# Carbon Transport in TEXTOR-94 Aspects of Long Term Operation

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#### Abstract

The paper describes an experiment carried out in TEXTOR-94 where <sup>13</sup>C containing methane was puffed into the discharge through a small hole in a graphite sample. The local deposition of <sup>13</sup>C could be distinguished from the global transport of <sup>12</sup>C in the machine by SIMS depth profiling analysis. The <sup>13</sup>C travels several cm and becomes deposited on the sample surface with low efficiency (0.2%) but with ratios <sup>13</sup>C/C  $\approx$  0.42. The co-deposited amounts of H and D were measured by NRA and ERDA. The results can be simulated quantitatively by ERO-TEXTOR code calculations if very high re-erosion is assumed for the deposit formed initially by the CH<sub>x</sub><sup>+</sup> radicals. The majority of the puffed <sup>13</sup>C travels to the plasma facing graphite limiters and seems to create a stationary <sup>13</sup>C/C ratio at the surface after the total plasma exposure time of 108 s. If the value <sup>13</sup>C/C  $\approx$  0.27 found on a collector probe is representative for the whole surface it would correspond to an interaction depth of about 27 nm which is involved into the carbon interchange. Since plasma facing surfaces are net erosion areas the carbon transport finally ends on the side walls of the poloidal limiters and on other obstacles in TEXTOR.

#### **Keywords:**

carbon transport,  ${}^{13}CH_4$  injection, deposition efficiency, simulation calculations, TEXTOR-94, long term exposure

#### 1. Introduction

Steady state operation of nuclear fusion devices is one of the next challenging goals [1], and machines like LHD, W7X or ITER are already planned for long pulse operation. Beside major concerns as e.g. the gas fuelling cycle and the heat removal, the transport of impurities will become a serious problem. They will continuously be eroded from plasma facing wall components by particle impact and deposited at other locations. This will reduce the lifetime of the components and lead to unwanted material accumulation and flaking at places

Erosion, transport and re-deposition of carbon have been studied in several machines, e.g. TEXTOR-94 [3], ASDEX-U [4] or JET [5]. Flakes with thicknesses in the range 10–100  $\mu$ m are observed in the divertor,

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difficult to access. Deposited layers consist of mixtures of all elements present in the machine [2]. These are mainly carbon, but also boron, tungsten, beryllium, steel components and oxygen. Moreover, tritium will be accumulated in these layers by co-deposition in unacceptable amounts.

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especially in shadowed areas, but also on components in the main chamber near the LCFS on plasma facing surfaces of limiters, and on areas deep in the shadow perpendicular to the magnetic field [6]. The balance between carbon erosion and deposition is highly sensitive to the circumstances of the exposure. The exposure time and operation conditions are as decisive important as the transport length, the shape of the components and possibly the surface morphology [7], or the presence of other plasma impurities (metals, oxygen).

Because plasma facing components made from graphite or CFC will still be used in the machines, the emphasis of this paper is put on the investigation of the transport of carbon as the major impurity. In the following an experiment will be described where <sup>13</sup>C containing methane (<sup>13</sup>CH<sub>4</sub>) has been puffed through a hole of a wall element in order to distinguish the local transport and deposition of the <sup>13</sup>C from the <sup>12</sup>C deposition out of the background. Of special interest were the <sup>13</sup>C deposition efficiency and the simulation calculations by means of the ERO-TEXTOR code [8] taking into account the local plasma conditions. The influence of the exposure time has been investigated additionally by means of long term exposed samples.

#### 2. Puffing Experiment

Because a part of the observations has already been presented elsewhere [9], only a brief description will be given. For the experiment, a graphite block (115 × 75 × 70 mm<sup>3</sup>) was moved from top of TEXTOR-94 and positioned in the scrape-off layer (SOL) with its outer end at the last closed flux surface (LCFS, r = 46 cm). It was kept there for 20 discharges ( $n_e = 2.5 \times 10^{13}$  cm<sup>-3</sup>,  $I_p$ = 350 kA,  $B_T = 2.25$  T) in deuterium without auxiliary



Fig.1 Schematic of the puffing experiment.

heating (total exposure time 107.7 s). The surface of the block was covered with an aluminum target plate ( $108 \times 70 \times 3 \text{ mm}^3$ ). Because of the inclination by  $20^\circ$  in toroidal direction the target reached a radial distance of 50 cm from the plasma center (fig.1).

