

STICKING COEFFICIENT AND SIMS OF HYDROCARBONS ON FUSION RELEVANT PLASMA – SPRAYED TUNGSTEN SURFACES

W. Schustereder^{1,}, B. Rasul², N. Endstrasser², V. Grill², P. Scheier², T.D. Märk²*

¹ Max-Planck-Institut für Plasmaphysik, EURATOM Assoziation, Boltzmannstraße 2, D-85748 Garching, Germany

² Institut für Ionenphysik und Angewandte Physik, Universität Innsbruck, Technikerstr. 25, A-6020 Innsbruck, Austria

Abstract: In this work we concentrate on the quantification of the sticking coefficient of CD_3^+ on fusion relevant plasma-sprayed tungsten (PSW) surfaces in the collision energy range from about 0 up to 100 eV. PSW samples were cut from ASDEX Upgrade tiles and both untreated and electropolished surfaces are investigated. A collision energy, E_{coll} , selected CD_3^+ ion beam deposits hydrocarbon layers onto the target surfaces which are ex-situ analysed by Nuclear Reaction Analysis (NRA) via $\text{D}({}^3\text{He}, \text{p}){}^4\text{He}$ at 800 kV and 2.5 MeV, respectively. The sticking coefficient, S , for deuterium has been found to be collision energy dependent, decreasing with increasing energy. It is in the order of $S \sim 0.4$ for PSW and $S \sim 0.1$ for electro polished PSW at $E_{\text{coll}} = 10$ eV. Secondary ion mass spectra of charged particles are recorded after ion-surface collisions with the polished PSW surface for collision energies up to 100 eV. While at very low collision energies below about 10 eV only simple reflection of CD_3^+ is observed, at higher collision energies both fragmentation of the projectile and fragmentation of ion-surface reaction products are observed. Concerning the stability of the layers we prove that hydrogen exchange plays a role on the time scale of weeks.

PACS: 34.50.Dy

Keywords: sticking coefficient, hydrocarbons, tungsten, plasma-sprayed tungsten

Corresponding Author Address: Dr. Werner Schustereder, Boltzmannstr. 2, 85748 Garching, Germany, Phone: +49 89 - 3299 1468, Fax: +49 89 - 3299 1212

Corresponding Author e-mail: Werner.Schustereder@ipp.mpg.de

1. Introduction

One of the key issues regarding ITER operation is the tritium accumulation at the first wall materials. This fuel retention effect is of particular interest both for the fuel balance and radiation safety, as ITER is only allowed to accumulate 350 g tritium inventory. Three materials are foreseen to be used for plasma-facing materials (PFMs) in ITER: Beryllium in the main chamber, and carbon and tungsten in the divertor region. Fuel retention in ITER is expected to be dominated by erosion of the CFC (carbon fiber composite) target and subsequent co-deposition of tritium in hydrocarbon layers [1]. Retention of fuel in metallic materials is known to be much lower than in hydrocarbon deposits [2], therefore, co-deposition of tritium in hydrocarbon layers is likely to dominate the retention of fuel in ITER. This can lead to a serious restriction of ITER operations with a carbon divertor target due to the restricted tritium accumulation. The net observed hydrocarbon co-deposition is the result of the balance of many complex processes and, thus, predictions of fuel retention in ITER are carried out with sophisticated models of the erosion/redeposition balance [3]. These models require as input the physical parameters that determine the reflection properties of the hydrocarbon radicals as they interact with the surface, namely their sticking probability and their probability of transformation into other molecular species. Although little is known on these properties they are expected to have a significant influence on the magnitude and spatial distribution on the final hydrocarbon deposition and tritium retention in ITER [4, 5].

In the current design of ITER the area covered with tungsten is 140 m² [1]. A technical feasible and cost effective method for covering such a large area with tungsten would be plasma spraying of tungsten on some feasible substrate. Tungsten coated graphite tiles were used at different regions (divertor strike points, central column, auxiliary limiters, etc.) of the tokamak experiment ASDEX Upgrade in several experimental campaigns [6, 7]. Plasma spraying of tungsten turned out to be a suitable method of tungsten layer deposition for tokamak application [8]. The advantages of plasma spraying in comparison with other

deposition methods are the homogeneously distributed voids, which stop crack propagation under high heat loads.

