to estimate the total chemical-sputtering yield, we have to take into account the total amount of carbon atoms. In this case, the carbon amount produced by the C_2D_x in this range of incident D^+ flux is equal to that produced by CD_4. Additionally, the carbon produced by the C_3D_y molecules is not negligible since it is only a factor 3 lower than the carbon produced by CD_4. In that way, C_2D_x and C_3D_y contribute significantly to the total chemical erosion yield and cannot be neglected. The carbon production versus incident flux is plotted in Fig. 3.

From the CD_4 production and the total pressure measurements, we are now able to investigate the variation of the CD_4 emission yield versus the incident deuterium ions flux. These measurements are made in the plenum of the OPL and not directly at the neutralizer surface, neglecting possible variation in ionisation and dissociation mean free path of CD_4. With this approximation, it appears that the CD_4 emission yield (Y_{CD_4}) decreases when the deuterium flux increases (Fig. 4), with the following flux dependence:

$$Y_{CD_4} \propto \varphi^{-0.23}.$$

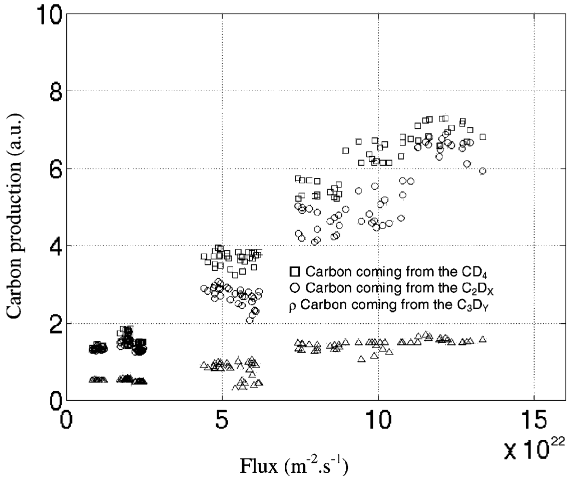

Fig. 3. Carbon production versus incident deuterium ions flux for each ohmic shots.

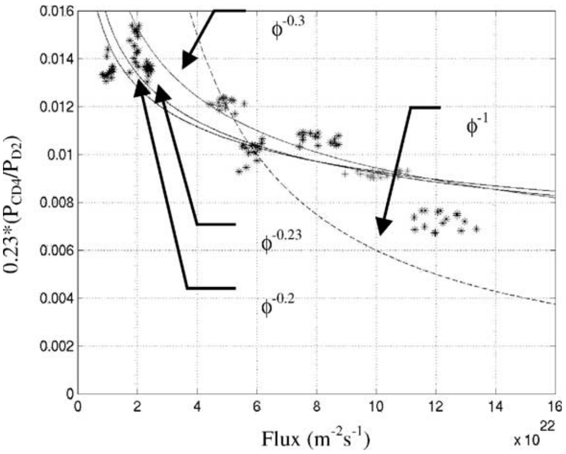

Fig. 4. CD_4 emission yield variation versus incident deuterium ions flux.

These results are similar to those found on JT-60 [16], and on the other hand it is far away from the theoretical model that presents a φ^{-1} flux dependence [17].

In order to compare the variation of the CD_4 emission yield at the neutralizer surface with the CD_4 emission yield measured in the plenum and then to study the effect of the dissociation process of CD_4, simulation of plasma wall interaction in the OPL throat, have been performed with a Monte Carlo code (BBQ). This code is described in detail in [18]. The input data, deuterium ions flux, electron temperature and density, are taken from the experimental data. The chemical sputtering yields are deduced from Roth's models [17], with and without the hydrogenation time taken into account. This process concerns hydrocarbon radicals formation by the hydrogenation of carbon atoms and depends on the incident flux. The Roth model does not take into account the formation of heavy hydrocarbons. The output data are the particles flux leaving the neutralizer, the CD_4 and D_2 partial pressures in the plenum, taking into account the ionisation and dissociation effects. We have compared the chemical sputtering yield at the surface, deduced from Roth's model (with or without the hydrogenation time) and the chemical sputtering yield calculated in the plenum from the CD_4 and D_2 partial pressure, determined by the BBQ code. The Fig. 5 show the proportionality between this two values. In this way, we demonstrate the validity of the technique used in the determination of the methane sputtering yield from the partial pressures measured in the plenum. At the highest

