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Removal of redeposited layers and hydrogen release by oxygen ventilation of TEXTOR-94

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Abstract

Ventilation of the TEXTOR torus with oxygen at pressures between 0.007 and 0.3 mbar and at wall temperatures between 500 and 700 K has been investigated to remove redeposited carbon material and to release the incorporated hydrogen. It has been observed that a significant part of the injected oxygen adsorbs on the walls due to formation of stable oxygen compounds. Part of the oxygen reacts with CO and CO_2. The CO reaction rate is independent of the filling pressure whereas the CO_2 production increases with increasing pressure. Plasma operation after the oxygen baking has been achieved after GDC in D_2 and He (15–30 min). The oxygen impurity content was initially about a factor of 2–3 higher than before but also showed a decreasing behaviour shot by shot. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Long-term retention of tritium fuel in the surface or bulk material of plasma facing components in fusion devices is one of the major problems in fusion technology. The objections are mainly due to tritium release in potential accident scenarios [1]. Four principal mechanisms can lead to retention of hydrogenic isotopes in plasma facing components (no difference is made here between different hydrogen isotopes for the retention and release behaviour): (i) direct implantation of ions escaping from the plasma, which leads to hydrogen retention primarily in a shallow surface region, (ii) diffusion into the bulk, (iii) codeposition of hydrogen isotopes with eroded wall material, which can produce codeposited surface layers with considerable hydrogenic content, (iv) production of tritium by transmutation nuclear reactions. It has been concluded from various investigations, that with carbon walls the dominant mechanism for hydrogenic retention is codeposition of carbon with deuterium.

In TFTR, the fraction of deuterium fuel retained in the first wall was evaluated to be 44 ± 17%. Post mortem analysis showed that the deposits were dominantly located distant from the regions of plasma-limiter interaction in low flux regions of the SOL where codeposits up to 10 μm thickness were found. Remarkable codeposition was also observed in gaps between the limiter tiles [2]. Particle balance in TEXTOR performed on a daily basis (outgassing over night and weekends are not counted) revealed a similar retention fraction of about 50%. Deposits of up to several hundred micrometer thicknesses have been built on limiters in TEXTOR in the SOL (≈5 cm behind LCFS) [3] and thick flakes were also found on the toroidal graphite limiter surface which defines the LCFS.

In the 3 year operation of TFTR in D-T, 2.7 g of tritium were introduced by NBI and 1.7 g by gas-puffing while 1.6 g remained in the vessel. The retained fraction of tritium (36%) was in good agreement with that observed in deuterium-fuelled experiments [4,5].

In the preliminary D-T experiments in JET, 5.5 mg of tritium were injected via the NBI system, of which 67%

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were still in the torus two days after the experiment. Special clean-up procedures succeeded in reducing this fraction to ~10% [6]. A much higher fraction of tritium remained in the JET during the full D-T experiment: of the 36 g tritium introduced cumulatively, about 12 g remained in the torus, which could be reduced to about 8 g by means of various cleaning mechanisms, representing a long-term retention fraction of 22% [7].

Such high retention fractions are unacceptable for fusion reactors and the evaluation of tritium removal techniques is very important. Such techniques are based on isotope exchange by normal plasma operation in deuterium/hydrogen, or by GDC wall conditioning in D_2 and He. A new proposal to remove the tritium is the baking of the torus in an oxygen atmosphere at elevated wall temperatures. It has been demonstrated in several laboratory experiments that redeposited carbon layers can be oxidised to CO and CO_2 in an oxygen atmosphere leading to the release of hydrogen. Significant oxidation rates could be achieved in the temperature range between 520 and 750 K, depending on the type of carbon film or carbon deposit [8–11]. At these temperatures, the graphite bulk material is not significantly attacked by the oxygen. The main advantage is that all surfaces including hidden areas can be accessed by this technique.

This paper reports on the first experiments of baking the torus of TEXTOR-94 in an oxygen atmosphere. Emphasis is on the formation of carbon oxides (CO and CO_2), but also on the influence of the baking on the subsequent plasma operation which is one of the main objections for the use of oxygen baking in fusion devices.

