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Chemical erosion measurements from various carbon based limiters and coatings in TEXTOR-94

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Abstract

A well-diagnosed test limiter has been used in TEXTOR-94 to study — by means of spectroscopic techniques — the fluxes and the spatial distribution of hydrocarbons, which are formed by plasma interaction on carbon, silicon and titanium doped (SiC30, RGTi) graphites, B_4C as well as from carbon coated metal limiters. On a special graphite limiter, which could externally be heated up to temperatures of 1400 K, the methane formation remained approximately constant up to 1050 K with a yield of about 3.5% and dropped well below 1% at 1350 K. Simultaneously the carbon ion fluxes into the plasma decreased by about 50%. By inserting the limiters deeper into the plasma the D-flux increased between $10^{22}/m^2s$ and $3 \times 10^{23}/m^2s$ and the maximum methane production yield decreased to about 1%. On bulk doped graphites as well as Cu- and SS-limiters a similar methane formation behavior as pure graphite has been observed. However, a weak hydrocarbon formation rate is found for Mo- and W-limiters. No discrepancy between radial CH- and C_2-intensity profiles from injected and sputtered CH_4 and C_2H_4 gas could be detected. The measured radial intensity profiles for CD were compared with a local erosion and deposition model (ERO-TEXTOR) using measured plasma parameters and atomic data taken from the Langer model. The calculations showed a reasonable agreement with the experimental results both in radial and toroidal direction.

Keywords: TEXTOR-94; Limiter; Chemical erosion; Hydrocarbons; Molecular break-up

1. Introduction

Since the introduction of carbon based materials as wall coatings and limiters in fusion devices, a growing interest has been developed for the examination of chemically sputtered species from these surfaces. Only very qualitative information about the source function, the transport and the redeposition properties of released hydrocarbons has been obtained during the last decade because both of the lack of molecular data bases and the non sufficient diagnostic access to the locations of their origin. Therefore an attempt has been made both to check parts of the data base for the spectroscopic determination of the fluxes of the CH(D)-, C_2-, and BH(D)-radical and to broaden the experimental base for the different kind of limiters and limiter/divertor coatings used in present and possible future fusion devices. One major subject of these investigations is to find the temperature and flux dependence of the chemical erosion yield for high deuterium fluxes, as this knowledge is an important factor in the fuel efficiency of the molecular sources in a carbon surrounding.

2. Experiment, methods, and calibrations

The majority of the results obtained in these experiments were measured by optical spectroscopic techniques. In TEXTOR there is an ideal access for observations of optical emissions from test limiters (see Fig. 1). The impurity release from these limiters is studied using a 0.5 m spectrometer equipped with a 2D-CCD camera with image intensifier; the data evaluation is performed by the

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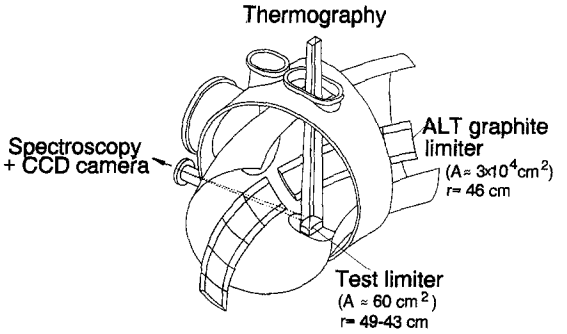

Fig. 1. Observation geometry with the lines used for spectroscopic measurements.

use of a video processing system. The spectrometer is capable of detecting line radiation in the range 210–850 nm (simultaneously observable range 53 nm), is absolutely calibrated, provides radially resolved intensity spectra over 10 cm with a resolution of 0.5 mm and was set up at a fixed toroidal position. For the latter position there was conventionally chosen the location of largest power loading (which was normally on the ion flux side) or a hole for gas blow experiments 15 mm from the center (which was on the electron flux side in order to obtain a better discrimination against background radiation). The same line of sight is shared by an intensified CCD-camera with interference filters in front, which allows the simultaneous 2D-observation of the cross section in the light of a specific spectral line or molecular band (CD: 430.7 nm ($\pm$0.75 nm); C_2: 515.75 nm ($\pm$0.75 nm); CI: 247.8 nm, CII: 514.5 nm, CIII: 464.7 nm). More experimental details can be found in [1]. The plasma parameters at the radial position of the limiter were measured by means of thermal He- and Li-beams [2,3].