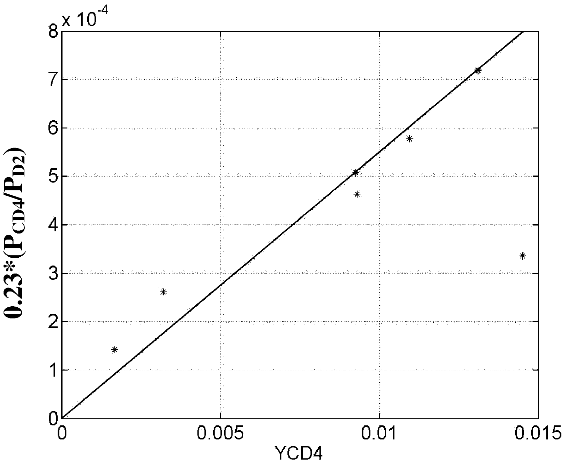

Fig. 5. CD_4 emission yield calculated from the CD_4 and D_2 partial pressures obtained in the plenum from the BBQ code versus the CD_4 emission yield deducted from Roth's model.

incident D^+ flux the theoretical model do not allow to reproduce the chemical sputtering (last point of the curve).

5. Chemical erosion yield variation versus surface temperature

In order to increase the surface temperature of the neutralizer, 2 MW LH heated discharges have been performed and the duration of the discharges has been increased up to 30 s. For such discharges, the electronic density increases from 1 to 1.5×10^{18} m^{-3} and the electronic temperature decreases from 40 to 20 eV during the LH power injection. At the end of the discharge, the increase of the electronic density is attributed to the impurities production. At the end of the power injection the temperature reaches more than 550 °C (20 s after the start of the shot).

At the same time, the CD_4, C_2D_x, C_3D_y partial pressures and the total pressure increase and particularly when the surface temperature exceeds 450 °C (15 s after the start of the shot). Between 15 and 20 s the CD_4 partial pressure is multiplied by 2, the C_2D_x partial pressure by 1.6 and the C_3D_y by 1.2. The CD_4 partial pressure rise, measured by mass spectrometry is correlated to the CD band emission evolution given by optical spectroscopy (Fig. 6). Thus the CD_4 production (CD signal) is proportional to the CD_4 partial pressure.

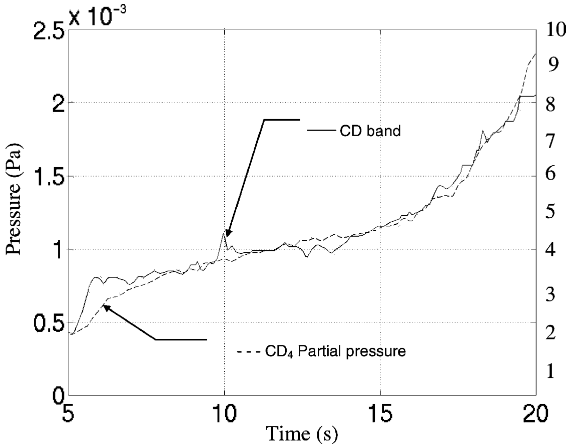

Fig. 6. CD signal (u.a.) and CD_4 partial pressure (Pa) versus time for a 2 MW LH heated discharge.

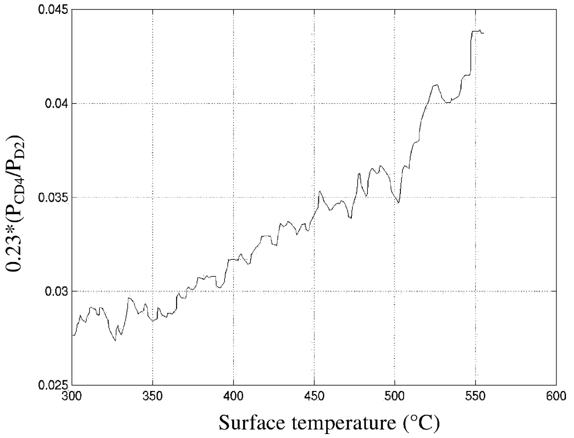

Fig. 7. CD_4 emission yield calculated from the CD_4 and D_2 partial pressures versus surface temperature.

For a surface temperature below 450 °C and if the hydrogenation time is not taken into account, as for the ohmic discharges, the CD_4 emission yield deduce from Roth's model and the one calculated from the CD_4 and D_2 partial pressures determined by BBQ are proportional. In that way, the experimental CD_4 emission yield is measured in the plenum, and as predicted by the model [19], increases with the surface temperature (Fig. 7). This result is confirmed by the simulations performed by the code BBQ. The surface temperature variation is similar with the one find with Roth's model in the case where the hydrogenation time is not taken into account (Fig. 8).