2. Experimental procedure

TEXTOR-94 is a medium-sized tokamak with a major radius of 1.75 m and a minor plasma radius of 46 cm. The spherical plasma is limited at $r = 46$ cm (LCFS) by a toroidal pump limiter (ALT), consisting of 8 blades with a surface area of 3.5 m^2. Behind the ALT limiter, a poloidal ring limiter is positioned at $r = 49$ cm with a total surface area of about 0.2 m^2. It covers about 30% of the poloidal circumference. The inner wall is protected with a bumper limiter at the same radius with an area of about 6 m^2. All limiters are made from fine grain graphite. The rest of the plasma facing surface is formed by the inconel liner which is a metallic torus with $r = 55$ cm and a plasma facing area of 35 m^2. About 15% of the liner surfaces is open for diagnostic access leading to a line of sight of the plasma to the vacuum vessel wall. The liner is heated by direct current flow and the inner wall bumper and the pump limiter are heated by thermal radiation. Temperature differences of typically ±50 K are measured at different locations on the liner and the temperatures given in this paper represent some average values. The temperatures of the pump and inner wall graphite limiters are typically about 70 K below the average liner temperature and much more uniform (±10 K). The vacuum vessel is heated separately to a temperature of 420 K.

The oxygen venting experiments have been performed by filling the torus with normal oxygen ^{16}O or with the oxygen isotope ^{18}O to a certain pressure and closing all external pumps. The evolution of the total and partial pressures were monitored by several differentially pumped quadrupoles. During the experiment, small amounts of inert gases have been added in order to ensure that no pump was active and to control the sensitivity of the quadrupoles. The stability of the quadrupoles is a critical issue when using oxygen as filling gas, probably due to a change of the secondary electron emission on electron multiplier by the impact of oxygen ions.

Standard ohmic plasma operation before and after the oxygen ventilation has been performed in order to investigate the influence of the oxygen venting on the oxygen impurity content and on the plasma performance. Standard GDC in D_2 and He (8 A, voltage of 400–800 V typically) has been used to remove the oxygen from the walls.

3. Results and discussion

3.1. Oxygen adsorption and CO and CO_2 production

Fig. 1 shows the typical behaviour of the evolution of the total pressure and the partial pressures of oxygen, CO and CO_2 following the filling with ^{18}O up to a

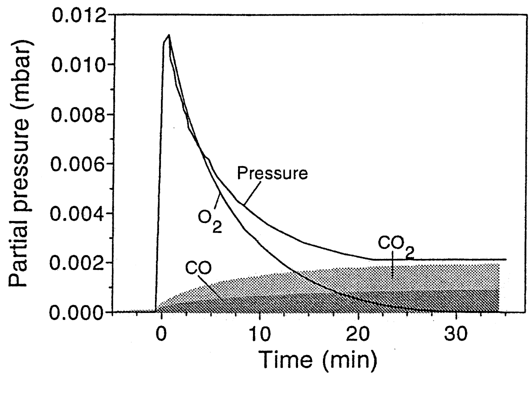

Fig. 1. Temporal behaviour of the partial pressures of $^{18}O_2$, the sum of the $C^{16,18}$ O and $C^{16,18}$ O_2 pressures and of the total pressure after a ventilation of TEXTOR with $^{18}O_2$ to an initial pressure of 0.011 mbar (this ventilation followed immediately after a ventilation to 0.007 mbar). All external pumps are closed. Plasma facing wall temperatures range from about 520 to 650 K. (For more details see inside text.)

pressure of 0.011 mbar in TEXTOR. The liner temperature was 610 K. This injection immediately followed a similar oxygen venting to a pressure of 0.007 mbar lasting for a similar timespan. It can be seen that the oxygen partial pressure decreases with an e-folding time of about 7 min. Simultaneously with the consumption of oxygen, the partial pressures of CO and CO_2 rise. After 30 min all of the oxygen is consumed and the CO and CO_2 becomes stationary at about 0.008 and 0.01 mbar, respectively. As can be seen, the evolution of these partial pressures explain well the evolution of the total pressure measured by baratrons. The strong decrease of the total pressure shows that at these conditions (low filling pressures) most of the oxygen adsorbs on the walls. Of the 4.6×10^{21} oxygen atoms injected about 3.7×10^{21} adsorbed on the walls, 8.7×10^{19} and 1.2×10^{20} are converted into CO and CO_2, respectively (see Table 1). The adsorbed amount of oxygen corresponds to about 1×10^{16} O/cm^2 uniformly distributed over the plasma facing liner surface area (35 m^2). Fig. 2 shows the same signals for an oxygen pressure of 0.32 mbar. Within 120 min the O_2 pressure decreases from 0.32 to 0.18 mbar and the CO and CO_2 partial pressure increases to 0.016 and 0.052 mbar, respectively. The oxygen pressure decays much more slowly with a time constant of about 4 h, more than 30 times longer compared with that at the low filling pressure. The adsorption corresponds to a coverage of 7×10^{16} O/cm^2 on an area of 35 m^2. A total number of 1.4×10^{22} C-atoms have been removed corresponding to 0.28 g of carbon.