The objective of this paper is to determine the reflection properties of CD_3^+ ions impinging on a plasma-sprayed tungsten (PSW) sample as used in the divertor region of ASDEX Upgrade. We investigate the sticking coefficient and the secondary ion mass spectra as a function of the incident energy from about 0 eV to 100 eV which covers well the energy range of hydrocarbons expected in the ITER divertor ($E \sim 5 - 25$ eV [9]).

2. Experimental

Plasma-sprayed tungsten (PSW) samples were cut from the front surface of an unused ASDEX Upgrade tile. The ASDEX Upgrade tiles consist of fine grain graphite with dimensions 160x80x30 mm, of which the front surface is covered with 500 μm tungsten deposited by high-density plasma spraying. To prevent carbon diffusion and subsequent tungsten carbide formation an intermediate layer of 10 μm thick Re produced by physical vapor deposition is placed between carbon and tungsten. The front surface of the tiles was cut to the dimensions 10x6x1 mm in order to fit on the probe holder of the mass spectrometer used for deposition of CD_3^+ . For comparison and due to the expectation of better reflection properties of secondary ions also electro polished PSW tiles were investigated.

The tandem mass spectrometer system BESTOF (described in detail in [10]) was used to deposit a quantified amount of CD_3^+ at certain well-defined energies on the PSW tiles. In a Nier-Type ion source CD_3^+ was produced from CD_4 vapour by electron impact ionization at an ion source pressure of $p_{\text{IS}} = 6 \cdot 10^{-6}$ torr and an electron energy of $E_e = 100$ V. The ions are extracted and accelerated for mass- and energy-analysis by a magnetic and electric sector field. The selected beam of CD_3^+ ions is refocused in front of the PSW surface at different collision energies and a typical ion current of $I = 70$ pA is measured by a Faraday cup at the exit of the first mass spectrometer. The collision energy of the incident ions is selected by the

difference between potential of the ion source and the surface and the distribution of the collision energy was in these experiments FWHM ~ 0.7 eV. These ions are undergoing elastic and inelastic collisions at an incident angle of 45° relative to the surface normal within an elliptical beamspot size of $\sim 1 \times 1.5$ mm. A factor of approximately 3 is lost in the deceleration optics, consequently a CD_3^+ ion beam of $I \sim 25$ pA interacts with the surface. The time for hydrocarbon deposition at each collision energy ($E_{\text{coll}} = 0.2, 10, 20, 30, 50, 100$ eV) is chosen to be 10 h, allowing a maximum film growth of some monolayers. On each sample surface deposition has been carried out only at two distinct energies are measured at well separated locations in order to avoid overlapping of the beamspots as well as edge effects of the surfaces. A fraction of the incident ions keeps sticking on the PSW surface, the reaction products coming off the surface are collected by a Time-Of-Flight mass spectrometer at 91° relative to the incoming ion beam. After exposure the probes are retrieved and transferred to IPP Garching for analysis. Thereby the samples are exposed to air for about 14 hours.

The areal densities of deuterium are measured by nuclear reaction analysis (NRA) using the reaction $\text{D}({}^3\text{He}, \text{p}){}^4\text{He}$ at 800 kV and 2.5 MeV, respectively. An ion charge of at least $100 \mu\text{C}$ is collected for each spectrum. Quantification is achieved by comparison to reference samples with a-C:D/Si layers of known thickness and composition.

3. Results

3.1. Interaction of D with PSW

After exposure to the CD_3^+ ion beam the sample is retrieved and transported from Innsbruck to IPP Garching. Thereby the PSW surface is exposed to ambient air for about 14 h. In the course of analysing the areal density of deuterium by NRA, the shape of the ion beam spot is determined to be elliptical with an area of approximately 2 mm^2 . At a distance of 4 mm to the beam spot centre no deuterium is detected, proving the purity of the sample surface. Taking an ion current of $I \sim 25$ pA the deuterium ion flux is $2.3 \cdot 10^{10} \text{ D cm}^{-2} \text{ s}^{-1}$ and after the

deposition time of 10 h a deuterium fluence of $8.4 \cdot 10^{14} \text{ Dcm}^{-2}$ is deposited onto the target. Comparison with the areal density of deuterium found by NRA yields an energy dependent sticking coefficient $S \sim 0.2 - 0.4$, see Fig. 2. The maximum value $S \sim 0.4$ is measured at a collision energy $E_{\text{coll}} = 10 \text{ eV}$ and with increasing energy the sticking coefficient decreases. At a very low collision energy ($E_{\text{coll}} = \sim 0.2 \text{ eV}$) an exception to that tendency is observed, which can be due to the uncertainty of the zero-point of the collision energy, i.e. at such low energies part of the ion beam is expected to leave the surface region without even hitting the target.