The limiters (10 cm long, 6 cm wide and 5 cm high) with a toroidal curvature of 8.5 and 6 cm, respectively, were introduced into TEXTOR through a limiter lock from the bottom of the torus. Silicon and titanium doped (SiC30, RGTi), B_4C and pure graphite (EK98) test limiters as well as metal limiters (Cu, SS, Mo, and W) were used for the experiments and had been positioned at the same plasma radius as the toroidal belt limiter ALT-II (r_{Lim} = 46 cm) or 0.5 cm behind. In order to become independent of the limiter heating by the plasma itself, which strongly depends on the discharge conditions chosen, one of the graphite limiters could be internally heated up to about 1400 K.

Specific care was taken for the conversion of measured photon intensities into molecular CD_4-fluxes. The observed CD transition ($A^2\Delta$–$X^2\Pi$ at about 431 nm) is well known and spectra have been already published in several journals (e.g., [4,5]). Very near to this band is also a BD-band (at 432.9 nm) and the D_γ-line (at 433.9 nm), the latter is very convenient to be used for the determination of the deuterium flux and without a major error in CD- to D-flux ratio measurements. Moreover CD^+-band emission can be identified at 420 nm (Fig. 2). Experimental [6] and theoretical [5] attempts have been made to determine the value S/XB (decay rate/photon) for the CD-band in order to correlate the measured photon ratios Φ_{CD}/Φ_D to the CD_4/D-flux ratios. These conversion factors are considerably dependent on T_e, n_e, and the ratio of molecular/atomic source of deuterium. Within the conventional experimental plasma parameter range all three factors together can lead to factors within the range of 0.06–0.13, with which the measured photon flux ratios have to be multiplied in order to obtain the particle fluxes CD_4/D. Therefore, not only the *local* plasma parameters have to be known (which they are mostly not) but also the source of the level population for D_γ. In [7] a number of S/XB = 100 for CD is adopted and in [5] S/XB = 800 for D_γ. We have raised the latter value to 1000 taking into account latest calculations, which also consider molecular sources for the atomic deuterium level population [8]. In addition we could on TEXTOR verify the measurements of [6] concerning the temperature dependence of S/XB for the CH-band (see Fig. 3) by blowing a fixed quantity of CH_4 through a hole into a plasma with changing boundary conditions, thus confirming the independence of this value in a wide range of plasma boundary parameters. From these experiments an excellent agreement is also obtained, when the simultaneous increase of CH and H_γ during the puff was measured and a source of 4 hydrogen atoms from the CH_4 was assumed. However, this method of calibrating atom line intensities with molecular compounds has to be treated with caution, as the photon efficiencies by dissociative excitation may differ from those by direct excitation!

When the limiter was moved to smaller plasma radii into a region of high electron temperature and density with the amount of CH_4 blown kept constant, the photon yield for CD dropped by about a factor of 2, which is more than expected from the electron temperature rise and may partly

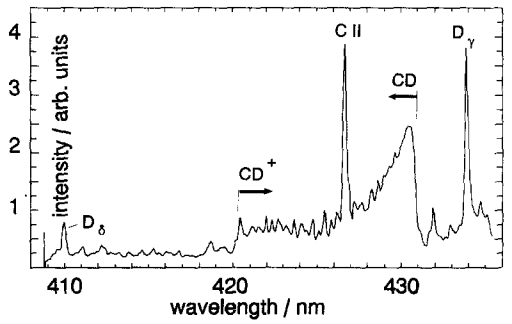

Fig. 2. Spectrum showing the transitions for CD: $A^2\Delta$–$X^2\Pi$ and CD^+: $A^1\Pi$–$X^1\Sigma^+$ during a CD_4-gas blow of 1×10^{19} molecules/s. The emission from both radicals can be clearly distinguished and has already been used for the measurement of radial intensity profiles [20].