It should be noted that the data represent values integrated over the total exposure time during which the oxygen pressure decreases. In contrast, Fig. 3 shows the temporal evolution of the CO and CO_2 production rate for the 0.32 mbar evaluated from the derivative of the pressure rise. A rapid decrease of the reaction occurs during the first 20 min showing that the reaction rate is transient in the beginning. Thereafter the removal rate remains nearly constant in time since it decreases linearly with the (slightly) decreasing oxygen pressure.

Table 1 also summarises the results from experiments not described here. It shows that the overall amount of the adsorbed oxygen is not linear with the filling pressure but shows a tendency to saturate. In contrast, the integrated fraction of oxygen transformed into CO stays roughly constant with pressure whereas CO_2 shows a clear tendency to increase with increasing pressure.

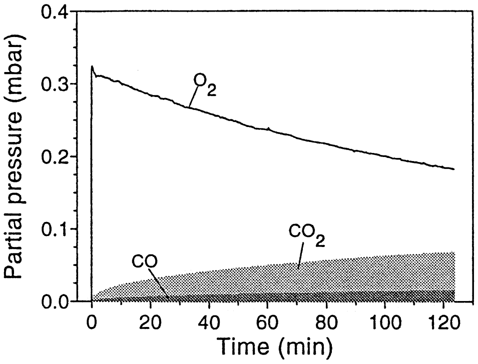

Fig. 2. Temporal behaviour of the partial pressures of O_2, CO and CO_2 after a ventilation of TEXTOR with $^{16}O_2$ to an initial pressure of 0.32 mbar. All external pumps are closed. Plasma facing wall temperatures range from about 520 to 650 K. (For more details see inside text.)

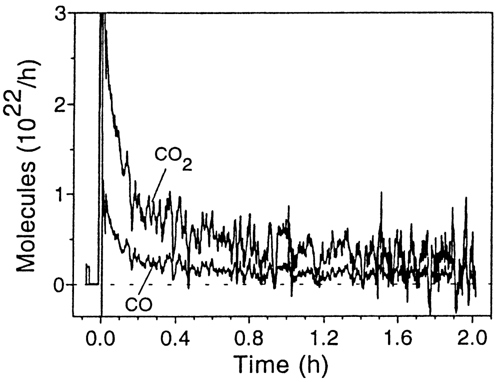

Fig. 3. Temporal behaviour of the CO and CO_2 production rate after ventilation of TEXTOR to 0.32 mbar with $^{16}O_2$. All pumps are closed. The behaviour of the oxygen partial pressure is shown in Fig. 2.

Table 1
Summary of all venting experiments performed at the same liner temperature of 610 K

Filling pressure (mbar)	Injected O-atoms $\times 10^{21}$	Exposure time (min)	Adsorbed O-atoms $\times 10^{21}$	Formed CO molecules $\times 10^{19}$	Formed CO_2 molecules $\times 10^{19}$	Fraction $CO/2O_2$ (%)	Fraction $2CO_2/2O_2$ (%)
0.007	2.88	7	2.72	6.3	4.8	2.2	3.3
0.011	4.6	20	3.7	8.7	12	1.9	5.3
0.093	38	100	22.8	51.3	93	1.35	4.9
0.32	130	120	28	330	1040	2.6	16

3.2. Isotope behaviour

An interesting feature is the observation that the injection of ^{18}O oxygen isotopes leads not only to the formation of $C^{18}O$ (mass 30) and $C^{18}O_2$ (48) but also to a significant release of $C^{16}O$ (28), $C^{16}O^{18}O$ (mass 46) and even of $C^{16}O_2$ (mass 44). This phenomenon is demonstrated in an exemplary manner in Fig. 4. For the CO_2 formation, the formation of the mixed molecule ($C^{16}O^{18}O$) was in all experiments the largest.