It is along with this high sticking coefficient probably due to the rough PSW surface that only marginal amounts of secondary ions can be detected by TOF mass spectrometry after the ion-surface collisions.

3.2. Interaction of D with electro polished PSW

For comparison the same measurements are carried out under similar experimental conditions with an electro polished PSW surface. This electro polished surface was chosen due to the higher reflectivity for secondary ions expected for the smoother surface. In the course of the transport and before the NRA measurements the treated sample was exposed to air for about 5 days and after 35 days the analysis was repeated. We observe during this time span of about 30 days a decrease of D areal density by a factor of 2.3 at the 10 eV spot and of 2.6 at the spot sampling energies from 10 – 40 eV. Consequently hydrogen exchange plays a role on the time scale of several days and weeks, but it is not crucial if the transport of samples takes place within several hours. The sticking coefficient for this surface is found to be $S \sim 0.1$ for $E_{\text{coll}} = 10 \text{ eV}$ and $S \sim 0.05$ for energies in the range from 20 – 40 eV, see Fig. 2. This is about 4 times less than for the non-polished PSW surface, but the tendency of decreasing sticking probability with increasing collision energy is observed for both surfaces.

Due to the smoothing of the surface by electro polishing a larger amount of product ions is detected after ion-surface collision.

The collision energy resolved mass spectra of the secondary ions after collision of CD_3^+ ions onto the electro polished PSW surface at different energies can be seen in Fig. 3. By doing an auxiliary experiment with an Ar^+ beam, we can see that the CD_3^+ ion beam secondary ion spectra are dominated by chemical sputtering products, i.e. secondary ions produced by charge transfer ionization of surface constituents. A minor contribution of exchange reaction can be observed in the spectra at low collision energies, i.e. CD_3H^+ with subsequent fragmentation to CD_2H^+ . By comparing Fig. 2 and Fig. 3 we can see that the decrease of sticking coefficient with higher collision energies seems to be correlated with an increase of secondary ion yield.

4. Discussion and Outlook

We presented first successful measurements on the sticking coefficient S of CD_3^+ on PSW and electro polished PSW, respectively, in the energy range from 0 – 100 eV. S is energy dependent, decreases with increasing energy and has its maximum around $E_{\text{coll}} = 10$ eV with $S \sim 0.4$ for PSW and $S \sim 0.1$ for electro polished PSW. Secondary ions are more abundant using the smoother electro polished PSW surface. Hydrogen exchange is observed to be a relevant process under air-atmosphere on a time scale of several days and weeks.

We are aware of the fact that the ion current measurement taking place at the Faraday cup includes a significant uncertainty for the quantification process of the sticking coefficient. A direct current measurement on the surface is planned to be installed in the next step of upgrading the collision chamber of BESTOF. We plan to extend our investigations both to higher hydrocarbons as well as to polycrystalline tungsten, beryllium and pyrolytic graphite surfaces. Furthermore also the use of different ion sources may be considered producing projectile ions with different initial internal energy.

Analysis of deposited layers may be extended to ellipsometry (when using a Si substrate). When depositing more material also the hardness of the films will be of interest.

Acknowledgements

This work, supported by the European Communities under the contract of Association between EURATOM/ÖAW and EURATOM/IPP, was carried out within the framework of the European Fusion Development Agreement. The views and opinions expressed herein do not necessarily reflect those of the European Commission.

References

- [1] Federici, G., et al., Nucl. Fusion **41R** (2001) 1967.
- [2] K. Krieger, H. Maier and R. Neu, ASDEX Upgrade Team, J. Nucl. Mat. **266-269** (1999) 207.
- [3] D. Naujoks, D. Coster, H. Kastelewicz and R. Schneider, J. Nucl. Mater. **266-269** (1999) 360.
- [4] J. N. Brooks, R. Causey, G. Federici and D. N. Ruzic, J. Nucl. Mater. **243-245** (1997) 294.
- [5] Kirschner, A., et al., Proc. 17th PSI Conference, Hefei, China, 2006. To be published in J. Nucl. Mater.
- [6] Mayer, H., Kötterl, S., Krieger, K., Neu, R., Balden, M., ASDEX Upgrade-Team, J. Nucl. Mater. **258-263** (1998), 921.
- [7] Neu, R., Rohde, V., Geier, A., Krieger, K., Maier, H., Bolshukhin, D., Kallenbach, A., Pugno, R., Schmidtman, K., Zarrabian, M., ASDEX Upgrade Team, J. Nucl. Mater. **290-293** (2001), 206.
- [8] Deschka, S., García-Rosales, C., Hohenauer, W., Duwe, R., Gauthier, E., Linke, J., Lochter, M., Malléner, W., Plöchl, L., Rödhammer, P. and Salito, A., J. Nucl. Mater. **233-237** (1996), 645.
- [9] R. Dux, A. Herrmann, A. Kallenbach, R. Neu, J. Neuhauser, H. Maier, R. Pugno, T. Pütterich, V. Rohde and ASDEX Upgrade Team, J. Nucl. Mater. **337-339** (2005), 852.
- [10] C. Mair, T. Fiegele, F. Biasioli, R. Wörgötter, V. Grill, M. Lezius, T.D. Märk, Plasma Source Sci. Technol. **8** (1999) 191.