The first two discharges were carried out without gas puffing. In the following 18 discharges puffing began at 1 s with a calibrated amount of 3.8 mbar·l<sup>13</sup>CH<sub>4</sub>-gas (corresponding to 9.2 × 10<sup>19</sup> molecules) through a 1.7 mm wide hole, which ended at r = 47.3cm. The CH and CII light emitted between the surface of the sample and the plasma edge region was observed in poloidal direction. Both intensities reached a maximum at 1.26 s after discharge begin and levelled off after 2 s. The CH light emission was brightest at the LCFS ( $r \approx 46$  cm), while the maximum of the CII radiation appeared about 1cm deeper in the plasma.

A part of the <sup>13</sup>C penetrated into the plasma and increased the CV signal by about 33%. The rest became re-deposited on the target plate together with the <sup>12</sup>C carried by the SOL plasma. Because hydrogen was codeposited, a transparent film was formed which showed thickness dependent interference colours in illuminating light.

This made in-situ colorimetry [10] possible, i.e. the two-dimensional determination of the thickness pattern assigned by the colour fringes. The fringe shift after each of the puffs yielded the incremental net deposition and, hence, the deposition rates. Nuclear reaction analysis (NRA), secondary ion mass spectroscopy (SIMS) and elastic recoil detection analysis (ERDA) were applied after dismounting of the sample in order to determine space and depth distributions of the carbon isotopes and of the codeposited deuterium and hydrogen.

Additionally, an aluminium sample was mounted perpendicular to the toroidal direction on the graphite block on the ion drift side well in the shadow of the puffed molecular  $^{13}CH_4$  cloud. The deposits found here were formed therefore mainly by the impurities travelling in the SOL. The long term samples were mounted on the Stockholm-TEXTOR collector probe system and exposed there for several weeks up to about 6000 s of plasma exposure time.

## 3. Deposition Pattern and Efficiency 3.1 Colorimetric analysis

Fig. 2 gives an black and white representation of the interference colours visible on the target plate after the end of the puffing experiment. The different orders

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Fig. 2 Top view of the target after 18 puffs and the total exposure time of 107.7 s.

of interference which can be recognised as dark stripes can be counted up to the 3<sup>rd</sup>, which corresponds to the maximum thickness of about 300 nm (circle).

Note, that because of the inclination of the plate each toroidal location corresponds to another radial distance from the plasma center. In the left part ('plasma far') the deposition is mainly due to the  ${}^{12}C$  in the SOL. It causes contour lines of constant thickness which show little variation in poloidal direction. The thickness mainly increases in direction to the plasma near side (right) due to the increasing background carbon flux. Beyond the puffing hole the contours are bent back due to the additional <sup>13</sup>C deposition, resulting in a 'butterfly' like pattern. This area of additional <sup>13</sup>C deposition is shifted poloidally by about 6 mm (in direction to line 2) due to the  $E \times B$  forces and is extended over about 8.7 cm<sup>2</sup>. Other areas (e.g. along line 1) are almost unaffected by the <sup>13</sup>C deposition, and the deposition is mainly due to the carbon flux in the SOL.

The differences of the thickness profiles along these two lines yield a rough estimate of the <sup>13</sup>C contribution alone. This has been done by colorimetry after each puff. The averaged incremental growth by <sup>13</sup>C deposition within the involved area is about 3.3 nm per puff. This corresponds to the total number of  $1.9 \times 10^{17}$ <sup>13</sup>C atoms if a density of  $6.5 \times 10^{22}$  cm<sup>-3</sup> is taken into account. Compared to the  $9.2 \times 10^{19}$  molecules injected this is a deposition efficiency of only 0.2%. The scatter was found to be ±0.1%. Low efficiencies (few %) have also been observed earlier [11].