This result shows that significant amounts of ^{16}O atoms had been stored in the plasma facing components which are released later by the oxygen attack. Two principle mechanisms could be possible in order to explain this behaviour: (i) $C^{18}O$ might collide with an absorbed ^{16}O atom and undergo an isotope exchange process; (ii) the injected oxygen erodes the redeposited carbon layers by the formation of CO and CO_2 by which process a stored ^{16}O atom is released. This process is very likely since it is observed that oxygen containing carbon layers (prepared by implantation with oxygen ions) release the stored oxygen in the form of CO when the layer is eroded by exposure to thermal hydrogen atoms [9]. In the same manner an oxygen ventilation with ^{16}O which immediately followed a ventilation with ^{18}O has led to measurable amounts of released $C^{18}O$ and in particular $C^{16}O^{18}O$.

3.3. Hydrogen release

In laboratory experiments of the oxidation of carbon films, it has been concluded that the layer is removed by formation of CO and CO_2 and the incorporated hydrogen is released in the form of water molecules [10]. No significant release of hydrogen molecules or hydrocarbons has been observed. Similarly, no release of molecular hydrogen by oxygen venting could be identified within the sensitivity of the present measurement. Release of hydrogenic species in the form of water is expected as $D_2^{18}O$ (mass 22) and $HD^{18}O$ (21) in case of ^{18}O injection and as mass 20 and 19 in case of ^{16}O injection (It should be noted here, that the signals of mass 22 are corrected with respect to the CO^{2+} ion signal which is a fraction of about 3.5% the normal CO_2 (mass 44) signal). However, ^{18}O attack might also lead to release of hydrogen bonded on ^{16}O as it is observed in case of CO and CO_2 formation. Fig. 5 shows as an example the temporal evolution of the signals of mass 44, 21, 22 by injection of $^{18}O_2$ which is then followed by a ventilation with $^{16}O_2$. The ^{18}O injection leads to a rapid increase of mass 21 and 22, which has to be interpreted as $HD^{18}O$ and $D_2^{18}O$. Although the formation of CO and CO_2 proceeds progressively, the signals stay nearly constant or even decrease in time as it can be seen on mass 21. The decrease of the signal at mass 21 shows that water is adsorbed on the wall surfaces (all pumps are closed!), most probably on colder areas of the TEXTOR first wall [vacuum wall and in particular portholes, valves etc., which are kept at much lower temperatures (<350 K)]. The following ventilation with $^{16}O_2$ injection shows that the formation of mass 21($HD^{18}O$) and 22 ($D_2^{18}O$) decreases fast after some initial increase in the beginning of the injection, whereas $HD^{16}O$ and $D_2^{16}O$ now increases strongly (not shown in the figure). The behaviour shows that the oxygen ventilation leads clearly to the formation of water molecules. A quantitative interpretation of the mass spectroscopic signals remains, however, difficult and seems to us not meaningful at the moment. More work is planned to better understand the adsorption/desorption behaviour of water on the walls, the temporal response of the differentially pumped quadrupole systems and of background signals in the mass spectrometer, which might originate from processes occurring in the ionisation source of the mass spectrometer.

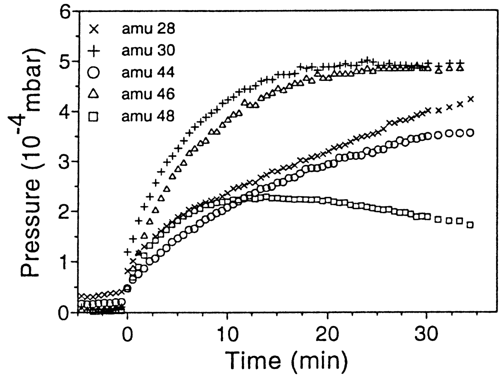

Fig. 4. Temporal behaviour of the partial pressures of all the carbon oxide isotopes after ventilation of TEXTOR with ^{18}O to an initial pressure of 0.011 mbar. All pumps are closed. The oxygen pressure behaviour is shown in Fig. 1.

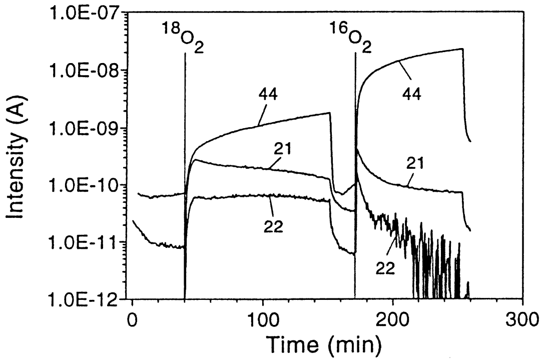

Fig. 5. Evolution of the signals at 21, 22 and 44 amu after two subsequent ventilations of TEXTOR with $^{18}O_2$ (first) and $^{16}O_2$ (second). All pumps are closed.