Figure captions

Fig. 1: Schematic of the double-focussing mass spectrometer BESTOF. A mass and energy analyzed CD_3^+ ion beam interact with a PSW surface, product ions are detected by a TOF mass spectrometer. The surface layers are analyzed ex-situ by NRA.

Fig. 2: The sticking coefficient S of deuterium on PSW (squares) and on electro polished PSW (circles). The line represents an average value for the energies of 20, 30 and 40 eV.

Fig. 3: Collision energy resolved mass spectra for ion surface collisions of CD_3^+ cations with an electro polished PSW surface for collision energies up to 100 eV.

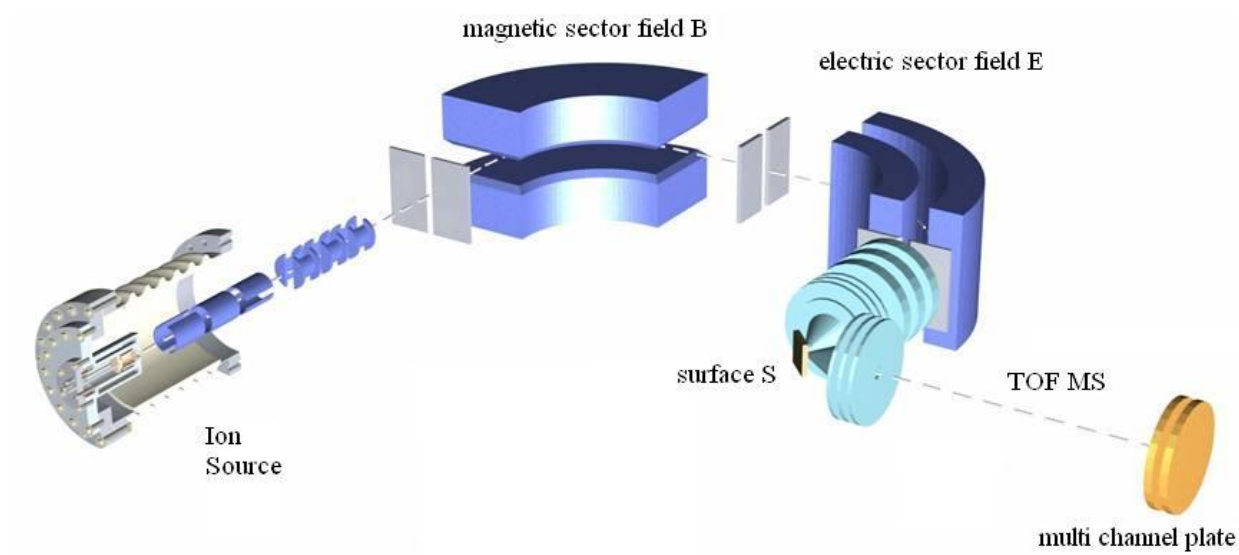
Figures

Fig. 1

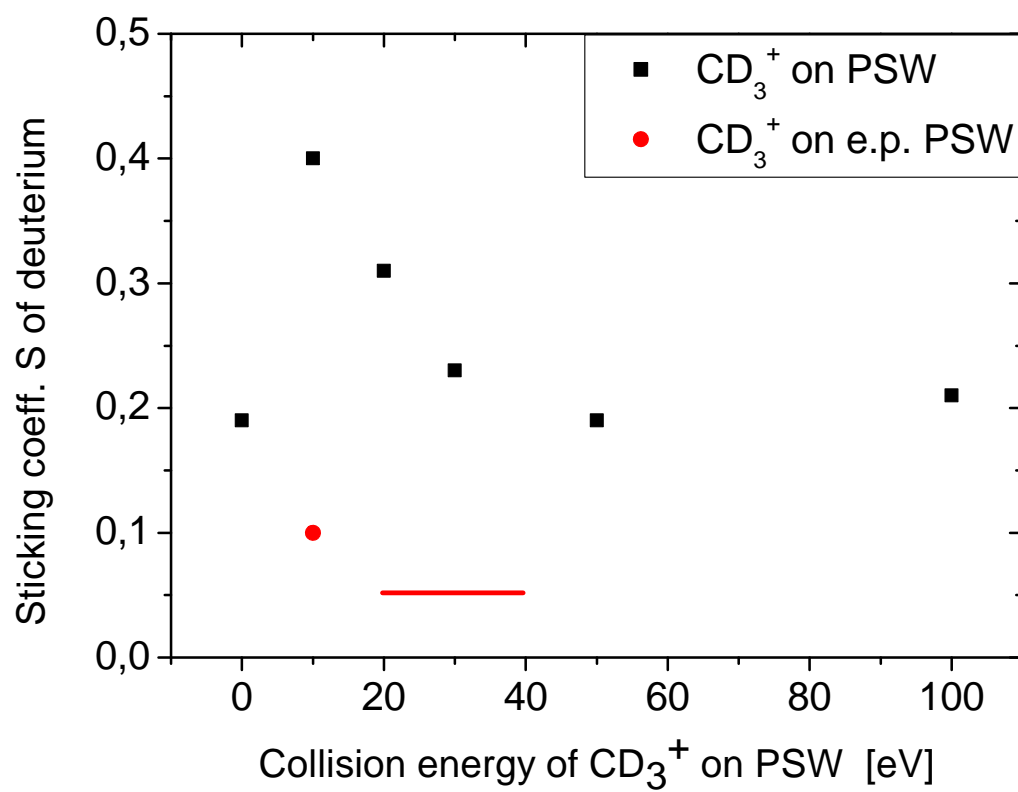


Fig. 2

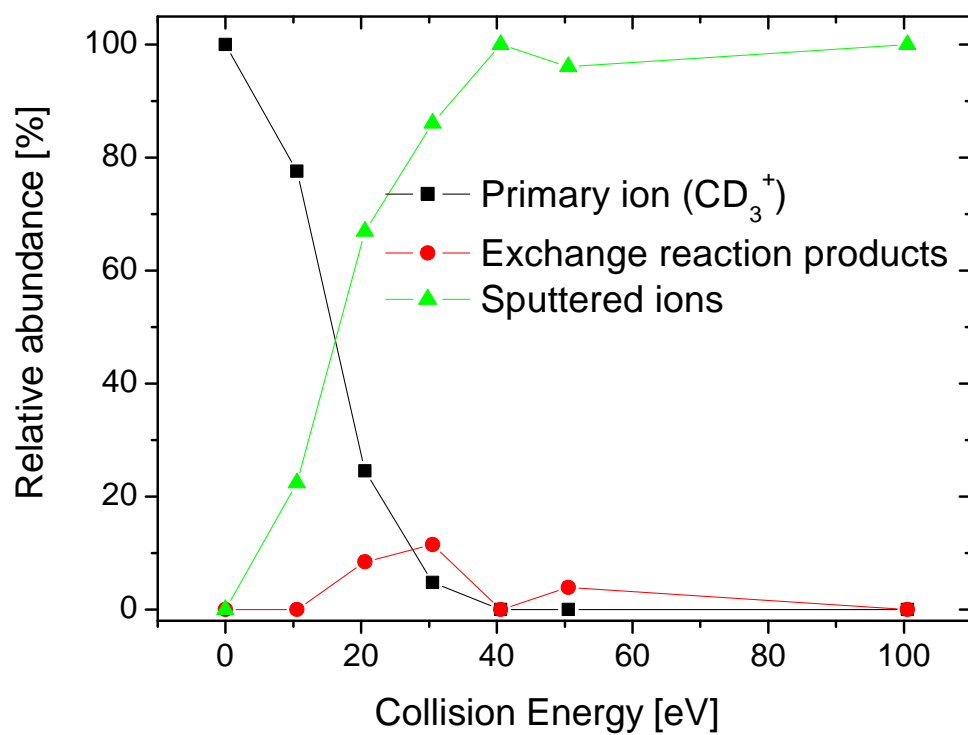


Fig. 3