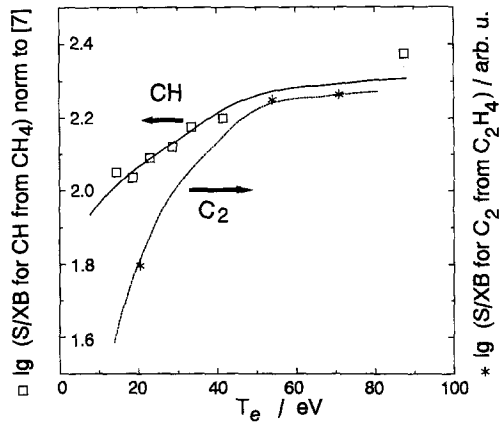

Fig. 3. Ionization (decay) rate/photon for CH from CH_4/C_2H_4 and for C_2 from C_2H_4. The curve for CH was normalized at 30 eV to the corresponding one in [6] (note the stronger temperature dependence for C_2!).

be the result of the shadowing of the light by the limiter curvature and (more probable) a change in the dissociation chain of CH_4 and a variation of S/XB with electron density. In the latter case only a collisional radiative model for CH may lead to more clarification. This discussion shows that it is at present improbable to derive hydrocarbon production yields by optical means more accurate than a factor of 2.

It is interesting to compare the photon yield from the CH-band for injected CH_4 and C_2H_4 under tokamak conditions. By blowing the same amounts of these gases into identical plasma discharges it was found that the CH-band intensity was practically equal. This is in good agreement with [6]; this also means that higher order hydrocarbons can in principle contribute a considerable fraction to the CH-photon yield. Therefore this amount has to be checked by spectral observations of other molecular compounds or radicals e.g. C_2 from higher order hydrocarbons (see below).

3. Results and discussion

3.1. Experiments with a heated graphite limiter

In [1] experiments have been presented, where parts of a graphite limiter have been heated up by plasma impact to about 2200 K. The experiments showed a clear drop of the methane production yield but indicated a temperature delay of this drop compared to beam data. Since the line integrated optical observation averages over the inhomogeneous heating, we performed experiments on a graphite limiter, which could be homogeneously heated up to 1400 K from the rear by a resistively heated meander from pyrolytic graphite. In this temperature region a maximum of the production yield (more or less strongly dependent on the impinging ion energy) was found in laboratory experiments [9]. The plasma conditions were kept constant in the course of the heating ($\bar{n}_e(0) = 2.5 \times 10^{13}$ cm^{-3}, $n_e(46\text{ cm}) = 3 \times 10^{12}$ cm^{-3}, $T_e(46\text{ cm}) = 30$ eV, $\Gamma_D = 2 \times 10^{18}$ cm^{-2} s^{-1}). As the experiments were started about 20 discharges after a fresh boronization (which took place *without* limiter!), also the appearance of B and BD could be seen in the course of the discharges (Fig. 4b). It is remarkable that from the observation of a BII- ($\lambda = 412.0$ nm) and a CII-line ($\lambda = 426.7$ nm) a B/C flux ratio of 1.2–0.8 could be derived just after inserting the limiter for the first time. In the course of the experiment the boron fluxes decrease slowly until a quasiconstant value is reached after about 100 plasma shots; this was already reported in [19]. The BD radical, which could also be seen since the first discharge, shows quite the same behavior as the CD but is about two orders of magnitude smaller using