4. Oxygen removal and influence on plasma operation

The influence of the oxygen ventilation on plasma operation has been investigated after two subsequent oxygen ventilations with $^{18}O_2$ to 0.007 and 0.011 mbar and after that with ^{16}O up to 0.32 mbar. As shown in Table 1 the total amount of adsorbed oxygen was 6.4×10^{21} O-atoms and 2.8×10^{22} O-atoms, respectively. Standard ohmic discharges with averaged densities of 2.5×10^{13}–$2.8 \times 10^{13}/cm^3$ at 350 kA have been used. The impurity content has been investigated from local emission spectroscopy at the ALT limiters, VUV spectroscopy and bolometry. Fig. 6 shows as an example the behaviour of OI and CI light emission normalised to that of the H_α intensity (a) and the oxygen VI line radiation and the total radiation (b) after the first injection campaign [0.007 + 0.11mbar]. Without further wall conditioning, the first two attempts of plasma build up failed due to disruptions already in the current ramp-up phase. This is expected for strongly contaminated plasmas. He-glow discharge cleaning (0.005 mbar, 400–600 V, 8 A) was performed for about 15 min and an amount of 1.1×10^{20} and 4.8×10^{20} ^{16}O and ^{18}O atoms was removed in the form of $C^{16,18}$ O (≈90%) and CO_2 (≈10%), respectively, as determined by quantitative mass spectroscopy. This represents only about 1.5% of the adsorbed ^{18}O atoms. The He-GDC show a rapid decrease of the CO removal rate with time. After that, plasma operation was possible, but the relative oxygen flux increased by about a factor of 3 together with some slight increase of the carbon release. The shot by shot behaviour shows some decrease of the oxygen impurity flux. In order to reduce the oxygen content furthermore, D_2-GDC was performed for 15 min. This removed about 6.5×10^{20} ^{18}O and 2.4×10^{21} ^{16}O, representing 7.2% of the adsorbed ^{18}O demonstrating again that the D_2-GDC is much more effective to remove oxygen compared with He-GDC. The reason for this is that the oxygen is also removed from deeper layers inside the carbon films due to chemical and physical erosion by hydrogen impact whereas He-GDC releases preferentially the oxygen which is adsorbed on the top surfaces. The D_2 GDC reduced the oxygen influx and oxygen impurity content furthermore. Again some self-cleaning effect of the plasma is seen. After this, a routine boronisation of TEXTOR was performed and the figure also shows the initial behaviour of oxygen and carbon fluxes, however, at slightly higher density. The boronisation reduced the oxygen flux somewhat more, but in general the oxygen flux was not significantly changed compared with that just before the oxygen ventilation. The ohmic density limit was at about $6 \times 10^{13}/cm^3$ ($I = 350$ kA) demonstrating that a clean plasma situation was restored. The plasma behaviour after the ventilation to 0.32 mbar was very similar: plasma operation failed in the first attempt and D_2-GDC was performed immediately. The enhancement of the oxygen impurity level was very similar to that described above although the amount of adsorbed oxygen was about a factor of three larger.

