ELSEVIER

Journal of Nuclear Materials 313–316 (2003) 354–359

journal of nuclear materials

www.elsevier.com/locate/jnucmat

Chemical erosion behaviour of carbon materials in fusion devices

V. Philipps [a,*], M. Stamp [b], A. Pospieszczyk [a], A. Huber [a], A. Kirschner [a], E. Vietzke [a]

[a] *Institut für Plasmaphysik, Forschungszentrum Jülich GmbH, EURATOM Association, Trilateral Euregio Cluster, D-52425 Jülich, Germany*

[b] *Euratom/UKAEA Fusion Association, Culham Science Centre, Abingdon, Oxon OX14 3DB, UK*

Received 27 May 2002; accepted 17 September 2002

Abstract

Published data on chemical erosion of graphite in present fusion devices obtained by spectroscopy are discussed in view of existing ion beam data. In addition, new data from TEXTOR for an intermediate flux range and from JET at different wall temperatures are presented. A distinction of data measured under erosion dominated, generally attached plasma conditions and under deposition dominated, mostly detached conditions is proposed. Under attached conditions yields are between 1% and 5% with only a moderate temperature dependence. Under deposition conditions connected with cold plasmas chemical erosion depends more strongly on the target temperature and the yields are more uncertain. In general, yields evaluated by CD spectroscopy still suffer from significant uncertainties of the effective photon efficiency (D/XB) of the CD radical. A definitive conclusion on a flux dependence of the chemical erosion is not possible from the present database.

PACS: 52.40.H
Keywords: Chemical erosion; Graphite

---

1. Introduction

Graphite wall materials are used in present day fusion devices in order to optimise the plasma core performance and to enable access to a large operational space. A serious concern about graphite is the chemical interaction with hydrogen which leads to significant erosion yields even under cold plasma conditions but, even more seriously, also to the formation of hydrogen rich carbon deposits which can lead to unacceptable amounts of tritium retention. Thus future devices like ITER foresee graphite only for the high heat flux areas in the lower divertor region. There graphite is presently considered to be indispensable due to its ability to withstand transient heat fluxes in disruptions or ELMS.

A reasonably consistent database exist for the chemical erosion of graphite under ion beam and thermal hydrogen impact (see e.g. [1–3]) but large uncertainties still exist about the chemical erosion under fusion reactor conditions, although various tokamak experiments have been carried out over the last years. This paper discusses the present database of hydrocarbon chemical erosion of graphite under fusion conditions. It is restricted to dedicated tokamak data based on spectroscopy rather than to provide a re-view of the field. More information is available from mass spectroscopic measurements in tokamaks [4,5] or from plasma simulators [6,7].

---

[*] Corresponding author. Tel.: +49-2461 61 6331; fax: +49-2461 61 2660.

E-mail address: philipps@fz-juelich.de (V. Philipps).

PII: S0022-3115(02)01344-2

2. Measuring conditions of chemical erosion in fusion devices

The database of chemical erosion under fusion relevant conditions suffers from difficulties to diagnose the release of hydrocarbons. Spectroscopic approaches, which are discussed here, are based on the light emission of the CD radical ($A^2\Delta$-$X^2\Pi$) [8] resulting from the dissociation chain of CH_4 or higher hydrocarbons. Higher hydrocarbons are evaluated from C_2 band emission (516 nm). The light depends on the emission properties of the CD and the molecular data of all the hydrocarbon precursors in the dissociation chain towards the CD, which depend on plasma density and temperature. Sticking or reflection of the hydrocarbon precursors decrease or increase the CD light emission showing the importance of surface processes (Fig. 1). These processes determine then the loss events per photon of the molecular CD or C_2-band (D/XB) which are needed to calculate from the light emission in particle source rates. Fig. 1 shows as an example the effective D/XB of CD depending on plasma parameters and sticking assumptions for a constant plasma background (column 1) and in front of a TEXTOR limiter with n_e, T_e defined at the LCFS and experimentally measured profiles, calculated with ERO-TEXTOR using the atomic data for methane [9]. Also, spectroscopic observations integrate often over areas with different flux densities and plasma parameters. CD light is also produced by the release of higher hydrocarbons, however with a lower efficiency (higher D/XB), for C_2H_4 e.g. about 0.7–0.5 of that of methane in the range 10–20 eV according to [24]. In JET, higher D/XB for CD originating from C_2H_4 have been found. The scatter in chemical yield measurements published from different tokamaks originates largely from different assumptions on the effective D/XB values. An isotope effect in chemical erosion exists (see chapter 6) but in this paper only data from plasma operation with a majority of deuterium are considered. Isotope effects between H and D are seen in JET [18], ASDEX [17], JT-60 [19] and TEXTOR [16] in beam experiments and are thus clearly proven. Enhancements factors are between 1.3 and 2 but possible dependencies of the isotope effect on hidden parameters (like flux or target temperature) have not been identified so far.