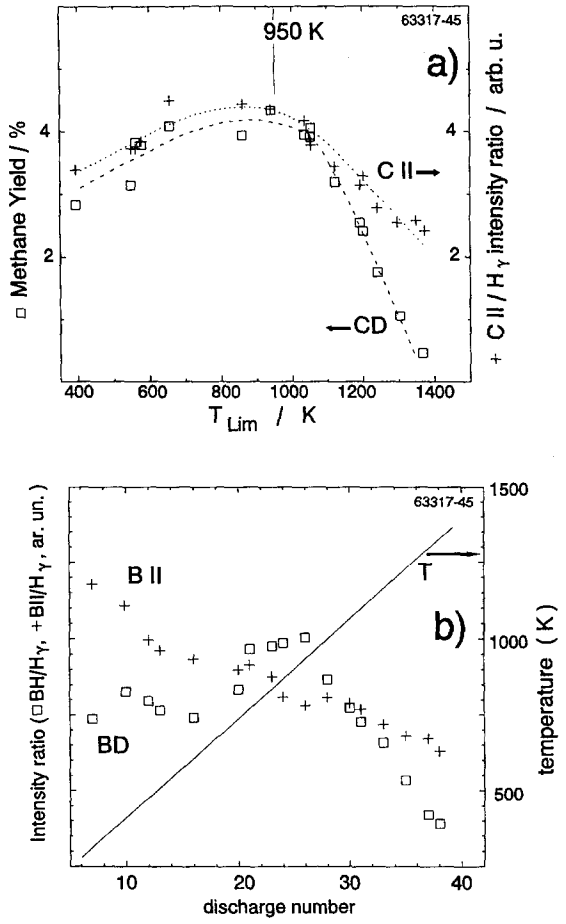

Fig. 4. (a) Methane yield and C II/D-flux ratio and (b) BD-yield and B II/D-flux ratio for a graphite limiter as a function of limiter temperature for identical plasma discharges ($\bar{n}_e(0) = 2.5 \times 10^{13}$ cm^{-3}, $n_e(46\text{ cm}) = 3 \times 10^{12}$ cm^{-3}, $T_e(46\text{ cm}) = 30$ eV, $\Gamma_D = 2 \times 10^{18}$ cm^{-2} s^{-1}). The temperature rise in the discharge sequence was practically linear (see (b))).

a number for S/XB of 1.2 for BD [7]. As can be seen in Fig. 4b the drop of the BD signal at high surface temperatures down to very small values does not affect significantly the BII line, showing the fact that indeed the BD emission plays only a minor role in the overall boron release.

In Fig. 4a the variation of both the methane yield and the relative CII fluxes can be seen during the limiter heating. Two important facts can be derived: (1) The maximum yield is only 50 K higher than the room temperature values and (2) a drop of the yields takes place only beyond 1050 K. A strong broadening of the chemical peak towards lower temperatures is always seen, when the impact energy of the hydrogen ion falls below about 150 eV [10]. Such a shift of the decrease of the methane production by about 100 K is not seen so far in any experiment and must be attributed to the high flux densities in the boundary layer impinging on the graphite limiter [11]. Similar measurements have already been made in the past on a tokamak (DITE), plasma simulators (PISCES) and ion beams. A comprehensive collection of references hereof can be found in e.g. [12], however, either the hydrogen fluxes were much weaker or the observation geometry was not as favorable as in TEXTOR.

An important observation is the simultaneous decrease of the carbon fluxes by about 40% at high target temperatures. This is a clear demonstration that 40% of the CII fluxes can be attributed to hydrocarbon formation on the limiter surface. The penetration depth of the CII-ions as measured at 1350 K increased by about a factor of 2 to about 3 cm compared with that at 900 K. The profile of the difference of both distributions is attributed to C^+ originating from hydrocarbons and has a penetration depth of 1 cm. Similar small penetration depths have been found for CD and BD (see also Section 3.4).

3.2. Flux dependence of erosion of graphite limiters

Fig. 5a shows the dependence of the CD/D-flux ratio on the impinging deuterium flux Γ_D. The variation of Γ_D was achieved by changing the plasma edge parameters. It should be mentioned that the variation of hydrogen fluxes is inherently connected with changes in n_e and T_e and methane formation as well as CD and D_γ photon efficiencies might change as discussed in Section 2. Therefore, the measured photon intensities have been corrected for the different electron temperatures during the variation of the deuterium fluxes. A value of about 1% was found for the highest fluxes, which increases to low Γ_D. The error bars shown indicate both the uncertainty in the conversion factor (see above) and in the experimental measurements, which is higher at lower fluxes (i.e., lower intensities). The values for the high fluxes are probably a lower limit and may be slightly higher (see Section 2). Thus the flux dependence might be weaker. This may also be indicated by mass spectrometric measurements in the TEXTOR scrape-off layer; however, one should have in mind that the radial and toroidal locations of the latter and the optical observations are different, but a convincing explanation for the differences has not been found yet.

3.3. Experiments with different C-based and metal limiters

Surprisingly the CD/D-flux ratio in front of boron, silicon, and titanium doped graphites does not show a significant deviation from that for pure graphite limiters (Fig. 5b), although in these cases a reduction of about a factor of 5 and more had been expected from laboratory experiments with plasma simulators (see [12]) and ion beams [13]. An explanation for this behavior may be found in experimental findings, when stainless steel and copper limiters have been exposed to the plasma in the limiter position. Those limiters, when they were first inserted into the boundary layer, displayed immediately a similar CD/D-flux ratio during the first exposure discharge. This