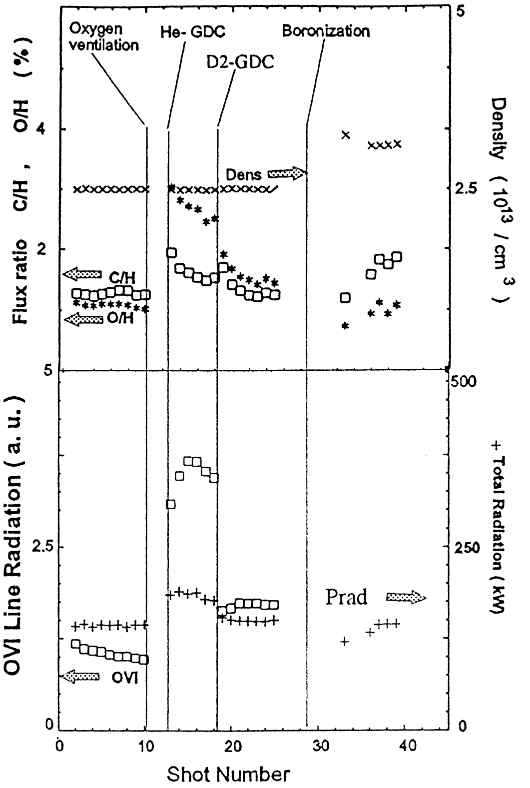

Fig. 6. Evolution of the oxygen and carbon flux ratios measured at the ALT limiter, the total radiation, the brilliance of an oxygen VI line radiation and the line averaged density before and after oxygen ventilation of TEXTOR. Two subsequent ventilations to 0.007 and 0.011 mbar were done. The first two discharges after the ventilations disrupted in the early start-up phase. The figure indicates the steps at which He- and D_2-GDC was done for about 15 min. Standard boronisation was performed afterwards at the same wall temperature.

5. Discussion and summary

The laboratory experiments on the oxidation of carbon films show that a significant reaction of oxygen with the layers proceeds in the range between 500–750 K with lower temperatures necessary for soft layers and higher temperatures for hard films.

The experimental conditions for the baking of TEXTOR-94 in oxygen are more complex: the hot area of the wall components facing the plasma is about 40 m^2 with a similar area not facing the plasma. The vacuum vessel has an even larger area but is ≈420 K. However, the temperature distribution of the components facing the plasma is large ranging from about 520 to 650 K. Thick carbon deposits with the appearance of hard films [3] are found on surfaces in TEXTOR but also soft-like deposits may be formed. In the vessel, inspections seem to reveal more soft-like films on parts of the liner surface in the vicinity of the NBI ports. Due to the distribution of the temperatures and also of the structure of deposits, it is difficult to correlate the present observations to special surface areas in TEXTOR.

The present results show that the main reaction product during baking of TEXTOR in pure oxygen is CO and CO_2 molecules which can be pumped easily. In the investigated pressure range (0.007–0.32 mbar), the reaction rate to CO is constant with the pressure whereas that of CO_2 increases with increasing pressure. The reaction decreases with time in the initial phase after the injection and then levels off and stays constant. During that quasi steady-state phase the pressure is about 0.25 mbar and the overall reaction rate to CO and CO_2 about 2.5×10^{18} CO + CO_2/s. This rate is similar to that removed in a D_2-GDC in TEXTOR (400–600 V, 8 A) and corresponds to an overall removal of carbon of about 2 g C/day. This is equivalent to a carbon layer of 0.1 μm on a plasma facing area of 40 m^2. If this removal rate would proceed linearly with increasing pressure a carbon removal rate of about 0.8 kg/day would result for TEXTOR for an oxygen filling pressure of 100 mbar.

A significant part of the injected oxygen is adsorbed on the walls forming a stable compound. At low filling pressure (0.01 mbar) about 90% of the injected oxygen is adsorbed. With increasing pressure and exposure time, adsorption tends to saturate. At the filling pressure of 0.32 mbar about 20% of the injected oxygen is adsorbed within 2 h. Part of the oxygen might penetrate in the porous structure of the carbon deposits as observed in recent measurements [12]. However, formation of oxygen compounds with metallic surfaces in TEXTOR cannot be ruled out.

Formation of water by oxygen injection could be measured clearly by the differentially pumped quadrupoles. A quantification in terms of released hydrogen needs more experiments and better understanding of the water adsorption and desorption behaviour from the entire wall. Work in this field is under way.

Reaction of oxygen ^{18}O isotopes also leads to the release of ^{16}O containing carbon oxides. This is most probably due to adsorption of ^{16}O in the carbon structures which is released in the form of oxides by ^{18}O oxidation. Formation of CO is also the mechanism by which oxygen is released from oxygen containing carbon layers during etching in hydrogen (e.g. during D_2-GDC).

After the venting experiments, the plasmas disrupted in the early start-up phase indicate strong impurity contamination. Both He- and D_2-GDC was successful to re-establish the plasma operation, but D_2- GDC was found to be much more effective to remove the oxygen compared with He-GDC. D_2-GDC removed about 7–10% of the adsorbed oxygen within about 15 min. After the wall conditioning, the oxygen impurity content was about a factor of 2–3 higher than before but the increase was similar after venting to 0.01 and 0.3 mbar, respectively. The enhanced oxygen impurity content decreased slightly shot by shot indicating some self-cleaning process by plasma operation. Boronisation was performed about 25 discharges after the first venting to 0.01 mbar oxygen. It reduced the impurity content to a level as it is usual after a boronisation of TEXTOR.

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