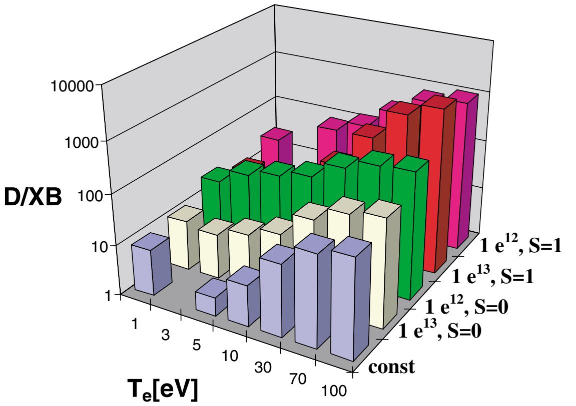

Fig. 1. Effective D/XB values for methane depending on T_e, n_e and sticking probability (S) calculated with ERO-TEXTOR. Column 1 is for constant plasma conditions the other data for erosion dominated connected in general with higher TEXTOR edge plasma conditions with the given n_e, T_e at the LCFS.

3. Parametric dependence of chemical erosion

Chemical erosion of carbon by hydrogen impact is a complex process, for which some of the atomistic mechanisms have been clarified recently [10,11]. The data show that the formation rates depend on the target temperature, particle impact energies and fluxes, isotope and surface condition of the carbon material. These parameters influence each other which can lead to synergistic effects: the simultaneous impact of one component (e.g. thermal atoms) together with a high energy component (e.g. hydrogen or other ions) enlarge the erosion of the thermal component ('synergistic erosion') [12].

At higher impact energies (>200 eV) the dependence on target temperature is strong in beam experiments but with a decreasing tendency with decreasing impact energy (flattening of the temperature dependence). However, for thermal atom impact (0.2 eV), (or cold, detached-like plasmas) the target temperature dependence is again strong indicating a transition of the parametric dependence of the erosion on target temperature between about 5 and 10 eV ion impact energy. This is of importance for the understanding of chemical erosion under cold plasma conditions. For this conditions the absolute erosion is in addition very sensitive to the 'surface structure' of the graphite which is of no significant importance at higher hydrogen impact energy (or attached plasma condition).

4. Erosion and deposition dominated surfaces

The important difference between erosion in beam experiments and under fusion conditions is that in ion beams the eroded carbon material is lost and erosion occurs on 'fresh' material whereas in fusion devices the eroded carbon returns to the surface and can be re-eroded again (carbon re-cycling). A reasonable ordering parameter is to separate the chemical erosion on erosion and deposition dominated areas. The outer divertor under attached conditions and the limiters are mostly erosion dominated connected in general with higher plasma temperatures, typically 10–30 eV (divertor) or

25–70 eV (limiter). The areas in the inner divertor (often detached) and the outer SOL regions in limiter machines are deposition dominated connected with lower plasma temperatures and also higher fraction of background carbon fluxes. On these areas the structure of the deposited carbon depends on the impact energy turning the deposited film from a so called hard film (density > 1.6, refractive index > 2, hydrogen content at 400 K = 0.4) to a more soft-like film (density < 1.6., refractive index < 1.8, hydrogen content at 400 K > 0.4). For these conditions the chemical erosion is strongly temperature dependent and largely enhanced for soft, polymerlike carbon films compared with hard films. Fig. 2 shows chemical erosion data normalised to 570 K by thermal hydrogen impact for different types of films [13,14] with the density as ordering parameter and in the lower part the relative erosion of hard and soft like films depending on temperature [15].