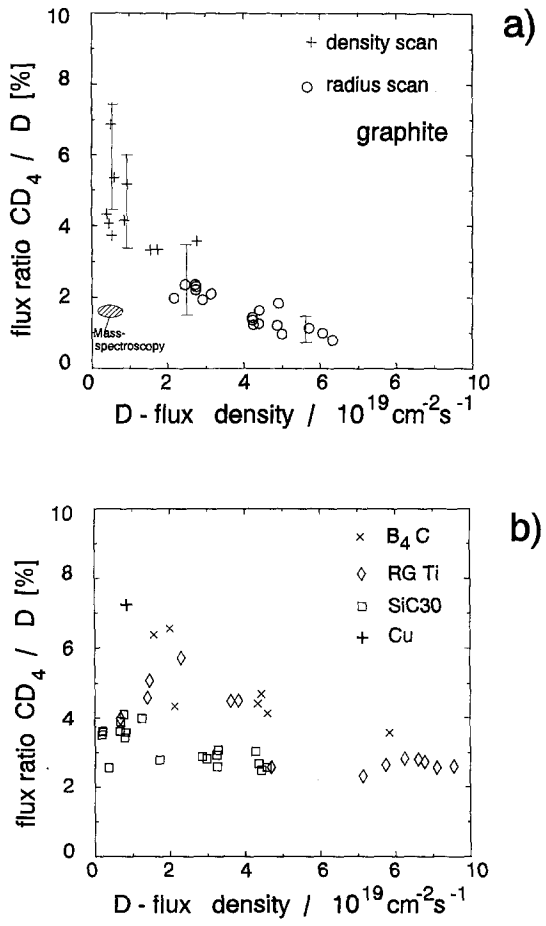

Fig. 5. CD_4/D-flux ratio as a function of D-flux density for a graphite limiter ($T_{Lim} \approx 550$°C) measured by optical and mass spectroscopy (a) for different limiter materials (b). For the discussion of the error bars see text!

shows that the yield from copper and doped carbon limiters reflects the carbon to deuterium ratio in the scrape-off layer.

However, Mo- and W-limiters hardly displayed a formation of hydrocarbons as the CD band in these cases was reduced at least by one order of magnitude. Obviously more carbon particles are directly reflected at the surface and the deposited carbon atoms do not form volatile hydrocarbons but are physically sputtered. A reason for this behavior may be found in the fact that Mo and W metals can form carbides with the impinging carbon, which is not the case for stainless steel.

3.4. Molecular intensities and penetration depths for different discharge conditions

An important question in tokamak molecular physics is the nature of the original molecule where the CH or C_2 stems from. We have tried to elucidate this problem by injecting C_1- and C_2-hydrocarbons (CH_4 and C_2H_4) and compared the penetration depths of the CH- and C_2-band before and during the blow phase. Moreover the 2D-emission pattern of CH was modelled with the ERO-TEXTOR interpretation code [14] (a derivative of the ERO code [15]) assuming CH_4 as the starting molecule and compared this with the measured one. If the assumption of CH_4 as starting molecules is not correct, this should then result in major discrepancies of measured and modelled distributions.

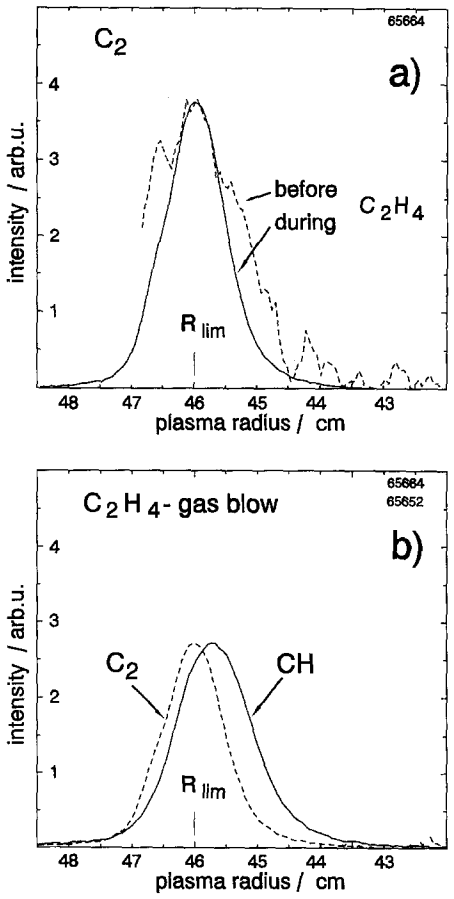

Fig. 6. (a) C_2 intensity profiles before and during a C_2H_4 gas injection and (b) a comparison for C_2 with CH.