#### 3.2 SIMS, NRA and ERDA

Depth profiling by sputtering with 3 keV oxygen



Fig. 3 <sup>12</sup>C and <sup>13</sup>C thicknesses as measured by SIMS depth profiling along line 3 in figure 2.

ions in connection with SIMS analysis was applied along line 3 (fig. 2) at 13 equidistant locations. This technique yields the total thickness of the deposit. Moreover, the ratio  ${}^{13}C/{}^{12}C$  within the layer can be determined and hence, the contributions of the two isotopes separately. For example, the  ${}^{13}C$  contributes with a thickness of about 100 nm at a distance between 2 and 4 cm from the blowing hole, (black columns in fig. 3), while the  ${}^{12}C$  deposition (hatched columns) is about 200 nm. The sum of the two (open columns) fully agrees with the total thicknesses determined independently by colorimetry.

The <sup>13</sup>C distribution shows, that <sup>13</sup>C has been transported several cm across the declined surface in toroidal direction towards the plasma near end of the target plate. The concentration <sup>13</sup>C/C = <sup>13</sup>C/(<sup>13</sup>C + <sup>12</sup>C) is not constant, however. It reaches a value of 0.42 near the blowing hole, but decreases to  $\approx 0.25$  with increasing distance. This might indicate the limited local transport of carbon. The slight decrease beyond the deposition maximum at the radial distance r = 46.5 cm is likely due to the increasing erosion by D<sup>+</sup> ions.

Deuterium is co-deposited with ratios D/C  $\approx 0.2$ rather independent on distance as found by NRA. Additionally, hydrogen from the injected molecules is co-deposited up to ratios H/C  $\approx 0.45$  in the deposition maximum as revealed by ERDA (not shown in the figure). Both values drop to  $\approx 0.1$  adjacent the end of the target plate ( $r \approx 46$  cm) due to the temperature excursions. This value might therefore indicate an upper limit for the possible hydrogen uptake from air.

#### 3.3 Simulation calculations

The surprisingly low deposition efficiency of less

than 1% seems to contradict to the spectroscopic observation that the maximum of the CH light emission occurs between the target and the LCFS. Since most of the <sup>13</sup>CH<sub>4</sub> molecules have to break up and become ionised within that small gap in order to reach the CH stage, a high fraction of them must travel along the magnetic field lines towards the declined target plate and should become deposited there. This is obviously not the case and points to the direction that the reerosion probability of the CH<sub>x</sub> radicals is very high. This is not unlikely because of the high hydrogen content.

In order to prove this idea simulation calculations with the ERO-TEXTOR Monte-Carlo code [12] have been performed. This code models three-dimensionally the local transport and re-deposition of eroded impurities under the influence of a background plasma, taking magnetic and electric field forces, friction, cross field diffusion, and thermal forces into account. Major input parameters were the measured electron density and the temperatures of electrons and ions at the LCFS and their decay lengths. These values result in an ion flux  $(\Gamma_{\rm D}^{+} = 1.33 \times 10^{19} \, {\rm cm}^{-2} {\rm s}^{-1}$  at the LCFS,  $\lambda_{\rm D} = 17 \, {\rm mm}$ ) which hits the declined surface and liberates CD<sub>4</sub> or C by chemical and physical erosion with the according yields. The fraction  $0.036\Gamma_D^+ = \Gamma_C^{4+}$  was assumed as <sup>12</sup>C impurity flux. The injection of <sup>13</sup>CH<sub>4</sub> gas which lasted roughly 1s per plasma discharge was replaced by a constant inlet rate of  $1.5 \times 10^{19}$  s<sup>-1</sup> of thermal molecules.

All hydrocarbon molecules break up, become ionised and travel either to the surface of the target or away. It is assumed that only ions can stick and change the ratio  ${}^{13}C/{}^{12}C$  within a certain interaction depth of 40 nm. Neutral hydrocarbons do not contribute to the deposition. Outputs of the code are e.g. the time evolution of the twodimensional pattern of  ${}^{13}C$  and  ${}^{12}C$ , or the light emission of the radicals around the blowing hole, which both can be compared directly with experimental observations.