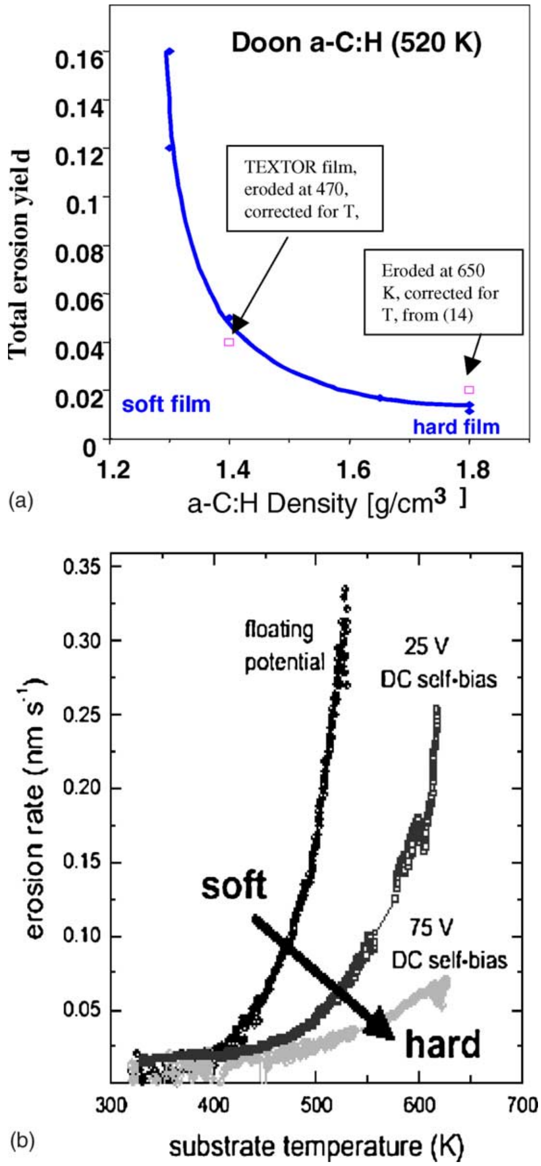

Fig. 2. Chemical erosion for thermal hydrogen atom impact for different types of carbon films, data from are from 13–15.

5. Temperature dependence of chemical erosion

In TEXTOR detailed measurements have been done under limiter conditions ($T_e \sim 50$ eV) with a stepwise heating of the limiter from the rearside up to about 1400 K [16]. The temperature enhancement of the yield from 400 K to 700 K is about 1.3, much weaker than beam data at comparable ion impact energies (200 eV). Fig. 3 compares these data with measurements in the outer divertor of ASDEX-U [17], JET [18] and JT-60U [19] and with ion beam data from [20]. The outer divertor of JET and ASDEX-U show no measurable increase with target temperature in the temperature range of 450–520 and 300–400 K, respectively whereas JT60-U found an increase by a factor of 1.6 from 420 to 550 K. The absolute yields will be discussed in chapter 6 and the CD band photon efficiencies used for the evaluation are listed in Table 1. Interestingly, on the deposition dominated areas in the inner divertor of JET [21], the normalised molecular CD and C_2 and carbon ion signals

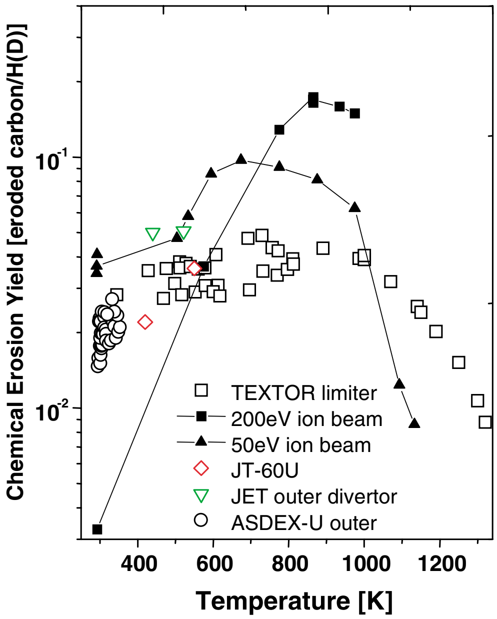

Fig. 3. Temperature dependence of chemical erosion measured in different fusion devices together with ion beam data. For data evaluation see also Table 1.