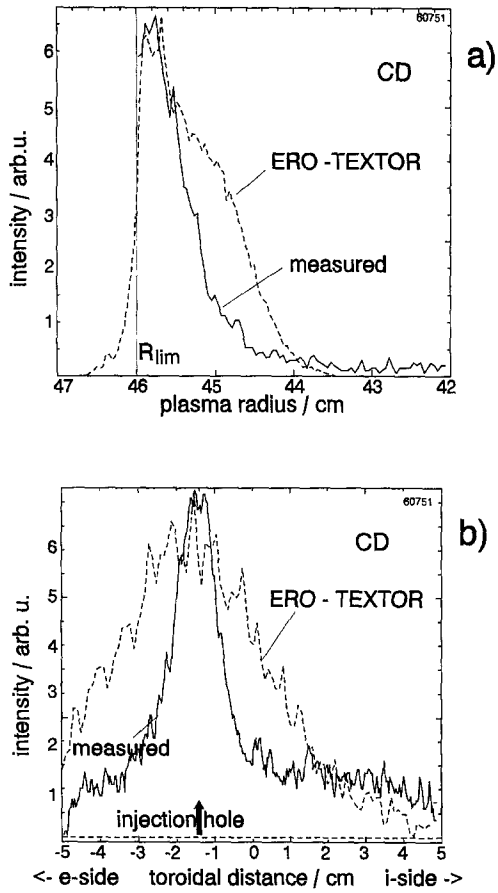

Fig. 7. Comparison of measured and modelled radial (a) and toroidal (b) CD-intensity distributions for $n_e(46\text{ cm}) = 2.3 \times 10^{12}$ cm^{-3}, $T_e(46\text{ cm}) = 28$ eV).

A comparison of the CD-fluxes with other molecular and/or atomic fluxes shows that in the TEXTOR limiter case the release of higher hydrocarbons plays a minor role. The C_2-band at 516.5 nm revealed always a low intensity except for the case of a detached plasma, when the radiation-of this band rose dramatically by a factor 10 (however, one should have in mind the variation of the photon efficiency of C_2 with T_e). This latter condition obviously resembles more those in the throat of the pumped limiter at TORE-S [16] or in the ASDEX-U divertor [7,17], where a considerably larger contribution from higher hydrocarbons has been routinely detected. By comparing known quantities of injected CH_4 and C_2H_4 into identical plasma discharges ($\bar{n}_e(0) = 3.5 \times 10^{13}$ cm^{-3}, $n_e(46\text{ cm}) = 5 \times 10^{12}$ cm^{-3}, $T_e(46\text{ cm}) = 23$ eV) it was found that in the

TEXTOR boundary plasma the contribution of C_2H_4 (measured via C_2) is about 1/3 of CH_4 (via CH). For lower densities it is probably even less.

Fig. 6a shows a comparison of the penetration depth of C_2 before and during the C_2H_4-gas blow. There is no obvious discrepancy between both profiles concerning their decay length, which justifies the assumption that C_2 is actually formed by volatile higher order hydrocarbons. The same was found in the case of CH, which is predominantly produced from CH_4. However, the penetration profiles of C_2 and CH differ slightly in the case of injection (see Fig. 6b). The difference is similar under plasma impact.

The penetration depths of CH are also modelled by the ERO-TEXTOR code, which simulates the many reaction paths by taking the reaction rates of [18] into account (Fig. 7). Also in 2D-observation geometry the model calculations show a good qualitative agreement in the radial direction with experimental observations, which have been obtained by detecting the light emission with a CCD-camera connected with a filter for the CH-band emission. From these measurements fuelling efficiencies for CH_4-blow and sputtering can be derived. They are relatively large (about 80%), which means that a major part of the discharge can actually be fueled by molecules. In the toroidal direction a better agreement can be obtained when a local cooling of the plasma by about a factor of 2 is assumed. This assumption is not unrealistic as the local hydrocarbon density can be considerably high and similar effects have been found in [19] for the injection of silane; however, more experiments have to be performed in this respect.

4. Summary and conclusions

– No pronounced maximum in the CD/D-flux ratio could be found for graphite limiters up to 1050 K. A shift of about +100 K in relation to beam experiments for the decrease of the chemical production rate is observed.

– The CD/D-flux ratio for pure graphite limiters decreases significantly with increasing flux. It is about 1% at $\Gamma_D = 10^{20}$ cm^{-2} s^{-1}.

– No differences in methane formation could be found for doped graphites, Cu- and SS-limiters. Mo- and W-limiters show a very low hydrocarbon formation.

– The ERO-TEXTOR code allows a qualitatively good modelling of the observed molecular emission pattern, but more experiments are needed.

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