Usually, 50% of the ionised radicals hitting the surface are assumed to stick, while the rest leaves it in the form of <sup>13</sup>CH<sub>4</sub>. This is described by a 'reflection coefficient'  $R_{ion} = 0.5$  in the code. This assumption leads, however, to insuperable disagreements with the experimental results: The efficiency for <sup>13</sup>C deposition reaches 40% in this case and causes thicknesses of 80–160  $\mu$ m directly adjacent the blowing hole while in the experiment a slight maximum of 300 nm was found several cm away. The concentration <sup>13</sup>C/C approaches even unity in the calculated deposition maximum, again in contradiction to the experiment (0.42).



Fig. 4 <sup>13</sup>C deposition pattern as calculated with the ERO-TEXTOR code for the case of 'high re-erosion'.

Obviously, the high sticking probability assumed is responsible for the dramatic <sup>13</sup>C deposition. Varying other input parameters, such as density, puffing rate, sputter yields, ionisation probabilities, did not much lower the deposition efficiency or yielded results far off the experimental findings. An optimum agreement was found, however, if a direct erosion was assumed for the deposited  ${}^{13}CH_{x}$  + radicals. This can mean, that the initial radicals (mainly CH<sub>4</sub><sup>+</sup> and CH<sub>3</sub><sup>+</sup>) carry too little energy to form a stable deposit when hitting the surface in a first attempt. It can also mean that the surplus of hydrogen causes the high re-erosion. Drastically increased erosion by hydrogen is also observed when a-C:H films are formed in glow discharges with self-bias voltages below 200 eV [13]. The case of 'high reerosion' was simulated in the code by setting the reflection coefficient to the extreme  $(R_{ion} = 1)$ . This immediately reduced the efficiency to the much lower value of 2.5%, because the remaining deposition is mainly due to carbon ions. The <sup>13</sup>C concentration is lowered to 0.5. Moreover, the deposition maximum is shifted by about 3 cm towards the end of the target plate. The calculated <sup>13</sup>C pattern (fig. 4) fully agrees with the experimentally observed deposition area. In addition, the calculated distributions for the CH and CII light emission agree well with the measured ones within the statistical errors.

According to the deposition efficiency of 2.5% the maximum thickness is  $1.4-2.3 \ \mu\text{m}$ . This is still 4-7 times too high. Possibly, erosion by oxygen impurity ions or by hydrogen released from the hydrocarbons, which were not taken into account, decrease the film thickness further in the experiment.

#### 4. Global Carbon Transport in TEXTOR-94

More than 99% (1.66  $\times$  10<sup>21</sup> molecules) of the

introduced <sup>13</sup>C is deposited at other locations inside the machine, most likely on the graphite tiles of the toroidal belt limiter ALTII (3.4 m<sup>2</sup>). This amount corresponds to a thickness of 7.5 nm and should create a ratio <sup>13</sup>C/C  $\approx$  0.2 provided the interaction depth is 40 nm as assumed in the calculations.

Because of the co-deposition of <sup>12</sup>C a mixed layer will be formed within the interaction depth. Since these are areas of net erosion the mixture is re-eroded and feeds the carbon flux in the SOL with the according isotopic ratio. The re-deposition will eventually end in net deposition areas, as e.g. on the side walls of the poloidal limiter ring or on other obstacles deep in the shadow.

If such a perception is true, the carbon deposits in the SOL should show an isotopic ratio independent of the radial distance from the plasma center. This was indeed found on the target mounted perpendicular to the toroidal direction and outside the influence of the puffing cloud. The value  ${}^{13}C/C \approx 0.27$  was determined by SIMS and NRA. It seems that the  ${}^{13}C$  concentration in the limiter surface has reached quasi-stationarity within the total exposure time (107.7 s) despite the continuous net erosion. Obviously, only a limited depth of the bulk is affected by the in- and outflowing  ${}^{13}C$  and  ${}^{12}C$ . If the observed ratio  ${}^{13}C/C \approx 0.27$  is representative for the whole plasma facing limiter surface (3.4 m<sup>2</sup>) it suggests an interaction depth (= mixed layer thickness) of  $\approx 27$  nm.