Table 1
Measuring conditions and published chemical yields for various fusion experiments

Attached conditions	Target temperature (K)	n_e ×10^{19}/m^3	T_e (eV)	D/XB (CD)	Yield % (methane)
TEXTOR limiter	300–1500	0.1–1	30–100	100 (increasing for fluxes >10^{23}/m^2 s → 250)	3–5 decreasing to 1% for fluxes >10^{23}/m^2 s
ASDEX outer divertor	370–470	1–5	5–15	50	3–1 decreasing with flux (10^{22}–10^{23} #/m^2 s)
JET outer divertor	400–550	1–7	20–8	70–130	5 no flux dependence
Jt-60U outer divertor	420–550	2–20	10–20	90–100	3.6–2.5 small decrease with flux (10^{22}–10^{23} #/m^2 s)
DIII-D outer divertor	<450	0.5–1	25–40	50	1–2.5
Detached conditions					
JET inner divertor	400–550	0.2–2	1–6	>100	5
DIII outer div	<400			5	0.01

(CIII) increase by almost a factor of two during a moderate increase of the target temperature from 450 to 520 K, observed both in L-mode and H-mode plasmas. (Fig. 4). The almost identical increase of the CD and C_2 molecular light with ion signals (C^{++} and C^{+}) shows the dominance of the chemical erosion sources in the inner divertor. This sharp temperature dependence is in good agreement with erosion of soft carbon films measured in beam experiments as shown in Fig. 3. Obviously the erosion due to low energy hydrogen ion and atom impact is below the transition energy at which the temperature dependence changes as discussed in Section 2. Large overall chemical yields of about 20% are estimated with large contributions of C_2-hydrocarbons in reasonable agreement with the yields shown in Fig. 2 for soft carbon layers.

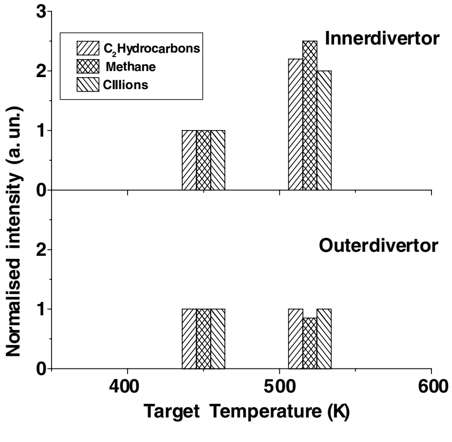

Fig. 4. Temperature dependence of the normalised C_2, CD and CIII light emission for the inner and outer divertor of JET depending on target tile temperature.

6. Chemical erosion yields

6.1. Attached conditions

Various data have been published for attached conditions on erosion dominated areas in the outer divertor or on limiters, although at different target temperatures [16–19,22,23]. The temperature dependence under these conditions is weak (see Section 4) a comparison even at different temperatures is reasonable. The data, measuring conditions and D/XB values are listed in Table 1.

In TEXTOR limiter data are obtained with a well defined observation geometry allowing local measurements, with flux densities from power fluxes and n_e, T_e data. A fixed D/XB (CD) of 100 ($H\gamma$: S/XB 1000) is used up to flux densities of about 10^{23} H/m^2 s and for higher fluxes relative D/XB values (adopted to 100 for lower fluxes) up to about 250 have been obtained by methane injection under identical conditions. As seen in Fig. 5 yields are about 3–5% up to 10^{23}/m^2 s, decreasing down to 1% with higher fluxes using the relatively adopted D/XB data [16]. The contribution of the hydrocarbons to the C^{++} light is about 40% of the total light. Few measurements have been done on C_2 hydrocarbons with the conclusion that C_2 hydrocarbons contribute to about 50% of the methane for attached conditions, but more data are needed in this area. In JET [18] the yields are integrated over the whole outer or inner divertor and fluxes are from I_{sat} and H_α which matches well (with S/XB (H_α) = 20). CD light is calibrated by methane injection but based on the important assumption that the D/XB of the intrinsic sources is similar to the injected although the location of the source does not match. With D/XB varying between 70 and 130 yields of about 5%, very similar as in TEXTOR (with similar D/XB) are deduced. Similar yields are deduced in the inner divertor but the flux determination is more difficult due to regular detachment. Higher hydrocarbons (C_2-band)