6. Conclusion

The investigation of the chemical sputtering performed on the OPL show that the impurities production (CD_4, C_2D_x, C_3D_y) increase with incident deuterium flux in the range of 1×10^{22}–16×10^{22} $m^{-2}\,s^{-1}$. CD_4 and C_2D_x have the same flux evolution, and the carbon production from the C_2D_x is as important as the one

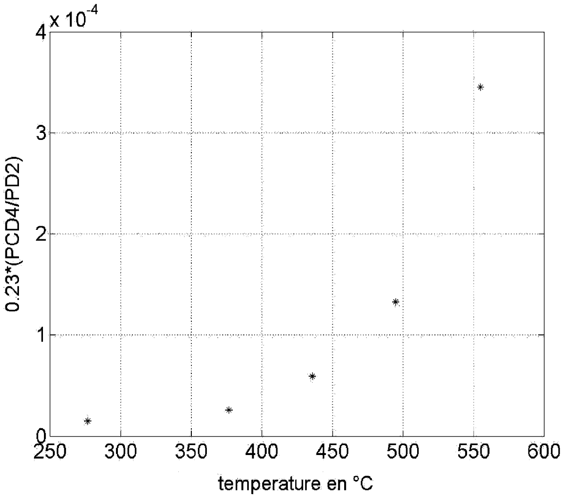

Fig. 8. Ratio of the CD_4 and D_2 partial pressure versus surface temperature.

produced via the CD_4. The contribution of the C_3D_y to the carbon production is significant since it is only a factor 3 lower that for CD_4. The variation of the CD_4 emission yield (Y_{CD_4}), measured in the plenum of the OPL, versus incident deuterium flux has been found to be in the form of $Y_{CD_4} \propto \varphi^{-\alpha}$ with $0.2 < \alpha < 0.3$. Y_{CD_4} increase with surface temperature, and specially from 450 °C.

Numerical simulations have been performed with a Monte Carlo code BBQ, using Roth's model, without the hydrogenation time. These simulations allow the validation of the method of calculation of the CD_4 emission yield in the plenum, showing that the CD_4 emission yield simulate at the neutralizer surface is proportional to the CD_4 emission yield calculated from the CD_4 and D_2 partial pressure in the plenum. This proportionality is verified except at the highest values of deuterium flux and high surface temperature. The simulation taking Roth's model without hydrogenation time shows good agreement with experimental data concerning the temperature variation.

References

[1] G. Federici et al., J. Nucl. Mater. 266–269 (1999) 14.

[2] B. Meslin et al., J. Nucl. Mater 266–269 (1999) 318.

[3] C.S. Pitcher, P.C. Stangeby, Plasma Phys. Control. Fusion. 39 (1997) 779.

[4] J. Roth, E. Vietzke, A.A. Haasz, Atomic and Plasma-material Interaction Data for Fusion (supplement to Nucl. Fusion.), vol. 1, IAEA, Vienna, 1991, p. 63.

[5] D.G. Whyte et al., Nucl. Fusion. 41 (2001) 47.

[6] M.F. Stamp et al., J. Nucl. Mater. 290–293 (2001) 321.

[7] A. Kallenbach et al., J. Nucl. Mater. 266–269 (1999).

[8] A. Pospieszczyk et al., 22nd EPS Bournemouth, 1995, p. II-309.

[9] P. Kornejew et al., Phys. Scr. T 91 (2001) 29.

[10] R.D. Monk et al., Phys. Scr. T 81 (1999) 54.

[11] A.A. Haasz, J.W. Davis, J. Nucl. Mater. 175 (1990) 84.

[12] T. Loarer et al., J. Nucl. Mater. 196–198 (1992) 1078.

[13] A. Cambe, thèse, Université de Provence, Marseille, 2002.

[14] A. Cornu, R. Masson, compilation of mass spectral data, Universitaires de France, 1996.

[15] R. Ruggiéri, E. Gauthier, J. Hogan, J.M. Layet, T. Loarer, J. Nucl. Mater. 266–269 (1999) 660.

[16] N. Hosogane et al., J. Nucl. Mater. 220–222 (1995) 415.

[17] J. Roth, J. Nucl. Mater. 266–269 (1999) 51.

[18] J. Hogan, C. Klepper, J. Harris, et al., Proc. 16th Int. Conf. on Plasma Physics and Control. Fusion, Montreal, IAEA, Vienna, 1996.

[19] J. Roth, C. Garcia-Rosales, Nucl. Fusion. 36 (12) (1996) 1647.