This is a lower limit, however, because the limiter exhibits a complicated pattern of erosion and deposition regions, which is caused by the magnetic field ripple and the complex shape of the plasma facing surface [14]. Only roughly one half of the area shows net erosion, the other half net deposition. In deposition areas, carbon can accumulate after the erosion and the local transport over short distances. Flakes of 45  $\mu$ m thickness were observed in such areas after 14000 discharge seconds [15]. The life time of the component will therefore not be determined by the average carbon loss of the graphite tile, but by the local erosion rate in the net erosion areas.

A long term exposure experiment was set up, in order to estimate the average carbon loss from the limiter surface. In TEXTOR, we can exclude the boronized liner (r = 55 cm) as a sink for carbon because it does not seem to contribute significantly to the carbon interchange. After its erosion the carbon is travelling through the SOL. The transport eventually ends on obstacles in the shadow. Here, samples were exposed to



Fig. 5 Carbon flakes grown after long term exposure in TEXTOR.

plasma discharges up to about 6000 s by means of the Stockholm-TEXTOR collector probe system.

Two samples exposed 4 cm deep in SOL for 1260 s and 5846 s are shown in fig. 5. They yield net deposition rates of approximately 2–3 nm/s. Growth rates of 10 nm/s at the LCFS could be deduced from the sample exposed in the SOL along with the puffing experiment. Provided these rates are not much depending on the poloidal location, they would correspond to a permanent flux of eroded carbon in the SOL of roughly  $1.5 \times 10^{19}$  s<sup>-1</sup> or to an equivalent of 0.3 mg/s of carbon lost from the limiters. The deposits permanently grow with time and reach e.g. thicknesses of 130  $\mu$ m on the side walls of the poloidal limiters [6]. Flaking of the thick deposits as shown in fig. 5 might limit the final thickness, but cause the formation of macroscopic particles ('dust').

### 5. Summary and Conclusions

The injection of  ${}^{13}C$  containing methane ( ${}^{13}CH_4$ ) into TEXTOR-94 through a hole in a wall element allowed to discriminate the local deposition and the

global transport of carbon. In a surrounding of mainly graphite ( $^{12}$ C), the isotope  $^{13}$ C behaves as a 'tracer impurity'. It was transported several cm across the target plate and deposited there with a low efficiency (0.2%) together with  $^{12}$ C out of the background. The ratio  $^{13}$ C/C reached values up 0.42. These findings as well as the CH and CII light emissions observed near the blowing hole could be modelled by means of the ERO-TEXTOR code if a high re-erosion probability for the hitting CH<sub>3</sub><sup>+</sup> and CH<sub>4</sub><sup>+</sup> radicals is assumed. They, obviously, form a film which is not very stable against erosion. Enhanced re-erosion by the hydrogen out of the radicals can not be excluded. Deuterium from the plasma and hydrogen are co-deposited into the film.

The majority of the  ${}^{13}C$  is first transported to the plasma facing surfaces of the toroidal graphite limiter ALTII. It seems to create an isotopic ratio in the <sup>12</sup>C bulk within an interaction depth. Since ALTII is the major carbon source in TEXTOR the flux of the reeroded carbon should have the same isotopic ratio. It is transported through the SOL to obstacles. The value <sup>13</sup>C/ C = 0.27 found independently on the radial distance on a collector in the SOL suggests an interaction depth of higher than  $\approx 27$  nm and that quasi-stationarity has been achieved despite the permanent erosion of the surface. Long term exposed samples yielded a rough estimate of about 0.3 mg/s as the average carbon rate leaving permanently the limiter surface. The carbon can eventually accumulate on limiter side walls and other obstacles no longer attacked by erosion and create permanently growing thick (e.g. 130  $\mu$ m) deposits and flakes.

The local carbon transport depends not only on plasma parameters. It is also sensitively influenced by outer circumstances like magnetic field ripple and shape of the surface. This may cause high local erosion rates on the limiter surfaces despite a low average carbon loss. Since the erosion rates determine the life time of the components predictions can hardly be made as long as the processes are not better quantified. This suggests further experiments in TEXTOR. Investigations in other machines may benefit from the finding that <sup>13</sup>C behaves like a 'test impurity', the transport of which can nicely be distinguished from the isotope <sup>12</sup>C of the background.

The ERO-TEXTOR code has been shown to be a very helpful tool in order to quantitatively simulate the experimental observations and in order to understand the processes of local transport of carbon.

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