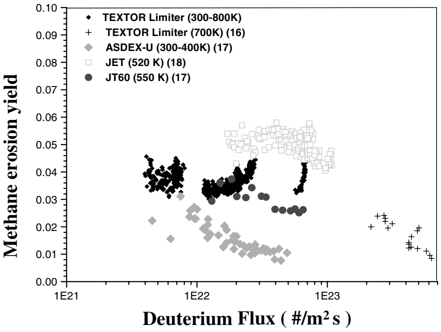

Fig. 5. Chemical erosion yields evaluated in different devices depending on ion flux density.

contribute to about 50% of methane in the outer divertor (same C-release) and about the same in the inner (twice C-release), resulting in overall yields of 10% and 20% in the outer and inner respectively. In ASDEX-U yields in [17] have been evaluated for the outer divertor based on fixed D/XB of 50. Fluxes are from probes and H_α, which agrees well. The data are for lower temperatures $T < 470$ K) with values of about 3% at lower fluxes (10^{22}/m^2 s) decreasing with fluxes down to 1% (8×10^{22}/m^2 s). In DIII-D [22,23] yields for the outer divertor region for attached conditions are evaluated with a D/XB of 50 and H-flux from spectroscopy. Yields are 1–2.5% for virgin targets and at low temperatures similar to ASDEX-U. Surprisingly the yields decrease by almost a factor of 10 in the course of long term plasma operation (20 000 shots). This has been attributed to consecutive boronization (30 boronizations in 10 years operation, which is, however, not seen in devices like TEXTOR, ASDEX or JT-60 applying also boronization with a comparable amount of boron deposition. This needs further clarification. In JT-60U [19] data for L-mode plasmas for the outer divertor at different target temperatures are obtained with D/XB values from [24] ranging between 90 and 100 and fluxes from probes and spectroscopy. Yields are between 3.6 (550 K) and 2.2% (450 K), showing a comparable strong temperature dependence when compared with JET, ASDEX and TEXTOR for attached conditions (Fig. 3).

6.2. Detached conditions

Larger uncertainties exist for detached plasma conditions, the reason for this is twofold: (1) D/XB values are more uncertain, (2) the target temperature is more important for the yields thus data can only be compared for the same temperature. DIII claims a drastically reduced chemical source under detached conditions, 0.01%, [22] at a cold target (<450 K) and with a D/XB of 5 resulting from model calculations. In contrast, the D/XB values obtained in JET by gas injection in the private flux region in the inner divertor are only about half of those in the outer divertor, about 30. This together with higher target temperature (450–550 K) results in a much larger chemical yields, about 5% and 7% for methane and C_2 hydrocarbons, respectively.

7. Flux dependence

Fig. 5 shows a compilation of published data on chemical erosion depending on flux density. In addition new data points from TEXTOR for lower flux densities evaluated with fixed D/XB of 100 are displayed. All the conditions are listed in Table 1. In a flux range of 10^{22}–10^{23} #/m^2 s a flux dependence is deduced in ASDEX [17], $Y \sim \Gamma^{-0.7}$ (fixed D/XB) and somewhat weaker in JT-60 [19], $Y \sim \Gamma^{-(0.2\text{–}0.3)}$, (D/XB 90–100). This is not clearly seen in TEXTOR [16] ($D/XB = 100$), and JET [18] (D/XB, 70–130). For higher fluxes exceeding 10^{23}/m^2 s and under conditions of large n_e and T_e ($>10^{13}$/m^3 and $T_e > 100$ eV) a clear decrease of the yield is seen in TEXTOR (with D/XB values relatively calibrated). A clear statement on the flux dependence from fusion based data is thus still not possible presently.

8. Conclusions

On erosion dominated areas the temperature dependence of chemical erosion is weak and yields based on CD spectroscopy show a certain consistency with values between 1% and 5% using D/XB values for the CD band between 50 and 200. On deposition dominated areas the plasma is colder and the surface temperature enters more strongly. Under those conditions yields from different devices show large differences with low (<0.1% DIII) and large yields (>5% JET) which is largely due to different D/XB values and differences in the target temperature. Measurements of a possible flux dependence in an intermediate flux range (10^{22}–10^{23} #/m^2 s) give $Y \sim \Gamma^{-n}$ with n ranging from 0.7 (ASDEX-U), 0.3 (JT-60U) to zero (TEXTOR, JET) allowing no clear statement about a possible flux dependence in this flux range. As a conclusion, quantitative data on yields and a possible flux dependence need to be re-visited. A careful in situ calibrations for CD spectroscopy that matches the plasma conditions, viewing geometry and hydrocarbon source location as much as possible is necessary. Averaged D/XB values depend on the experimental conditions and are not transferable easily from one device to the next without accurate modelling based on the atomic and surface data of all the species in the hydrocarbon chain and using measured plasma parameters. New experiments are needed based on in situ calibrations of the CD band emission and further work on the atomic database and model validation in dedicated experiments is indispensable.

References

[1] E. Vietzke, A.A. Haasz, in: W. Hofer, J. Roth (Eds.), Physical Processes of the Interaction of Fusion Plasmas with Solids, Academic Press, 1996.

[2] M. Balden, J. Roth, J. Nucl. Mater. 280 (2000) 39.

[3] B.V. Mech, A.A. Haasz, J.W. Davis, J. Nucl. Mater. 255 (1998) 153.

[4] V. Philipps et al., Phys. Scr. T64 (1996) 71.

[5] R. Ruggiéri, E. Gauthier, J. Hogan, et al., J. Nucl. Mater. 266–269 (1999) 660.

[6] D.G. Whyte, G.R. Tynan, R.P. Doerner, J.N. Brooks, Nucl. Fusion 41 (2001) 47.

[7] H. Grote, W. Bohmeyer, P. Kornejew, et al., J. Nucl. Mater. 266–269 (1999) 1059.

[8] K. Behringer, J. Nucl. Mater. 176&177 (1990) 606.

[9] A. Kirschner et al., these Proceedings.

[10] E. Vietzke, J. Nucl. Mater. 290–293 (2001) 158.

[11] A. Horn, A. Schenk, J. Biener, et al., Chem. Phys. Lett. 231 (1994) 193.

[12] E. Vietzke et al., J. Nucl. Mater. 111&112 (1982) 763.

[13] E. Vietzke et al., JNM 145–147 (1987) 443; E-MRS Strasbourg (1987) 351.

[14] T. Schwarz-Seeliger et al., J. Vac. Sci. Technol. A18 (2000) 995.

[15] A.W. Keudell, W. Jakob, J. Appl. Phys. 79 (2) (1996) 1092.

[16] A. Pospieszczyk et al., J. Nucl. Mater. 241–243 (1997) 833.

[17] A. Kallenbach et al., J. Nucl. Mater. 266–269 (1999) 343.

[18] M.F. Stamp, S.K. Erents, W. Fundamenski, et al., J. Nucl. Mater. 290–293 (2001) 321.

[19] T. Nakano et al., 14th Iter expert group meeting, Naka, July 2001.

[20] M. Balden, J. Roth, J. Nucl. Mater. 280 (2000) 39.

[21] J.P. Coad, N. Bekris, J.D. Elder, et al., J. Nucl. Mater. 290–293 (2001) 224.

[22] D.G. Whyte et al., Nucl. Fusion 41 (2001) 1243.

[23] D.G. Whyte et al., J. Nucl. Mater. 290–293 (2001) 356.

[24] A. Pospieszczyk, Y. Ra, Y. Hirooka, et al., Report UCLA-PPG-1251 (1989).